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Geographically Differentiated Life-cycle Impact Assessment of Human Health

by

Sebastien Humbert

A dissertation submitted in partial satisfaction of the

requirements for the degree of

Doctor of Philosophy

in

Engineering-Civil and Environmental Engineering

in the

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of the

University of California, Berkeley

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University of California, Berkeley

Geographically Differentiated Life-cycle Impact Assessment of Human Health

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ABSTRACT

Geographically Differentiated Life-cycle Impact Assessment of Human Health

by

Sebastien Humbert

Doctor of Philosophy in Engineering-Civil and Environmental Engineering

University of California, Berkeley

Professor Arpad Horvath, Chair

In this dissertation, I explore how life-cycle assessment (LCA) results can potentially be influenced by the location of emissions, also called regionalization. The current practice in life-cycle assessment increasingly stresses the importance of being “geographically differentiated” or “regionalized.” Regionalization can be important for cases that have extended supply chains distributed across different regions or for processes situated in locations with environmental conditions and population densities far from “average”. A spatially resolved model for North America, called IMPACT North America, is developed here. Geographically differentiated intake fractions and characterization factors are evaluated for the North American continent for human toxicity. Special attention is given to the modeling of intake fractions and characterization factors for particulate matter. These factors can be used to adapt damage-oriented impact assessment methods. Parameters influencing intake fractions and characterization factors are identified, such as population densities and agricultural production. The importance of modeling an “urban box” and a “remote area” is also shown. The case of fly ash concrete is studied through the lens of regionalization, and critical parameters influencing the environmental trade-offs between fly ash concrete and Portland cement concrete are identified.

Through the development of the new model, along with the different applications, various questions related to spatial life-cycle assessment can be studied, and several conclusions can be drawn about what is important to consider when performing regionalization in life-cycle assessment.

I show that particulate matter dominates damage to human health and therefore the importance of devoting more detailed attention to the character of particulate matter when modeling the fate, exposure and effect of pollutants. I also demonstrate that when addressing the issue of regionalization in inventory and impact assessment, priority should be placed on the development of the archetype-based (i.e., situation-dependent) regionalization; spatialization (geography-based regionalization) should only be used secondarily and in occasional cases when results from key processes need to be expanded.

*To my entire family,
And especially to my aunt and godmother, Madeline, who enabled me to enter the world of
studies,
As well as to the future generations*

*A toute ma famille,
Et particulièrement à ma tante et marraine Madeline, qui m'a permis d'entrer le monde des
études,
Ainsi qu'aux générations futures*

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Acronyms and Nomenclature

The different acronyms used in this dissertation are the following:

CF	Characterization Factor (“impact” per amount emitted)
DALY	Disability Adjusted Life Years: a unit expressing impacts to human health (Murray and Lopez 1996)
ED ₁₀	Effect Dose inducing a 10% response over background
EF	Effect Factor (“impact” per amount intake)
EIO-LCA	Economic Input-Output-based Life-cycle Assessment (Hendrickson et al. 1998)
iF	Intake fraction (also known as exposure efficiency — Evans et al. 2002) (Bennett et al. 2002)
ISO	International Organization for Standardization
LCA	Life-cycle Assessment
LCI	Life-cycle Inventory
LCIA	Life-cycle Impact Assessment
NO _x	Nitrogen oxide. NO _x is composed of NO and NO ₂ but with a significant dominance of NO ₂
PAF	Potentially Affected Fraction of species (Payet 2005); a unit expressing impacts to ecosystem quality used in several damage oriented life-cycle impact assessment methods (e.g., Jolliet et al. 2003)
PDF	Potentially Disappeared Fraction of species; a unit expressing impacts to ecosystem quality used in several damage oriented life-cycle impact assessment methods (e.g., Jolliet et al. 2003)
PM	Particulate Matter
RfD	Reference Dose
SETAC	Society of Environmental Toxicology and Chemistry
uF	Uptake fraction (term introduced in this dissertation)
UNEP	United Nations Environmental Program

Units: Unless specified, all units follow the international system. Note that in United States conventions “tons” refers to the United States short ton, which is 2,000 lbs. In the United States, one uses “tonne” or “metric ton” or “metric tonne” to refer to 1,000 kg. In all cases, “t” refers to “1000 kg”. “Year” is abbreviated to “y”, “day” to “d”, “hour” to “h” and “second” to “s”.

Throughout this dissertation the following nomenclature is used (broadly accepted terms are not presented).

- “Multipathway” instead of the traditional “multi-pathway” to be consistent with the more accepted and used term “multimedia.” “Multipathway” is used in a sense that encounters “multiexposure.”
- “Geographically differentiated,” “spatially differentiated,” and “spatially resolved” are used as synonyms. Indeed, the roots “spatial” and “geographical” refer to a specific place in the world.

- “Geographically differentiated” refers to the differences among geographical locations such as continents, countries, or regions throughout the world, e.g., South versus North America, Germany, North Sea, etc. The geographical location where the impacts occur is sometimes captured in the information related to the source of the material or product, e.g., crude oil from Venezuela. (Sedlbauer et al. 2007)
- “Spatially resolved” refers to an application (e.g., a model) that considers different levels or zones in the space.
- “Situation dependent” (or “archetypical”) refers to archetypical situations leading to important variations in the characterization modelling and its results and therefore justifying a differentiation. It then refers to the type of environment the pollutant is emitted into or where the inventory flow is occurring, e.g., high versus low population density area, agriculture intensive area, upstream versus downstream of a lake. (Sedlbauer et al. 2007). The term “situation dependent,” or “archetype,” is therefore independent of the geographical region (i.e., urban, rural, etc. zone wherever it is in the world). It is defined by its characteristics and not by its location.

Summary

Life-cycle assessment (LCA) is increasingly used to help understand the overall environmental consequences of any subject of interest (a product, a service, a company, an entity, a policy, etc.). Within life-cycle assessment, impact assessment is a step that allows the expression of environmental impacts through a reduced number of indicators, thus simplifying interpretation of the results of a life-cycle assessment. The magnitude of the impacts associated with the emission of pollutants can vary depending on the locations of emission throughout the world. The current life-cycle assessment practice stresses the importance of “regionalization.”

In this dissertation, I explore how human health damage factors, or characterization factors, of pollutants and life-cycle assessment results are influenced when the location of emissions is taken into account. This issue can be important for emissions that are situated in zones with conditions far from average or have significant and extended supply chains that may easily change location.

In Chapter 1, I revisit the general concepts of life-cycle assessment as well as the state of the art of geographic differentiation in life-cycle impact assessment. I describe the objectives of this dissertation and the chosen method. I also describe the nomenclature used throughout this dissertation, with a suggestion for the life-cycle assessment community to improve communication about geographic differentiation of life-cycle assessment.

In Chapter 2, I present IMPACT North America, a spatially resolved, multimedia, multipathway, fate, exposure and effect model for North America developed to evaluate geographically differentiated intake fractions and characterization factors for the North American continent for human toxicity, aquatic ecotoxicity and terrestrial ecotoxicity. The IMPACT North America model is evaluated using monitored data, as well as by comparing results with other spatially resolved models. Results show that intake fractions and characterization factors can vary by up to eight orders of magnitude depending on where the pollutant is emitted in North America.

In Chapter 3, I use the model developed in Chapter 2 to explore how regionalization influences the intake fractions of pollutants, to suggest how generic intake fractions and characterization factors should be computed, to compare geography-based and archetype-based intake fractions, and to draw recommendations on how regionalization of fate and exposure should be performed in life-cycle assessment. In Chapter 3, I conclude that archetype-based regionalization has substantial advantages over geography-based regionalization, therefore making archetype-based regionalization a more powerful approach to perform regionalization in life-cycle assessment. As suggested in Chapter 3 generic intake fractions and characterization factors should be calculated as emission-weighted average intake fractions and characterization factors. However, when no emission data are available, in Chapter 3, I suggest using population distribution as a proxy for emissions distribution.

In Chapter 4, I review and recommend intake fractions, effect factors and characterization factors for primary and secondary particulate matter (PM). This work is performed in addition to that in Chapter 2 because the IMPACT North America model is appears not suited to evaluate intake fractions of secondary PM. Because PM is often found to dominate the human health damage in life-cycle assessment, a special emphasis is put on making intake fractions, effect factors and characterization factors of PM consistent for a complete set of emissions sources and locations, introducing the source-location matrix concept. I conclude that intake fractions and characterization factors of PM can vary by several orders of magnitude, depending on the

population density patterns surrounding the location of emission, the stack height and meteorological conditions. This chapter serves as a basis for the recommended European Commission life-cycle impact assessment method as well as for the UNEP-SETAC Life Cycle Initiative task force on damage to human health from PM.

In Chapter 5, I use the intake fractions and characterization factors developed in Chapters 2 and 4 as well as the archetypes developed in Chapters 2 and 3 to evaluate which pollutants dominate the total human health damage in the United States (U.S.), as well as explore the influence of regionalization on human health damage of different processes. In Chapter 5, I evaluate whether regionalization is important and compare the two approaches, geography-based or archetype-based, to perform regionalization. I conclude that PM smaller than 2.5 μm ($\text{PM}_{2.5}$) is dominating the damage to human health caused by airborne pollutant emissions in the United States and therefore the regionalization scheme chosen should be well suited to PM emissions. Furthermore, I also conclude that the archetype-based approach is more practical than the geography-based approach.

In Chapter 6, I introduce and define the concept of uptake fraction for PM, the fraction of emitted PM that will eventually deposit in the respiratory tract. Indeed, because PM is often found to dominate the total human health damage, I provide ideas on how to improve the fate, exposure and effect modeling for PM, leading to the concept of uptake fraction. In this chapter, I find that for an identical intake fraction, the uptake fraction of PM in the respiratory tract can vary by a factor of three depending on the particle size distribution of PM, therefore making this new metric a substantial improvement in quantification of human exposure to PM.

In Chapter 7, I present UFPM (Uptake Fraction of Particulate Matter), a model developed to evaluate the uptake fraction, a concept defined in Chapter 6, of primary PM, especially considering the influence of the particle size distribution of the different types of PM. The uptake fraction concept provides a complementary approach to the rather simplified approach used in the IMPACT North America model and the recommendations of Chapter 4 for the evaluation of intake fraction, uptake fraction and characterization factor of PM. I find that the ratio of uptake fraction to intake fractions is on average 0.3 kg deposited per kg inhaled. I also find that intake fractions and uptake fractions of PM can vary by several orders of magnitude, mainly influenced by the population density patterns downwind of the emission, and that population situated hundreds or even thousands of kilometers downwind from the PM source will still be exposed to the PM.

In Chapter 8, I use the concept of uptake fraction developed in Chapter 6 and calculated in Chapter 7 to calculate the effect factors and characterization factors of different types of PM, for different population density patterns and using different proxies for damage such as mass and surface area of particles deposited in the respiratory tract. I apply these characterization factors to the case of a diesel passenger vehicle. I show how population density of the location of emission influences the overall human health damage of different scenarios.

In Chapter 9, I apply the characterization factors developed in Chapters 2 and 4 to the case of fly ash disposal. Critical parameters influencing the results of fly ash disposal are identified. I show that although regionalization alters the absolute results for damage to human health, regionalization will not change the main observations, in this case that the preferred option (which is to use fly ash in concrete) is confirmed, because the differences among disposal alternatives are substantial. I also conclude that if fly ash is seen as a co-product of coal-fired power plants, using fly ash in concrete might not be as beneficial for the environment as it would be if fly ash were seen as waste.

In Chapter 10, I apply the characterization factors developed in Chapters 2 and 4 to the case of fly ash concrete. Critical parameters influencing the environmental trade-off between fly ash concrete and Portland cement concrete are identified, especially in light of regionalization of damage to human health. I conclude that damage to human health is the limiting factor when evaluating over what distance fly ash can be shipped. I show that regionalization will increase reliability of the results of damage to human health and therefore the confidence in final results when evaluating trade-offs among alternatives.

In Chapter 11, I summarize the main contributions of the dissertation as well as further research needs.

1. Introduction

1.1. Introduction to life-cycle assessment

Human activities and their associated impacts are often very challenging and thus difficult to evaluate. Life-cycle assessment is a concept and methodology that has been developed to analyze the consequences of human activities on the environment.

1.1.1. Basics of life-cycle assessment

Life-cycle assessment is a concept and methodology to study the environmental effects of a product, process, activity or service throughout the entire life cycle (from “cradle to grave,” taking into account the supply chains). One of the powerful advantages of life-cycle assessment is that it prevents “shifting” of the impacts from one life cycle stage to another or from one impact category to another. The general structure of life-cycle assessment is presented in Figure 1-1.

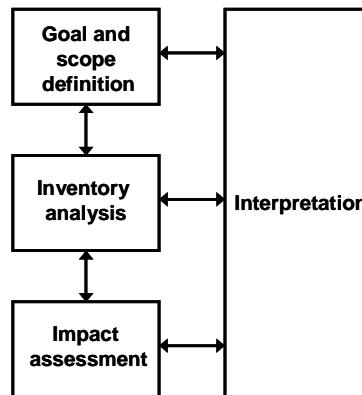


Figure 1-1: Life-cycle assessment methodology structure (based on ISO 2006a, 2006b).

Goal and scope definition: The goal and scope define what is studied, what the questions are, what the functional unit is, what the boundaries are, etc. A “reference flow” is a measure of the outputs from processes in a given product system required to fulfill the function expressed by the functional unit (ISO 2006a, 2006b).

Life-cycle inventory: Life-cycle inventory (LCI) aims at identifying what the inputs (resources) and outputs (emissions and wastes) are, their respective amounts, and the media of emissions over the entire life cycle. The inputs and outputs are generally referred to as “elementary flows.” Generally results are given in physical units (kg, m³, etc.) consumed or emitted. In practice, the location of emissions is often neglected. One objective of this dissertation is to promote the use of location-sensitive supply chain analysis in life-cycle assessment.

Life-cycle impact assessment: The life-cycle impact assessment (LCIA) aims at evaluating the impacts of the different inputs and outputs identified in the inventory on different targets (“impact categories”). The impact categories can be related to human health, ecosystems, but also to climate or resources. A typical structure of the impact assessment method is shown in Figure 1-2 using IMPACT 2002+ (Jolliet et al. 2003b) as an example.

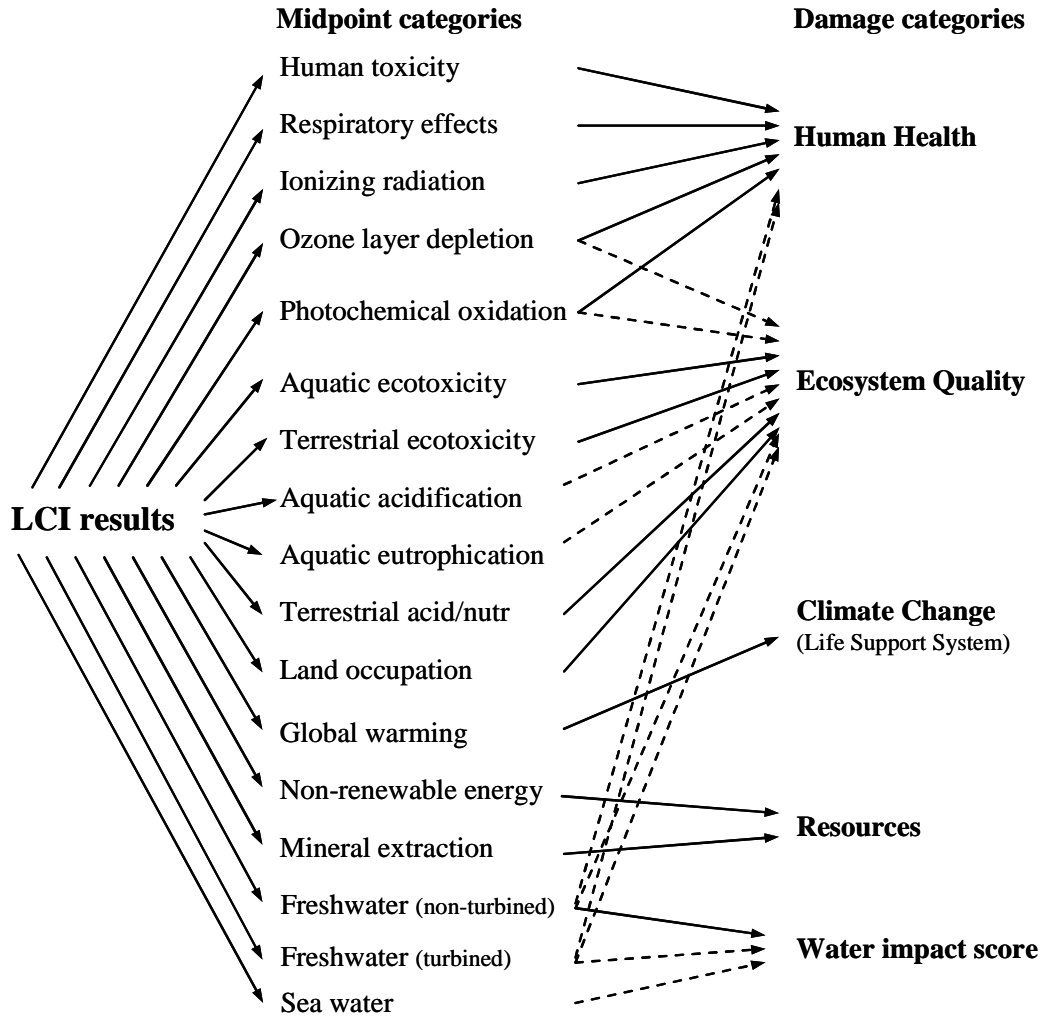


Figure 1-2: Typical structure of impact assessment method (based on Jolliet et al. 2003b).

The impact score: Life-cycle impact assessment provides characterization factors (CF) that express “impacts” per “amount” of inventory. The impact score (or damage score) on a target (humans, ecosystems, climate, etc.) can be estimated as the product of an emission (E) and a characterization factor (Rosenbaum et al. 2007):

$$Impact [impact] = E [mass emitted] \times CF \left[\frac{impact}{mass emitted} \right] \quad (1-1)$$

The characterization factor: The characterization factor (CF) can be expressed as the product of a fate factor (FF), an exposure factor (XF), a dose-response factor (DR) and a severity factor (SF) (Rosenbaum et al. 2007):

$$CF \left[\frac{impact}{mass emitted} \right] = FF \left[\frac{ambient concentration}{mass emitted} \right] \times XF \left[\frac{mass intake}{ambient concentration} \right] \times DR \left[\frac{case}{mass intake} \right] \times SF \left[\frac{impact}{case} \right] \quad (1-2)$$

The intake fraction: For human health, the fate factor multiplied by the exposure factor can be represented as the intake fraction (iF) (Bennett et al. 2002):

$$iF \left[\frac{mass intake}{mass emitted} \right] = FF \left[\frac{ambient concentration}{mass emitted} \right] \times XF \left[\frac{mass intake}{ambient concentration} \right] \quad (1-3)$$

The intake fraction is defined by Bennett et al. (2002) as:

$$iF = \frac{\sum_{\text{people, time}} \text{intake of pollutant by an individual (mass)}}{\text{mass released into the environment (mass)}} \quad (1-4)$$

The effect factor: For human health, the dose-response factor multiplied by the severity factor can be represented as the effect factor (EF). The effect factor is a factor quantifying the effect on human health that the intake of a certain amount of a specific pollutant will have. For example, it can be expressed as “DALYs” (disability adjusted life years – Murray and Lopez 1996 – the number of years of life that a person is losing because of mortality or morbidity caused by carcinogenic and non-carcinogenic effects) per mg of pollutant inhaled or ingested. Crettaz et al. (2002) and Pennington et al. (2002) developed effect factors for cancer and non-cancer effects for hundreds of pollutants based on the following approach:

$$EF_i = \left[\beta_{ED10-i} \times \frac{1}{BW} \times \frac{1}{LT_h} \times \frac{1}{N_{365}} \right] \times DALY_p \quad (1-5)$$

with EF_i : effect factor of substance i [DALY/mg_{intake}], β_{ED10-i} : slope factor of i [risk per mg_{intake}/(kg_{bw}·d)], BW : human body weight [kg_{bw}], LT_h : lifetime of a human [y], N_{365} : number of days in one year [d/y], and $DALY_p$ being the severity per person caused by the risk associated with substance i [DALY/risk].

Hence, the impact to human health can be expressed as:

$$Impact = E \times iF_{inhalation} \times EF_{inhalation} + E \times iF_{oral} \times EF_{oral} + E \times iF_{dermal} \times EF_{dermal} \quad (1-6)$$

1.2. Introduction to and state of the art of life-cycle impact assessment

This section introduces the main impact assessment methods used in the third phase of life-cycle assessment (ISO 2006a, 2006b). The aim of this chapter is to provide context for the contributions of this dissertation.

1.2.1. Life-cycle impact assessment

Life-cycle impact assessment aims to connect the elementary flows (extractions and emissions, land use, etc.) identified in the life-cycle inventory with different impact categories (human toxicity, global warming, acidification, etc.) to improve the interpretation of results by reducing the number of parameters that need to be considered. It does so by allowing one to select a set of impact categories, category indicators and characterization models (one per impact category), then assign (classify) the elementary flows to the different impact categories and then convert (characterize) them into the common indicator units within each category. The relative contribution of each elementary flow to each impact category can be assessed. Two optional steps are also possible: normalization, which translates the magnitude of the impact category indicator results relative to a reference scenario, and weighting, which converts the normalized results into the same unit, allowing for their aggregation into a single score. All of the steps up to normalization are based on natural sciences, whereas the weighting step takes value choices into account.

The impact category indicator result (also called impact score, I) is the sum of all elementary flows contributing to the specific impact category of the product between the life-

cycle inventory result (E) and its respective conversion factor, called characterization factor (CF):

$$I = \sum_i E_i \times CF_i \quad (1-7)$$

For example, knowing that the characterization factors (considering a 500-year time horizon, also known as GWP500) of CH₄ and N₂O are, respectively, 7.6 and 153 kg CO₂-eq/kg of emitted gas (IPCC 2007), the impact score for the global warming impact category (expressed in units of kg CO₂-eq) resulting from an emission of, e.g., 4 kg (biogenic) CH₄ and 0.5 kg N₂O is the following:

$$I = (4\text{kg}_{CH_4} \times 7.6\text{kg}_{CO_2\text{eq}}/\text{kg}_{CH_4}) + (0.5\text{kg}_{N_2O} \times 153\text{kg}_{CO_2\text{eq}}/\text{kg}_{N_2O}) = 30 + 77 = 107\text{kg}_{CO_2\text{eq}} \quad (1-8)$$

The chosen life-cycle impact assessment method provides the list of characterization factors.

1.2.2. Characterization versus life-cycle impact assessment methods

Different definitions exist. In the present work, the following definitions are used.

A **characterization method** calculates the characterization factors for a specific, single impact category for the different elementary flows contributing to the impacts described by this category. Methods such as the IPCC GWP100 (IPCC 2007), USEtox (Rosenbaum et al. 2008, Hauschild et al. 2008b), the method of De Schryver et al. (2009), or IMPACT 2002 (Pennington et al. 2005) are examples of characterization methods.

A **life-cycle impact assessment method** aims to address, as accurately and as consistently as possible, a large set of impact categories. A life-cycle impact assessment method then presents a consistent set of impact categories within a defined framework, each with their own characterization method. Methods such as IMPACT 2002+ (Jolliet et al. 2003a), TRACI (Bare et al. 2003), EDIP 2003 (Hauschild et al. 2004), ReCiPe (Goedkoop et al. 2008), or EU-LCIA (Hauschild et al. 2008b) are examples of life-cycle impact assessment methods.

The interaction between characterization methods and life-cycle impact assessment methods is presented in Figure 1-3.

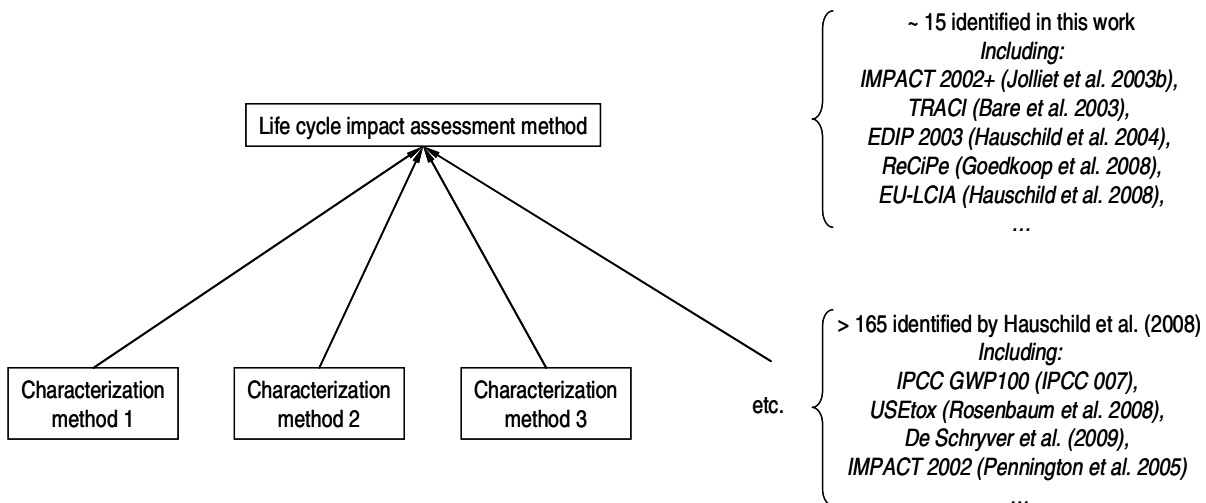


Figure 1-3. Characterization methods versus life-cycle impact assessment methods.

Method versus methodology

The terms method and methodology are often used interchangeably in the field of life-cycle assessment. To help the reader follow the terminology used in this dissertation, I decided to use the term “methodology” when referring to the general concept of life-cycle assessment and “method” when referring to specific characterization models or methods as well as life-cycle impact assessment methods based on my own interpretation of The American Heritage Dictionary of the English Language, Fourth Edition: “*Methodology can properly refer to the theoretical analysis of the methods appropriate to a field of study or to the body of methods and principles particular to a branch of knowledge. In this sense, one may speak of objections to the methodology of a geographic survey (that is, objections dealing with the appropriateness of the methods used) or of the methodology of modern cognitive psychology (that is, the principles and practices that underlie research in the field). In recent years, however, methodology has been increasingly used as a pretentious substitute for method in scientific and technical contexts, as in The oil company has not yet decided on a methodology for restoring the beaches. People may have taken to this practice by influence of the adjective methodological to mean "pertaining to methods." Methodological may have acquired this meaning because people had already been using the more ordinary adjective methodical to mean "orderly, systematic." But the misuse of methodology obscures an important conceptual distinction between the tools of scientific investigation (properly methods) and the principles that determine how such tools are deployed and interpreted.*”

Note that Hauschild et al. (2008a) use the term “method” for what I define as “characterization method,” and “methodology” for what I refer to as “life-cycle impact assessment method.”

1.2.3. Impact categories

Figure 1-4 presents the framework developed by the UNEP-SETAC Life Cycle Initiative (Jolliet et al. 2003a), giving a non-exhaustive list of suggested midpoint impact categories and their relationship to damage impact categories, also sometimes referred to as areas of protection. Note that “waste” is not to be considered an impact category as it is another process of the system that will lead to a certain amount of life-cycle inventory results.

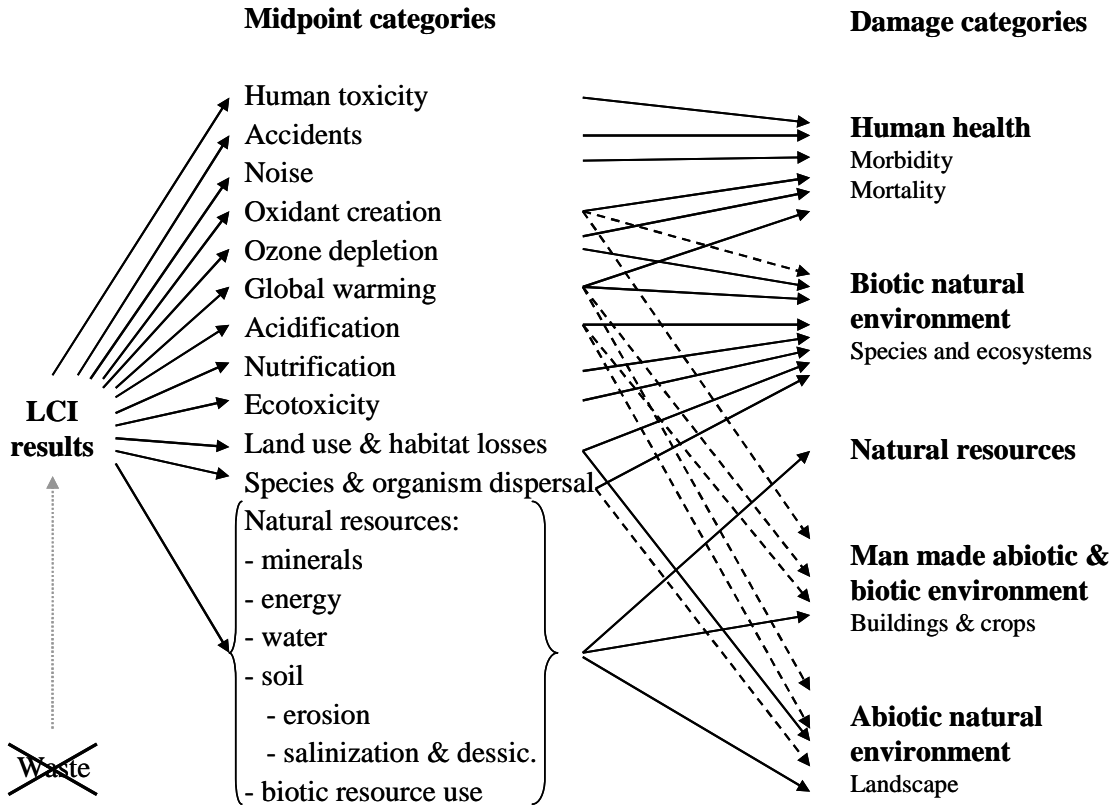


Figure 1-4. Overall framework, linking life-cycle inventory results via the midpoint impact categories to damage impact categories (Jolliet et al. 2003a).

1.2.4. Midpoint categories versus damage categories

Figure 1-5 presents the framework of life-cycle assessment from the process system through the inventory, the midpoint and damage impact categories to the single impact score. As one progresses down the cause-effect chain, the scientific precision specific to a single quantum of information (a piece of information, such as a quantified elementary flow or an impact category indicator result) decreases, but the relevance of this same information quantum for decision makers increases because of the very significant reduction in the number of single quanta of information that have to be considered simultaneously at each step.

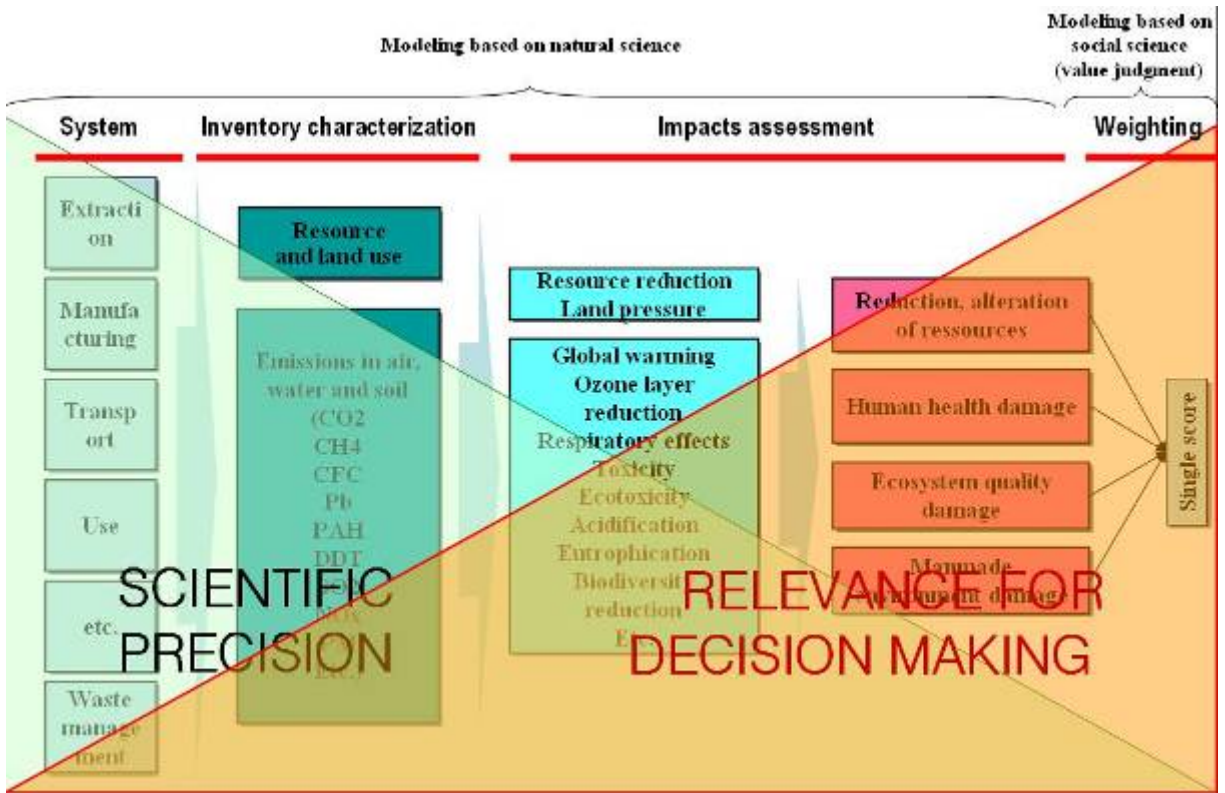


Figure 1-5. Life-cycle assessment framework: from product system to single impact score, through inventory, midpoint and damage impact categories.

1.2.5. Chronological development in life-cycle impact assessment

The chronological development of life-cycle impact assessment methods is presented in Figure 1-6.

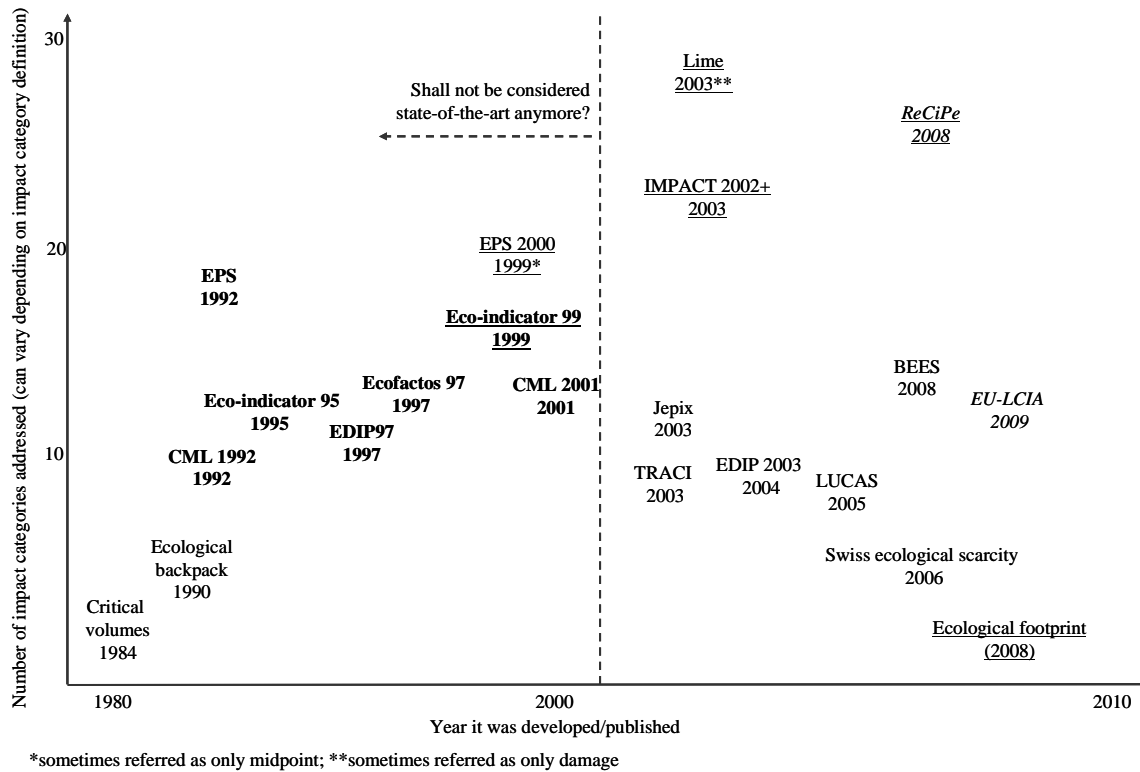


Figure 1-6. Chronological development of life-cycle impact assessment methods. Bold methods are officially outdated and have been replaced by updated versions. Underlined methods have both midpoint and damage impact categories. Italic methods are still under development.

1.2.6. Evaluation of the different life-cycle impact assessment methods

Several works have evaluated the different life-cycle impact assessment methods. In 2004, the UNEP-SETAC Life Cycle Initiative did an overview of ten life-cycle impact assessment methods (Eco-indicator 99, EDIP 97, EDIP 2003, EPS 2000d, CML 2001, IMPACT 2002+, JEPIX, LIME, Swiss Ecoscarcity, and TRACI) (UNEP 2008). More recently, at the request of the European Commission, a consortium of life-cycle impact assessment experts performed a thorough review of current life-cycle impact assessment methods with the goal of recommending a set of characterization methods for the European Platform on life-cycle assessment (Hauschild et al. 2008a). Hauschild and colleagues (2008a) identified 165 characterization methods and thoroughly reviewed 12 life-cycle impact assessment methods (CML 2001, Eco-indicator 99, EDIP 97, EDIP 2003, EPS 200, IMPACT 2002+, LIME, LUCAS, ReCiPe, Ecological Scarcity 2006, TRACI, and MEEuP). A summarized evaluation of the different methods identified in the present work is presented in Table 1-1.

Table 1-1. Evaluation of the different life-cycle impact assessment methods.

Name of the method and reference	Categories assessed	Geographical region assessed for non-global categories	Method description (especially whether it is a midpoint and/or a damage approach)	Advantages	Disadvantages
Critical volumes (1984) (BUS 1984)			Midpoint approach	Characterization model straightforward	No regionalization
Ecological backpack (http://playgreen.org/Wiki/EcologicalBackpack)			Midpoint approach	Characterization model straightforward	No regionalization
CML 1992 (Heijungs et al. 1992)	9	Europe	Midpoint approach	Characterization model straightforward	No regionalization, Damage missing, Outdated (ReCiPe is the updated version)
EPS 1993 (Steen 1992) (http://eps.esa.chalmers.se/)		Europe	Damage approach	Characterization model straightforward	Outdated (EPS 2000 is the updated version)
Eco-indicator 95 (Goedkoop 1995) (http://www.pre.nl)	11	Europe	Midpoint approach. It was developed under the Dutch NOH program by PRé consultants (The Netherlands) in a joint project with Philips Consumer Electronics, NedCar, Océ Copiers, Schuurink, CML Leiden, TU-Delft, IVAM-ER (Amsterdam) and CE Delft.	Characterization model straightforward	No regionalization, Midpoint missing, Outdated (ReCiPe is the updated version)
EDIP97 (Hauschild and Wenzel 1998) (http://ipt.dtu.dk/~mic/EDIP97)	11	Europe	Midpoint approach with normalization. Update available as EDIP2003 with site-dependent characterization for most non-global impact categories	Characterization model straightforward	Midpoint missing, Outdated (EDIP 2003 is the updated version)
Swiss ecoscarcity (Ecofactor 97) (Brand et al. 1997)	12	Europe	Weighting set based on environmental policy goals, to be used for midpoint categories and selected emissions/interventions. Update for 2004 Swiss values in progress. The "Swiss ecopoints" are based on the Swiss environmental policy; the method may be applied to other regions' environmental policy goals as well.	Characterization model straightforward	No regionalization, Outdated (Swiss ecological scarcity 2006 is the updated version)
Eco-indicator 99 (Goedkoop and Spriensma 2000) (http://www.pre.nl/eco-indicator99/)	15	Europe	Damage approach, including normalization and default weighting sets. Three versions are published (Hierarchis, Individualist, Egalitarian).	Allows single score calculation. Can have different perspectives	No regionalization, Midpoint missing, Outdated (ReCiPe is the updated version)
EPS 2000 (Steen 1999) (http://eps.esa.chalmers.se/)	13+4	Europe	Damage approach + weighting as willingness to pay (WTP) to avoid damage	Characterization model straightforward	No regionalization
CML 2001 (or Dutch Handbook on life-cycle assessment) (Guinée et al. 2002) (http://www.leidenuniv.nl/cml/ssp/projects/lca2/lca2.html)	14	Europe	Midpoint approach with normalization	Characterization model straightforward	No regionalization, Damage missing, Outdated

Name of the method and reference	Categories assessed	Geographical region assessed for non-global categories	Method description (especially whether it is a midpoint and/or a damage approach)	Advantages	Disadvantages
TRACI (Bare et al. 2003) (http://epa.gov/ORD/NRMRL/std/sab/iam_traci.htm)	9	United States	Midpoint approach with normalization	Includes some regionalization (states)	No damage
TRACI 2 (Bare et al. 2003) ((Bare et al. 2003) (http://epa.gov/ORD/NRMRL/std/sab/iam_traci.htm)	9	United States	Midpoint approach with normalization	Includes some regionalization (using geographic-based approach)	No damage
EDIP 2003 (Hauschild and Potting 2003) (http://ipt.dtu.dk/~mic/EDIP2003)	9	Europe	Midpoint approach with normalization. Site-dependent characterization for European countries for most non-global impact categories	Includes some regionalization (using geographic-based approach)	No damage
JEPIX (Miyazaki et al. 2003) (http://www.jepix.org)	10	Japan	Weighting set based on distance-to-target of environmental policy. Providing regionalized weighting factors based on specific environmental quality. Revision and update in progress under the Japanese government's Centre of Excellence program		No regionalization
LIME (Itsubo and Inaba 2003) (http://www.jemai.or.jp/lcaforum/index.cfm)	11+16	Japan	Midpoint and damage approach. All lists (midpoint, damage assessment, weighting) are downloadable from the web site Based on Japanese environmental conditions for most non-global impact categories		No regionalization
IMPACT 2002+ (Jolliet et al. 2003b) (http://www.impactmodeling.org)	17+5 (including water use)	Europe	Midpoint and damage approach, including normalization	Both midpoint and damage	No regionalization
LUCAS (Toffoletto et al. 2006)	8	Canada	Midpoint	Regionalization for Canada	Only for Canada
Swiss ecological scarcity 2006 (or Ecopoints 2006) (Frischknecht et al. 2006) (http://www.esu-services.ch/)	7	Switzerland	Midpoint approach. Ecological Scarcity 2006 is a follow up of the Ecological scarcity 1997 method, which is sometimes called Ecopoints 97 (CH)	Includes regionalized (using archetypes) assessment of water use	Not comparable with other life-cycle impact assessment methods
Ecological footprint (Wackernagel and Rees 1996, Ewing et al. 2008, Kitzes et al. 2008)	3	Global	Damage approach, single score. GHG emissions, uranium use, and land use	Very easy to communicate	No regionalization, Very limited in terms of the impacts it measures
BEES (NIST, 2008) (National Institute of Standards and Technology (NIST), U.S. Department of Commerce 2008) (http://www.bfrl.nist.gov/oea/software/bees/)	13	United States	Midpoint approach. Building for Environmental and Economic Sustainability	Characterization model straightforward	No damage. No regionalization
ReCiPe (Goedkoop et al. 2008) (http://www.lcia-recipe.net)	19+3	Europe	Midpoint and damage approach	Allows single score calculation. Can have different perspectives. Some regionalization using rough archetypes	Regionalization missing for some important categories

Name of the method and reference	Categories assessed	Geographical region assessed for non-global categories	Method description (especially whether it is a midpoint and/or a damage approach)	Advantages	Disadvantages
EU-LCIA (Hauschild et al. 2008a)	11	World for respiratory inorganics, Europe for others	Midpoint and damage approach	Consensus (widely accepted)	No regionalization

1.2.7. Methods sometimes described as life-cycle impact assessment

Confusion can arise when referring to life-cycle impact assessment methods (see above) and characterization methods. The main example of the latter is the IPCC's (2007) list of midpoint characterization factors that allow for the assessment of greenhouse gas emissions. The European Platform on life-cycle assessment (Hauschild et al. 2008a) identified more than 165 characterization methods. Table 1-2 presents a selected list of these methods that are sometimes referred to as life-cycle impact assessment methods, but that are not considered as such in the present assessment (customized explanations are presented in Table 1-2).

Table 1-2. Selected methods sometimes referred to as life-cycle impact assessment methods, but not considered as such in the present assessment.

Method and reference	Impact categories assessed	Type of method	Reason it is not considered a life-cycle impact assessment method in the present assessment	Life-cycle impact assessment methods that it is used in
Carbon footprinting (PAS2050) (BSI 2008)	Global warming	Midpoint	It is a characterization model with detailed guidelines on how to construct the LCI	Most in the global warming potential impact category
IPCC 2001 (IPCC 2001), a.k.a. GWP20, GWP100, and GWP500	Global warming	Midpoint	It is a characterization model	Most
IPCC 2007 (IPCC 2007), a.k.a. GWP20, GWP100, and GWP500	Global warming	Midpoint	It is a characterization model	Most
De Schryver et al (2009)	Global warming	Damage	It is a characterization model	ReCiPe
USEtox (Rosenbaum et al. 2008, Hauschild et al. 2008b)	Human toxicity, Aquatic ecotoxicity	Damage	It is a characterization model	EU-LCIA, ReCiPe
Pfister et al. (2009)	Water use (fresh, non-turbined)	Damage	It is a characterization model	None
Waterfootprint (Chapagain and Hoekstra 2007, Hoekstra and Chapagain 2008, Hoekstra et al. 2009)	Water use (fresh, non-turbined)	Inventory classification method	It is actually an inventory classification method, but it does not evaluate the impacts associated with water use	None

1.2.8. Recommendations

Tentative recommendations for use of life-cycle impact assessment methods based on the criteria above are presented in Table 1-3. This tentative recommendation is valid for the situation in October 2009. Development after October 2009 will most likely change the recommendations.

Table 1-3. Tentative recommendations for life-cycle impact assessment methods as of October 2009.

Main constraint	Other constraint	Recommended method	Advantage	Limits
Midpoint and damage oriented method	If it needs to be directly used and be published in a scientific journal	IMPACT 2002+ (Joliet et al. 2003b)	Fully implemented in most life-cycle assessment software	Will be surpassed by ReCiPe and EU-LCIA
	If not needed to be fully implemented in life-cycle assessment software	ReCiPe (Goedkoop et al. 2008)	Latest development for toxics, some regionalization included using archetypes	Not openly implemented in life-cycle assessment software yet (currently being fine-tuned)
	If one can wait six months	EU-LCIA (Hauschild et al. 2008a)	Consensus method (the widest consensus method)	Not operational yet and not necessarily consistent among impact categories

1.3. Scope of work of this dissertation

1.3.1. Problem statement

The term “geographically differentiated” signifies that a life-cycle assessment will not only sum the amount of emissions, but will take into account the “severity” of the impacts depending on where the different pollutants are emitted.

Current issues related to geographic differentiation in life-cycle impact assessment

Most life-cycle studies still stop at the life-cycle inventory phase. Some apply a life-cycle impact assessment method. However, the latter often uses default characterization factors, i.e., without differentiating among the different locations of emissions. Rare are the studies that try to account for the influence of the location of emissions. Thus far, no complete life-cycle assessment has been performed using geographical differentiation. One of the main reasons is that no clear and complete method exists. This area is still at its initial stage of development. The state of the art in life-cycle assessment stresses the importance of “geographical differentiation” and the identification of critical parameters. Indeed, regionalization is recognized as an important step towards improving accuracy, precision and confidence in life-cycle assessment results as well as its discriminatory power, especially for comparative assessments (Potting and Hauschild 2006, Sedlbauer et al. 2007, Reap et al. 2008, Margni et al. 2008).

Spatially differentiated life-cycle assessment can especially be important in the case of emissions of chemicals that have short characteristic travel distances, and processes that can be “moved” around a country or a region, such as extensive supply chains.

Supply chains

Currently, most life-cycle assessments consider only supply chains as aggregated into the main inventory. However, several questions specific to the supply chains exist: What type of technology do they use? How do they change over time? Where are they located? What routes and modes do they use for transportation? How does the location of supply chains influence the characterization factor of the different pollutants? What is the influence of geographic differentiation on final life-cycle assessment results when modifying the locations of the supply chains? This dissertation provides a practical approach to evaluate how the impacts associated with the emitted pollutants will change when changing the location of emissions of these pollutants.

1.4. Objectives of this dissertation

The objectives of this dissertation are to bridge the different areas of research in life-cycle assessment related to geographical differentiation and fill some of the different gaps that exist, with a focus on intake fractions and characterization factors in North America. In this dissertation, I aim at identifying what is important for regionalization of impacts, with a focus on damage to human health and how regionalization can be practically implemented in life-cycle assessment. The influence of geographically differentiated characterization factors on final life-cycle assessment results is evaluated for the extended supply chain of fly ash disposal and fly ash concrete.

1.5. Method

In this dissertation, I explore how life-cycle assessment results can be influenced when the location of emissions is taken into account.

In a first step, I aim to adapt existing (Chapters 2 and 4) — or developing new (Chapters 6, 7 and 8) — fate, exposure and effect modeling approaches to the case of North America to calculate regionalized intake fractions and characterization factors and to explore the importance of regionalization in life-cycle impact assessment (Chapters 2, 3, 4, 6, and 7).

In a second step, I explore how regionalization influences life-cycle assessment results using different case studies (Chapters 5, 8, 9, and 10).

1.6. Outline

In this section, I outline the present dissertation.

In Chapter 1, I have introduced the general concepts of life-cycle assessment as well as the state of the art regarding the issue of geographic differentiation.

In Chapter 2, I develop, describe and test IMPACT North America, a spatially resolved model that can calculate geographically differentiated population intake fractions and toxicological impacts in a North American context. This chapter demonstrates the difference between a spatially differentiated assessment and a generic assessment using a case study of diesel emissions.

In Chapter 3, I use the model developed in Chapter 2 to explore how regionalization influences the intake fractions of pollutants. I suggest how generic intake fraction and characterization factor should be computed and suggest archetypes for implementation.

In Chapter 4, I review and recommend intake fractions, effect factors and characterization factors for PM that are straightforward to apply to life-cycle assessment, in a manner consistent with currently used approaches for organic pollutants in Chapter 2.

In Chapter 5, I use the intake fractions and characterization factors developed in Chapters 2 and 4 as well as the archetypes developed in Chapter 3 to evaluate the influence of regionalization on the human health damage of different processes, comparing the two approaches geography-based or archetype-based to perform regionalization.

In Chapter 6, I explore the concept of uptake fraction (uF), an approach that explicitly addresses particle size distribution in the evaluation of environmental fate, human exposure and health effects of PM.

In Chapter 7, I present UFPM (Uptake Fraction of Particulate Matter), a model developed to evaluate the uptake fraction (the concept defined in Chapter 6) of primary PM.

In Chapter 8, I use the concept of uptake fraction developed in Chapter 6 and calculated in Chapter 7 to calculate the effect factor and characterization factor associated with different types of PM.

In Chapter 9, I apply the characterization factors developed in Chapter 2 and Chapter 4 to the case of fly ash disposal.

In Chapter 10, I apply the characterization factors developed in Chapter 2 and Chapter 4 to the case of fly ash concrete.

In Chapter 11, I summarize the main contributions of this dissertation.

1.7. Acknowledgments

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2. Assessing regional intake fractions and damage factors in North America

When starting to develop the model to assess regional intake fractions and damage factors in North America by reviewing the literature and assessing work under way in other institutions, it became apparent that the very similar work was being conducted at the École Polytechnique de Montréal for the development of such a model for Canada. Rima Manneh was developing the Canadian model, supported by her advisors, Manuele Margni and Louise Deschênes. Instead of competing, we decided to combine our work to create a better and more accepted model by both parties. I was in charge of developing the United States part of the model and Rima Manneh was in charge of developing the Canadian part of the model. Both parts would eventually be combined into one North American model. Furthermore, Shanna Shaked and Cedric Wannaz from the University of Michigan, supported by their advisor Olivier Jolliet, were calculating the air advection (i.e., flows of air, in m^3/h , in and out of each grid, for different layer of the atmosphere) at a $2^\circ \times 2.5^\circ$ grid level throughout the entire world. These data are information that will be needed in the present chapter to model the advection of pollutants in the air. This resolution would never be achieved by the work of one person only. Thus it was decided to use their part of the air layer to complete our model.

The outcome model is therefore a state of the art model, accepted by the institutions that participated in its development, three institutions playing a major role within the field of life-cycle assessment in North America.

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2.1. Summary

This chapter develops the IMPACT North America model, a spatially resolved multimedia, multipathway, fate, exposure and effect model that includes indoor and urban compartments. IMPACT North America allows geographic differentiation of population exposure of toxic emissions for comparative risk assessment and life-cycle impact assessment within United States and Canada. This model examines air, water, soil, sediment and vegetation media, and divides North America into several hundred zones. It is nested within a single world box to account for emissions leaving North America. It is a multiscale model, covering three different spatial scales – indoor, urban and regional – in all zones in North America. Model results are evaluated against monitored emissions and concentrations of benzo(a)pyrene, 2,3,7,8-tetrachlorodibenzo-p-dioxin (2,3,7,8-TCDD) and mercury. Most of the chemical concentrations predicted by the model fall within two orders of magnitude of the monitored data. The model shows that urban intake fractions are one order of magnitude higher than rural intake fractions. The model application and importance are demonstrated by a case study on spatially distributed emissions over the life cycle of diesel fuel. Depending on population densities and agricultural intensities, intake fractions can vary by eight orders of magnitudes, and even limited indoor emissions can lead to intakes comparable to those from outdoor emissions. To accurately assess

these variations in intake fraction, the essential three original features described in the present chapter are required: (i) inclusion of the continental model within a world box for persistent pollutants, (ii) addition of an urban box for short- and medium-lived substances (for grid size larger than 100 km), and (iii) assessment of intake from indoor emissions. This model can therefore be used to screen chemicals and assess regionalized intake fractions within North America for population-based human exposure assessment, life-cycle impact assessment, and comparative risk assessment. The model can be downloaded from <http://www.impactmodeling.org>.

2.2. Introduction

This chapter aims to develop a multimedia fate and exposure model that provides spatial intake fractions and damage factors for North America. It meets the need of accounting for long-range transport on a global scale while also capturing short-range variations in exposure related to differing population densities and agricultural production intensities.

Identifying pollutants that have potential adverse effects for the human population and setting priorities for emission controls require efficient and predictive screening tools that help us understand the potential risk of chemical releases into the environment. Comparative risk assessment and life-cycle impact assessment are typical examples of methodologies providing indicators of toxicological effects based on the relative risk and associated consequences of chemicals that are released into the environment (Pennington et al. 2004, ISO 2006a, 2006b, Rosenbaum et al. 2008). These consequences often occur as regional or local impacts (Potting et al. 2005, Potting and Hauschild 2006, Sedlbauer et al. 2007, Reap et al. 2008), making it important to evaluate them within a regional context.

Multimedia fate and multipathway human exposure models have been developed to support various scientific, regulatory, and educational purposes and are now widely adopted in assessments of the toxicological risks of chemical emissions at a regional scale (Cowan et al. 1994, European Commission 1996, Mackay 2001). Historically these single box models have provided estimates of risks for generic or non-spatial multimedia environments (e.g., CalTOX – McKone 1993, SimpleBox 2.0 – Brandes 1996, USEtox – Rosenbaum et al. 2008). However, single box models do not allow the differentiation of fate and exposure of pollutants based on emission location. To overcome this limitation, more recently several spatially resolved models have been developed to evaluate the impact of pollutants for different locations of emissions at a continental level. IMPACT 2002 (Pennington et al. 2005), a spatially resolved model for Western Europe, consists of 157 air cells, 136 watersheds and 125 ocean zones. BETR North America (MacLeod et al. 2001) consists of 25 air cells and watersheds/ocean zones. As demonstrated by these publications' results, disregarding spatial differentiation might under- or over-estimate certain intake fractions by several orders of magnitude and may therefore not be sufficient to distinguish the impacts between two emissions taking place in different distinct locations. Additional efforts at a global level (Huijbregts 2003, Toose et al. 2004, MacLeod et al. 2005, Sleswijk 2006, Rochat et al. 2006) have come to similar conclusions, although differences are less important because of a lower resolution scale (at a continental or a country level). An additional disadvantage of the single box models is that decision-makers and practitioners are generally more skeptical of impact assessment results using generic data instead of regional data. Thus, there is an increasing demand for methods reflecting regional concerns and adapted to regional conditions. (Potting and Hauschild 2006, Sedlbauer et al. 2007, Reap et al. 2008). Fulfilling this need is exactly the aim of this chapter, with three key differences from

the previous effort of MacLeod and colleagues (2001): (i) increased areal resolution by two orders of magnitude to capture effects occurring over a few thousand km², (ii) increased consistency in total pollutant damage by including an outer world box to account for emissions leaving North America, and (iii) the possibility to consistently model the intake fraction for indoor and urban emission within each zone. The increased resolution helps better describe short-lived pollutants, since Pennington et al. (2005) suggested that a 2° latitude by 2.5° longitude (2° × 2.5°) grid might still be insufficiently resolved to capture differences among population densities. Finally, by embedding North America within a world box, long-lived pollutants and their potential for damage outside North America or re-entry into North America are correctly addressed. Indeed, as shown by Margni (2003), intake of long-lived pollutants such as carbon tetrachloride and hexachlorobutadiene can be dominated by exposure outside of North America increasing total intake by up to one order of magnitude.

Note that in this chapter (in the model I am developing for North America), I am interested to evaluate what is the potential damage caused by emissions originating from North America. In the model developed in this chapter, I am therefore only looking at the marginal incremental damage from North American emissions and I am not looking at the actual damage to populations inside or outside North America from emissions originating outside North America. Therefore, in the model developed in this chapter, for emissions entering North America, I only look at the fraction that was actually emitted in North America and left North America before re-entering North America. However, if one wants to evaluate the damage caused by emissions occurring outside of North America (as I will do in Chapters 5, 9, and 10), one needs to adapt models or find alternatives models to also look at the damage from emissions occurring outside of North America. Extending the model developed in this chapter to the full world is outside of the scope of my dissertation but is a work in progress outside of this dissertation (e.g., Jolliet et al. 2008).

Objectives

This chapter has four main objectives. The first is to develop and describe a spatially resolved model that can calculate geographically differentiated population intake fractions and toxicological impacts in a North American context, covering the following three spatial scales: direct surroundings (indoor or outdoor); local scale (urban or non-urban); and the regional scale (covering North America at a resolution length of 100-200 km). The second objective is to evaluate the model results against monitored concentrations and empirical intake fractions in North America and against two other spatially resolved models for North America and Western Europe – BETR (MacLeod et al. 2001) and IMPACT 2002 (Pennington et al. 2005), respectively – to identify essential model features. The third objective is to show the importance of specifically considering urban areas in the modeling of intake fractions. Finally, this chapter aims to demonstrate the difference between a spatially differentiated assessment and a generic assessment using a case study of diesel emissions.

2.3. Methods

Model framework

Assessing the toxicological effects on human health of a chemical emitted into the environment requires a cause-and-effect chain assessment linking the emission source to damage through four intermediary parameters as depicted in Figure 2-1 and Equation 2-1 (based on Rosenbaum et al. 2007). The damage score (D , in units of impact) caused by a chemical emission (S , in mass_{emitted}) is obtained as the product of this chemical emission and a fate factor

(FF , in time), an exposure factor (XF , in time^{-1}), a dose-response factor (DR , in $\text{case}/\text{mass}_{\text{intake}}$), and a severity factor (SF , in $\text{impact}/\text{case}$):

$$D = SF \times DR \times XF \times FF \times S = EF \times iF \times S = CF \times S \quad (2-1)$$

The fate and exposure factors can be combined into one single metric, namely the intake fraction (iF , in $\text{mass}_{\text{intake}}/\text{mass}_{\text{emitted}}$). The intake fraction is the fraction of pollutant taken in by an exposed population, which is the mass taken in per unit of mass emitted (Bennett et al. 2002). The dose-response and severity factors can be combined into the effect factor (EF , in $\text{impact}/\text{mass}_{\text{intake}}$). In comparative risk assessment and life-cycle impact assessment, fate, exposure, and effect factors are usually grouped into characterization factors (CF) or human damage factors (HDF), which express the potential impact per unit of mass emitted ($\text{impact}/\text{mass}_{\text{emitted}}$). This latter metric is used by practitioners to weigh chemical emissions by their human toxicity potential.

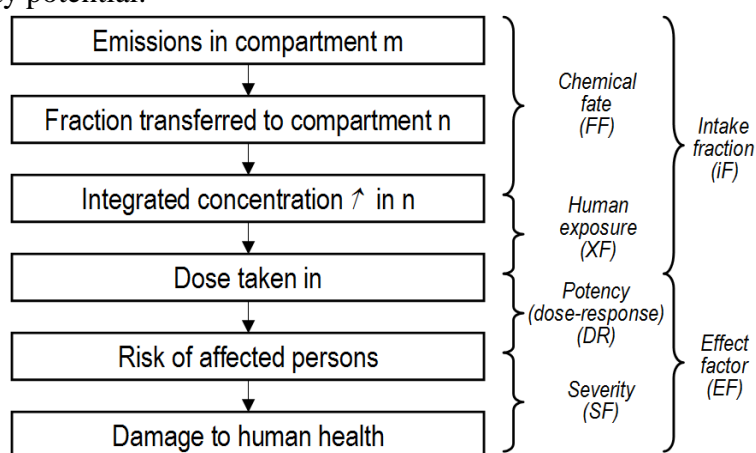


Figure 2-1: General source-to-impact framework for human toxicity (based on Pennington et al. 2005).

The IMPACT North America model

Starting with the model developed by Pennington and colleagues (2005), IMPACT North America extends this work by adding an indoor and an urban environment to create a multimedia, multipathway, fate, exposure, effect, and damage steady-state model (Figure 2-2). It predicts chemical concentrations in environmental media for direct indoor surroundings as well as at local (urban), regional and global scales. Furthermore, it predicts multiple exposure pathways that link chemical concentrations in the atmosphere, soil, surface water, and vegetation to human intake through inhalation and ingestion. It allows the calculation of human toxicity (carcinogenic and non-carcinogenic effects) and aquatic and terrestrial ecotoxicity caused by organic and inorganic pollutants. However, the modeling of aquatic and terrestrial ecotoxicity is not the focus of this chapter. The effect factors modeling is based on the following approach described in Jolliet et al. (2003): human toxicity is expressed in terms of disability adjusted life years (DALYs) using Crettaz et al. (2002) and Pennington et al. (2002) for carcinogenic and non-carcinogenic effects respectively; aquatic and terrestrial ecotoxicity is expressed in terms of the time- and space-integrated potentially affected fraction of species ($\text{PAF} \cdot \text{m}^3 \cdot \text{y}$) using Payet (2005).

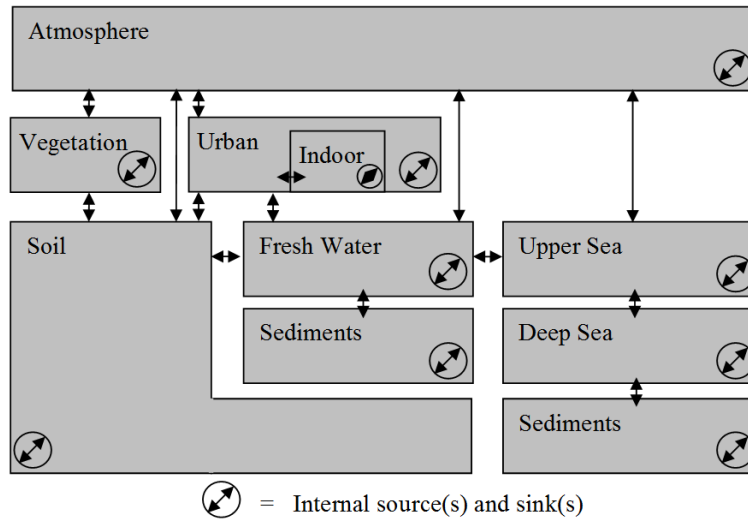


Figure 2-2: Framework for the fate modeling (based on Margni (2003), Margni et al. (2004) and Pennington et al. (2005)).

Model parameterization

In this dissertation, when referring to North America, I implicitly refer to only United States and Canada and exclude Mexico. Although Mexico can also be considered to be part of North America, because of lower availability of data for Mexico, I decided to focus my analysis on United States and Canada.

The IMPACT North America model (Figure 2-3) is nested in a world box. The three scales of the IMPACT North America model are: (i) direct surroundings (indoor or outdoor); (ii) local scale (urban or non-urban); and (iii) regional scale (which air cell, watershed or ocean zone is considered). The detailed map containing all of the regional labels is provided in section 2.8.

IMPACT North America

Version 1 (April 2009)

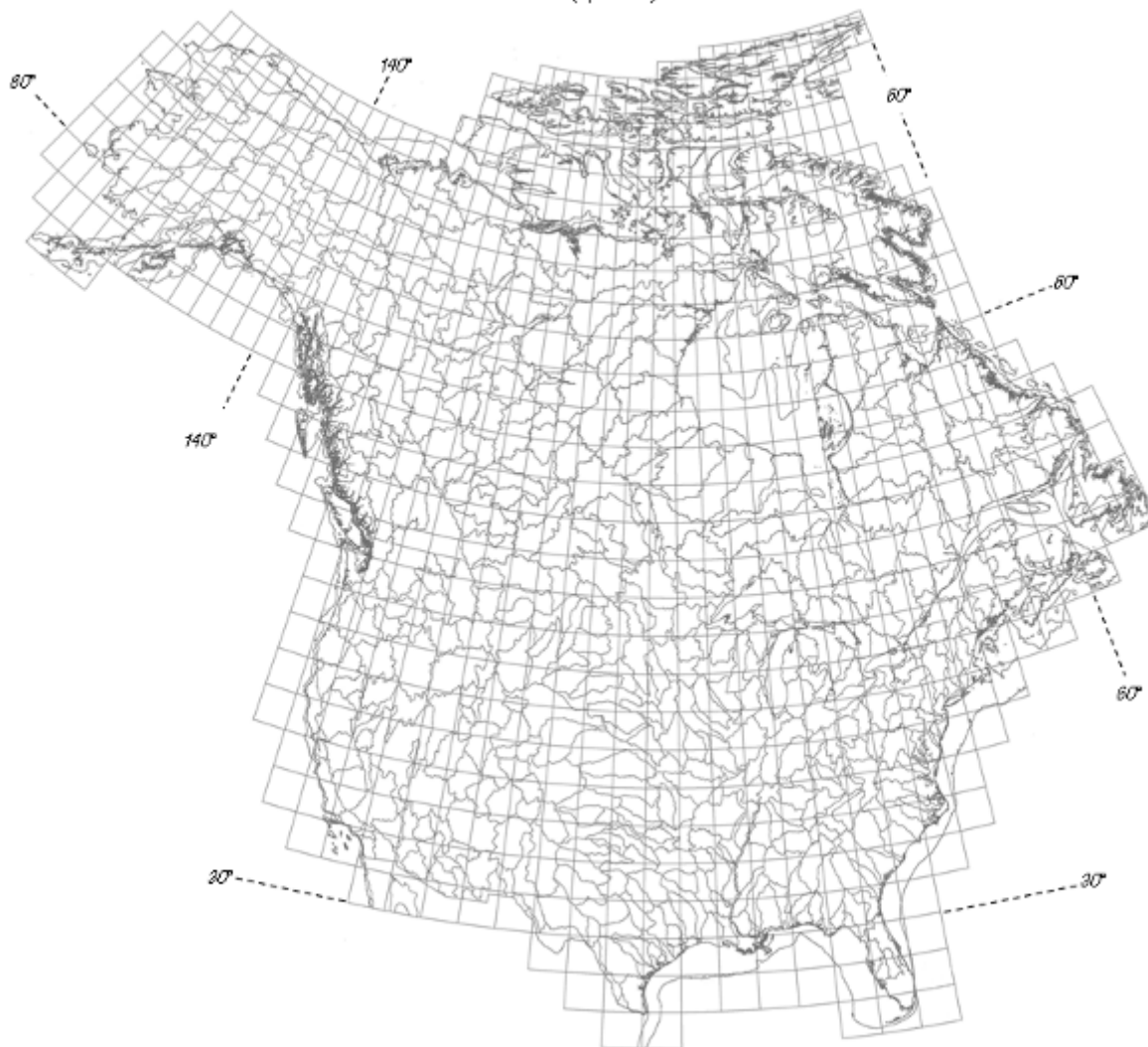


Figure 2-3: Overview of the geographical extent of IMPACT North America.

World box: The parameterization of the box representing the rest of the world outside of North America is based on the IMPACT World model (Shaked et al. 2008).

Air cells for North America: The North American air medium is divided into 831 air cells (version 1.0), each 2° latitude by 2.5° longitude ($2^\circ \times 2.5^\circ$). Atmospheric modeling is based on one air layer, with the mixing height set to 1,000 m (Rosenbaum et al. 2008). I used the wind transport simulated by Shanna Shaked from the University of Michigan by using 6-hour average wind speeds of the lowest three atmospheric layers (up to 962 m) from the GEOS-Chem (2007) global 3-D model of tropospheric chemistry driven by assimilated meteorological observations from the Goddard Earth Observing System of the NASA Global Modeling and Assimilation Office (GEOS-Chem 2007). The North, South, East and West wind speeds are each arithmetically averaged to yield $2^\circ \times 2.5^\circ$ resolution annually averaged wind speeds. (Note that by averaging annually, I remove the temporal resolution. Further research would be needed to evaluate the influence of time in addition to space when calculating fate and exposure to

pollutants.) The advection of each air cell is evaluated using the wind speed in each direction multiplied by the mixing height of the air cell.

Watersheds for North America: The 523 watersheds (version 1.0) are based on the sub-watersheds of North America, using NRCAN (2003) for Canada and USGS (1996, 2005)¹ for United States.

Coastal zones: Five coastal zones (version 1.0) are defined around North America (NGDC 2006): one in the West (Pacific), one in the East (Atlantic), one in the North (Arctic), and one in the South-West of Alaska (Bering), each linked to the ocean defined in the world box, plus the Hudson Bay, which is linked to the Arctic coastal zone.

Urban box: The air cells within the 48 contiguous states and lower Canada (i.e., where most of the North American population lives) are 30,000 km² to 50,000 km² in area, which provides inadequate resolution to capture differences between urban and rural emissions in areas that have a high variability in population density such as California (Pennington et al. 2005, Humbert and Horvath 2006). To overcome this limitation, the model provides the option of including an urban box within any air cell or watershed where the urban area is situated. The urban box is spatially differentiated to provide location-specific populations and areas for 292 urban areas (version 1.0) in both the United States and Canada (Demographia 2008), capturing 60% of North America's population. If no information is available about the urban area where a pollutant is emitted, a default urban area can be parameterized as follows (in version 1.0): a square area of 49 km × 49 km (population-weighted average value for United States urban areas — USDOT 2006), a population density of 753 persons/km² (USDOT 2006) (representing 1.8 million people), a mixing height of 250 m (harmonically averaged mixing height of 75 urban areas, based on the Marshall et al. (2005) analysis of USEPA SCRAM mixing height data (USEPA 2006), and a dilution rate of 610 m²/s (Marshall et al. 2005). Non-urban emissions are represented by emissions within the air cell or watershed, without including an urban box. The model provides the option to simultaneously run emissions in both the urban and non-urban boxes of a given region.

Indoor box: If emitted indoors, a typical pollutant can have an intake fraction up to three orders of magnitude higher than if emitted outdoors (Smith 1988). For indoor emissions, there is clearly a need to model the intake using an indoor box (Meijer et al. 2005a, 2005b, Nazaroff 2008, Hellweg et al. 2005, 2009). An optional, simplified indoor box is included in the model to allow for such emissions. The parameterization of the indoor box can be changed as desired. Default parameters for household, office and industrial indoor emissions are suggested in section 2.8, giving inhalation intake fractions of, respectively, 4.7×10^{-3} , 1.6×10^{-3} , and 3.3×10^{-5} kg_{inhaled}/kg_{emitted}.

The fate and exposure parameters that are adapted to the North American context are related to environmental conditions and human exposure. The *environmental parameters* include data on geographical (watershed area, fresh water area, amount of biomass, etc.), meteorological (mixing height, temperature, precipitation, air advection, etc.), and hydrological (fresh and coastal water depth, water flow rates, etc.) conditions. The *human exposure parameters* include data on demographics, fractions of drinking water coming from surface water, vegetable production, and animal production. Table 2-1 presents the major parameters that are adapted to the North American context. The detailed list of parameters is provided in section 2.8. Data can be contained and managed in ArcGIS.

¹ HUC6 level of the 1:2,000,000-Scale Hydrologic Unit Boundaries.

Table 2-1: Parameters adapted to the North American context (the complete list of parameters can be found in section 2.8).

Parameter adapted	Source (version 1.0)	Variability (version 1.0)
Population	CIESIN (2005)	<1 (Arctic area) up to ~2,000 (urban area) pers/km ² for watersheds; <1 to 440 pers/km ² for air cells (more than 100 pers/km ² in the cells capturing the top twelve big cities, with a peak at 440 pers/km ² in the cell capturing New York City)
Agricultural intensity	USGS (2005), FAO (2005), USDA, Agriculture Canada, Statistics Canada	<1 (Arctic area) up to ~400 (Midwest) t/km ² -y
Fresh water area	Natural Resources Canada, USGS	Fraction of water: <0.2% (desert area) up to 100% (Great Lakes, each considered as one watershed)
Precipitation	Environment Canada, Extrapolation from rainfall maps	90 (Arctic area) to 2,000 (North West Pacific and South Mississippi region) mm/yr
Fresh water fish production	Extrapolation from FAO, USGS, Statistics Canada	<1 (desert area) up to ~30,000 (coastal area) t/watershed-yr
Sea water fish production	Extrapolation from FAO, Statistics Canada. Extrapolation is based on the fact that 89% of seawater fish are caught in the coastal zones and 11% in open oceans	10,000 to 30,000 t/(coastal-zone-y)
Air advection	GEOS-Chem (2007)	0.1 to 7.6 m/s, with median of (in m/s) 2.7 (eastward), 1.8 (westward), 2.3 (northward) and 2.4 (southward)

2.4. Results and discussion

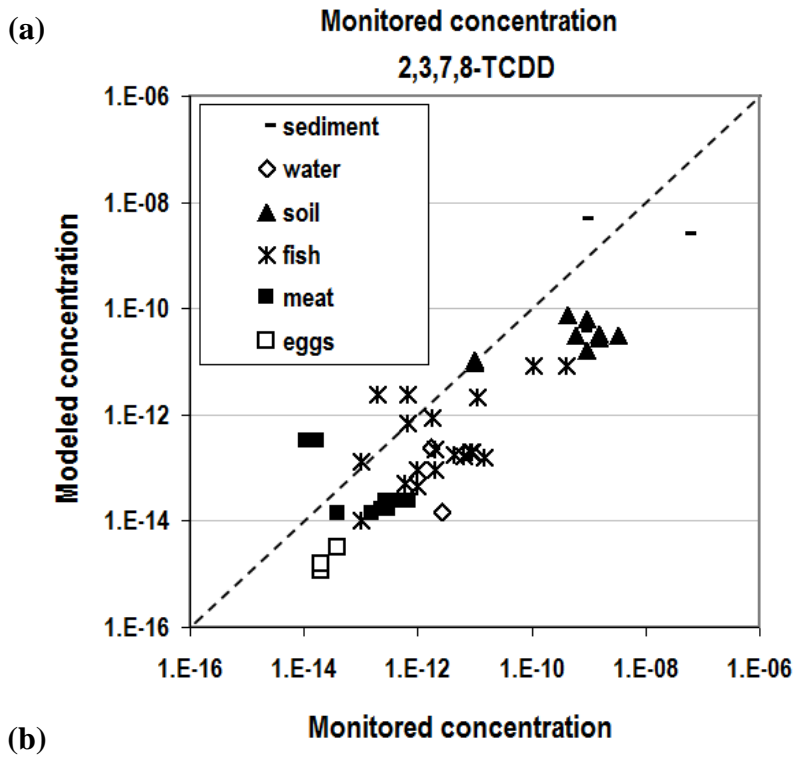
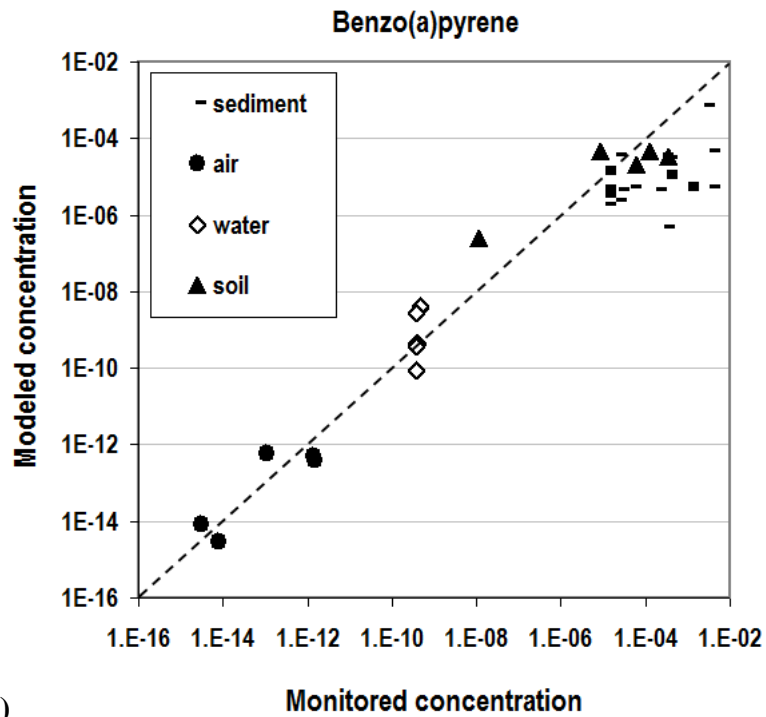
Model evaluation

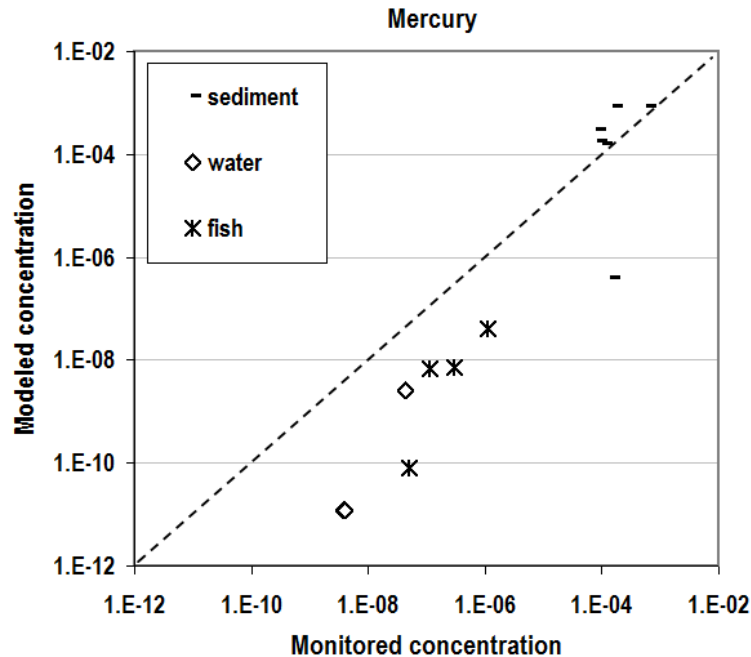
This model is compared with (i) monitored data of emissions and concentrations of benzo(a)pyrene, 2,3,7,8-tetrachlorodibenzo-p-dioxin (2,3,7,8-TCDD), and mercury in North America, (ii) results of the BETR North America (MacLeod et al. 2004), another spatially resolved model for North America with lower spatial resolution, and (iii) results of IMPACT 2002 (Pennington et al. 2005), a spatially resolved model for Western Europe, upon which the modeling framework of IMPACT North America is based.

Comparison with monitored data

Monitored emissions and concentrations of benzo(a)pyrene, 2,3,7,8-TCDD, and mercury are used to evaluate the model. The amount and distribution of emissions, the corresponding reported concentrations and their locations, and the list of physico-chemical properties of these substances are provided in section 2.8.

Figure 2-4 shows the concentrations modeled with IMPACT North America (version 1.0) versus the concentrations monitored in the environment.





(c)
Figure 2-4: Modeled versus monitored concentrations in the environment, for (a) benzo(a)pyrene, (b) 2,3,7,8-TCDD, and (c) mercury. Only the compartments where monitored concentrations are available are shown. Concentration in air and sediment is given in kg/m³, concentration in water, soil, meat, fish and eggs is given in kg/kg.

Most concentrations evaluated with the model fall within two orders of magnitude (maximum four orders of magnitude) of the monitored concentrations for sediment, air, water, soil, fish, meat and eggs.

Concentrations in sediment for benzo(a)pyrene show the highest difference between modeled and monitored concentrations – the latter being higher – which can be related to the persistency in sediment and the high uncertainty of the degradation rate in sediments. 2,3,7,8-TCDD also exhibits a weak correlation between monitored and modeled meat concentrations, which may be related to the fact that monitored concentrations do not necessarily report background concentrations or to the fact that meat is not necessarily produced in the watershed where the concentration is monitored. For benzo(a)pyrene and 2,3,7,8-TCDD, the natural background levels might not contribute significantly to monitored concentrations. However, for mercury, there may be a sizeable contribution from natural background to the monitored concentrations that is not included in the emission vector included in the model. This could explain why monitored concentrations of mercury in the water and fish are higher than modeled concentrations. However, apart from one data point, currently monitored and modeled concentrations in sediment are similar. This could be explained by the fact that the model is a steady-state model, with the sediment compartment as the ultimate sink for mercury emissions; in reality, the sediment compartment has likely not reached steady state, so the model is overestimating sediment concentrations, which compensates for the underestimation of not including natural background mercury to yield modeled sediment values that are similar to measurements. Further research is needed to better understand the reasons for discrepancies in modeled and monitored concentrations.

Because of the reduced number of pollutants in this dataset (only three), no general conclusions are drawn in this chapter on the accuracy of IMPACT North America to evaluate

different classes of pollutants. However, over a range of monitored concentrations of more than 12 orders of magnitude, modeled concentrations always fall within four orders of magnitude and 80% are within two orders of magnitude, with some of them being as close as a factor of two.

Comparison with other models

Comparison with BETR North America (MacLeod et al. 2004) and IMPACT Western Europe (Pennington et al. 2005). Intake fractions (i.e., fate and exposure) for benzo(a)pyrene, 2,3,7,8-TCDD, benzene, and carbon tetrachloride evaluated with IMPACT North America are compared to the values modeled with BETR North America and IMPACT Western Europe. The ranges of intake fractions obtained for the four chemicals mentioned above, as calculated by the three models, are reported in Table 2-2. For the calculation of the “average intake fraction,” emissions are distributed in the different zones of the model proportionally to their population. I use population-weighted average intake fractions since, as a first approximation, air emissions are assumed to be correlated with population. For example, using the data from Greco et al. (2007), I find that emissions of primary PM_{2.5}, NO_x, SO₂, and NH₃ are better correlated with populations of the different United States counties than with their surface areas (R² of 0.39, 0.11, 0.86, and 0.06 for population and 0.14, 1×10⁻⁵, 0.02, and 0.04 for surface area for PM_{2.5}, NO_x, SO₂, and NH₃ respectively).

To enable an adequate comparison with the BETR model, the physico-chemical properties for the four pollutants have been changed from the default values in the IMPACT North America database to match those from MacLeod et al. (2004) (see section 2.8 for values used). In addition to these four chemicals, intake fractions for particulate matter (PM) calculated with IMPACT North America are also reported.

Table 2-2: Average, minimum and maximum intake fractions (in ppm) for IMPACT North America (version 1.0) compared to the intake fractions modeled with BETR North America (MacLeod et al. 2004) and IMPACT Western Europe (Pennington et al. 2005).

Pollutant (emitted into air)	Intake pathway	IMPACT North America (present model)						BETR North America (MacLeod et al. 2004)			IMPACT Western Europe (Pennington et al. 2005)		
		Indoor	Min	Max (without urban box)	Max (urban box)	Ave. (without urban box) *	Ave. (with urban box) *	Ave.* *	Min	Max	Ave.*	Min	Max
Benzo(a)-pyrene	Oral		~0.01 ^a	1000 ^{d,h}		400		92	0.4 ^c	465 ^d	600	300 ^{j,k}	1000 ^l
	Inhalation	4700	~0.0001 ^a	0.4 ^e	30 ^{e,f}	0.09	5	0.2			0.2	0.03 ^k	0.5 ^l
2,3,7,8-TCDD	Oral		~700 ^a	20000 ^d		10000		1139	8 ^g	5293 ^h	3000	1000 ^j	5000 ^l
	Inhalation	4700	~0.1 ^a	3 ^{e,h,m}	40 ^{e,f}	2	7	0.5			5	2 ^{j,k}	7 ^l
Benzene	Oral		~0.001 ^a	0.03 ^{d,h}		0.05		0.003			0.007	0.007 ^{j,k}	0.01 ^l
	Inhalation	4700	~0.5 ^a	4 ^m	40 ^{e,f}	3	8	0.8	0.02 ^{c,g}	3.17 ⁱ	4	4 ^{j,k}	7 ^l
Carbon tetrachloride	Oral		~0.5 ^a	1 ^f		1		0.01			0.7	0.6 ^{j,k}	0.7 ^l
	Inhalation	4700	~300 ^a	~300 ⁿ	~300 ⁿ	~300	~300	3.1	0.74 ^c	7.12 ⁱ	400	300 ^{j,k}	400 ^l
Particulate matter	Inhalation	4700	0.002 ^a	2 ^e	40 ^e	1	6	n/a	n/a	n/a	n/a	n/a	n/a

**population-weighted average; **single North American box; ^aNorth Alaska/Canada; ^bChicago; ^cAlaska; ^dCalifornia; ^eN.Y.; ^fL.A.; ^gNunavut; ^hMidwest; ⁱMexico City; ^jSouth Spain; ^kScotland; ^lThe Netherlands/Ruhr; ^mDetroit; ⁿDenver*

Results show that the intake fractions evaluated with the IMPACT North America model are between a factor of two and 100 higher than the intake fractions as calculated by BETR

North America (MacLeod et al. 2004) and one order of magnitude from the intake fractions calculated by IMPACT Western Europe. Carbon tetrachloride has an intake fraction in BETR North America more than two orders of magnitude lower than in both IMPACT models. This discrepancy probably occurs because BETR North America does not consider the exposure in the world box nor the feedback from the world box to North America. This global effect dominates the impact of very persistent substances in air in IMPACT North America, thus demonstrating the importance of considering the transfer to and from the world box.

Note that for persistent pollutants, the annual intake fraction can be estimated to be 7 ppm/y, which is the ratio between the mass of air annually breathed by the global population (6.2×10^9 pers (value in the model) $\times 13 \text{ m}^3/(\text{pers}\cdot\text{d}) \times 365 \text{ d/y} \times 1.23 \text{ kg/m}^3$ (Wikipedia) = $3.5 \times 10^{13} \text{ kg/y}$) and the total atmospheric mass ($5 \times 10^{18} \text{ kg}$ — Wikipedia). The half-life of carbon tetrachloride considered in the model is 86,700 h (= 10 y) (Table 2-9), giving an atmospheric residence time of 14 y (i.e., half-life divided by $\ln(2)$) in a case of exponential decay). Therefore, this back-of-the-envelope calculation indicates that the total intake fraction that I should obtain is $7 \text{ ppm/y} \times 14 \text{ y}$, which is 100 ppm. In Table 2-2, I obtain 300 ppm for North America and 400 ppm for Europe. This factor of three increase obtained with the IMPACT North America model could partly be due to the fact that the mixing height of 1,000 m is an underestimate for persistent substances (persistent substances live long enough to mix throughout the free troposphere, up to 10-15 km). In the IMPACT Western Europe model, the mixing height of 800 m is even lower, further increasing the modeled intake fraction. Note that the ratio of intake fractions due to the underestimated height is not directly proportional to the ratio of heights due to the decreasing atmospheric density with height. Another source of high intake can be the short-term local effect from emissions in a densely population area that can reach a few tens of ppm depending on the local conditions.

The (population-weighted) average intake fraction of 10,000 ppm for 2,3,7,8-TCDD is within a factor of five of the empirically based estimate of 2,000 ppm reported by Bennett et al. (2002) for North America and of 3,500 ppm reported by King et al. (1999) for Western Europe. Regional intake fractions estimated by BETR North America are either within the range (for benzo(a)pyrene) or lower (for 2,3,7,8-TCDD and carbon tetrachloride, because of the absence of a world box in BETR) than those estimated with IMPACT North America. For benzene, a relatively medium-lived compound, it appears that the zones of BETR are too big to accurately capture exposure to emissions within high population density areas. The range of intake fractions modeled with IMPACT North America is higher than within Western Europe, mainly because the minimum intake fractions are lower in North America (because of a significant number of low-population density cells in Northern Canada and Alaska). For carbon tetrachloride emissions in IMPACT North America, the range between the lowest and highest intake fractions is insignificant, a result of the high persistence of this substance in air, leading to a somewhat uniform concentration worldwide regardless of its emission location. Finally, as expected, the population-weighted average intake fraction tends to be closer to the high values than the low values, because the population-weighted average intake fraction gives more weight to emissions occurring in high population density areas. For example, the population-weighted average intake fraction for PM is less than an order of magnitude lower than the maximum value but three or more orders of magnitude higher than the minimum value.

Urban box and indoor emissions

The comparisons to existing models have all been done on a regional scale (100-200 km for IMPACT North America), because the existing models do not have higher resolutions.

However, within IMPACT North America, when an emission occurs in an urban box rather than in the region surrounding this box, the intake fraction can be more than an order of magnitude larger (Table 2-2 and Figure 2-5). This finding, in agreement with the findings of Heath et al. (2006), suggests that it is important for models using grids larger than 100 km in width to add an urban box to properly evaluate the intake fractions for emissions of short- and medium-lived pollutants occurring in urban areas. As a matter of comparison, the indoor intake fractions are between one and three orders of magnitude higher than the outdoor urban intake fractions. Therefore, to get an accurate impact score on human health, life-cycle assessment needs to also consider indoor intake from indoor emissions whenever the proportion of release that occurs indoors exceeds about 1% of the total release.

Variation in intake fractions across North America

The variations in oral and inhalation intake fractions across North America are shown in Figure 2-5. For each pollutant, Figure 2-5 also shows the minimum (min) and the maximum (max) intake fraction as well as the 5th, 25th, 50th, 75th and 95th percentile of the distribution of intake fractions.

Variations of up to 8 orders of magnitude for oral intake fractions (e.g., 2,3,7,8-TCDD) and up to 5 orders of magnitude for inhalation intake fractions (e.g., PM) are observed. The variations are limited to less than 3 orders of magnitude for benzene, which has a higher residence time in air than PM. For the substances shown, 95% of the emissions have intake fractions within two orders of magnitude for all areas and within one order of magnitude for urban areas. Intake fractions in urban areas are about one order of magnitude higher than rural intake fractions for each substance. The population-weighted distribution of intake fractions shown reflects the exposure of North American population (i.e., the likely intake fraction that an emission will have).

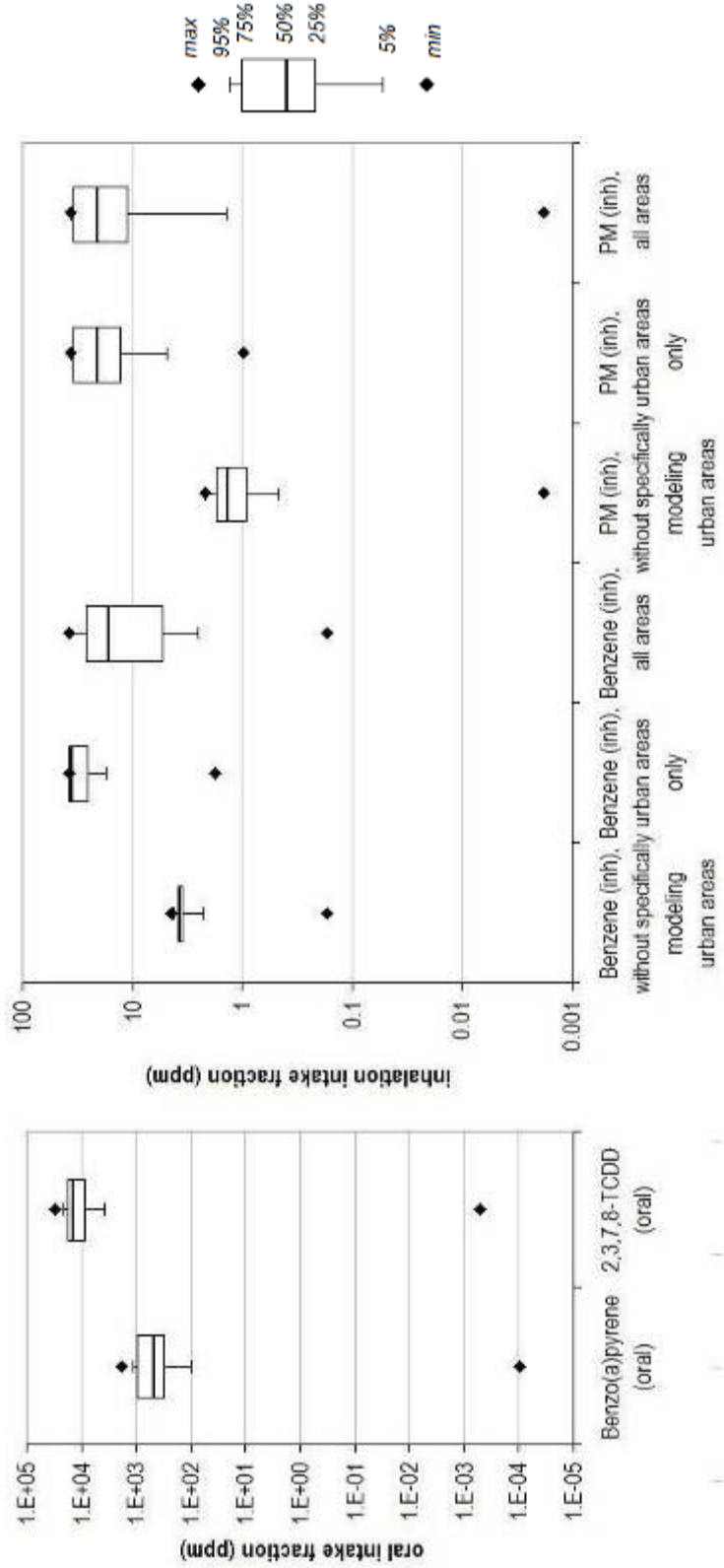


Figure 2-5: Variations in oral and inhalation intake fractions across North America. Percentiles are presented for the population-weighted distribution of intake fractions.

Application: Population intake of chemicals per km diesel car driven as a function of emission location and chemical properties, including a comparison between indoors and outdoors

Diesel car

To depict the variations in intake fractions within the North American continent when regionalization is included, I have applied the IMPACT North America model to three pollutants that are all emitted when driving a diesel car: PM_{2.5} (medium range, damaging through inhalation intake), benzo(a)pyrene (medium range, oral intake), and benzene (medium range, inhalation intake). Because the intake fraction from benzo(a)pyrene is dominated by oral intake, this intake fraction is proportional to agricultural production intensity in the locations of emission and deposition. Agriculture effectively pumps benzo(a)pyrene from the environment through vegetables and animals and into humans, so increased agricultural intensity can pump increased amounts of benzo(a)pyrene. This is in contrast to inhalation-dominated pollutants where lungs act as the pump and therefore increased concentrations of lungs (i.e., areas with higher population densities) have increased intake fractions.

These three chemicals are considered to be representative of a wide range of pollutants. Furthermore, PM_{2.5} is recognized as one of the most damaging pollutants to human health via inhalation (Kuenzli et al. 2000, Schwartz et al. 2008, Pope et al. 2009). These intake fractions are combined with the corresponding emissions, calculated per km driven by car, including both direct emissions and the fuel supply chain.

Fuel supply emissions are assumed to be emitted in a rural area in Texas, where both oil extraction and refining exists (and is characterized by medium population density and medium agricultural intensity), whereas tailpipe emissions from a diesel engine car are assumed to be emitted either in Chicago (characterized by urban area, high population density, situated in an area of high agricultural intensity) or in Alaska (characterized by remote area, low population density and low agricultural intensity). These three locations are fictive. The aim is to take three locations within North America with different population density and agricultural intensity patterns.

Table 2-3 reports the emissions and intake fractions used for the application example used in this case study. The life-cycle inventory for emissions during the supply chain and from the tailpipe is taken directly from ecoinvent (Frischknecht 2005). The intake fractions are modeled with IMPACT North America version 1.0.

Table 2-3: Emissions (life-cycle inventory (LCI)) and intake fractions (iF) for a diesel engine car.

Type of pollutant	LCI (g/km)		iF (generic, prop. to pop.)		iF (rural, Texas (cell AF29))		iF (urban, Chicago (cell AK25))		iF (remote, Alaska (cell N12))	
	supply	tailpipe	inh	oral	inh	oral	inh	oral	inh	oral
PM _{2.5}	0.0095	0.022	6.0E-6		4.6E-7		2.2E-5		2.9E-9	
Benzo(a)pyrene	1.4E-8	1.6E-5	5.1E-6	2.7E-4	1.2E-8	2.6E-4	2.1E-5	9.3E-4	3.0E-11	2.6E-10
Benzene	3.9E-4	0.0013	7.9E-6	1.2E-8	1.3E-6	2.4E-8	2.5E-5	2.0E-8	3.0E-7	9.6E-10

Note that the substance properties for benzo(a)pyrene are based on those reported in Table 2-9.

Reported in kg of intake per km driven by car, the total inhalation and oral intakes (Figure 2-6) can be calculated as the products of the emissions and the respective intake fractions (Table 2-3). I present the intakes calculated for a generic North American emission (i.e., an emission distributed proportionally to population), as well as those calculated using the regional emissions and intake fractions.

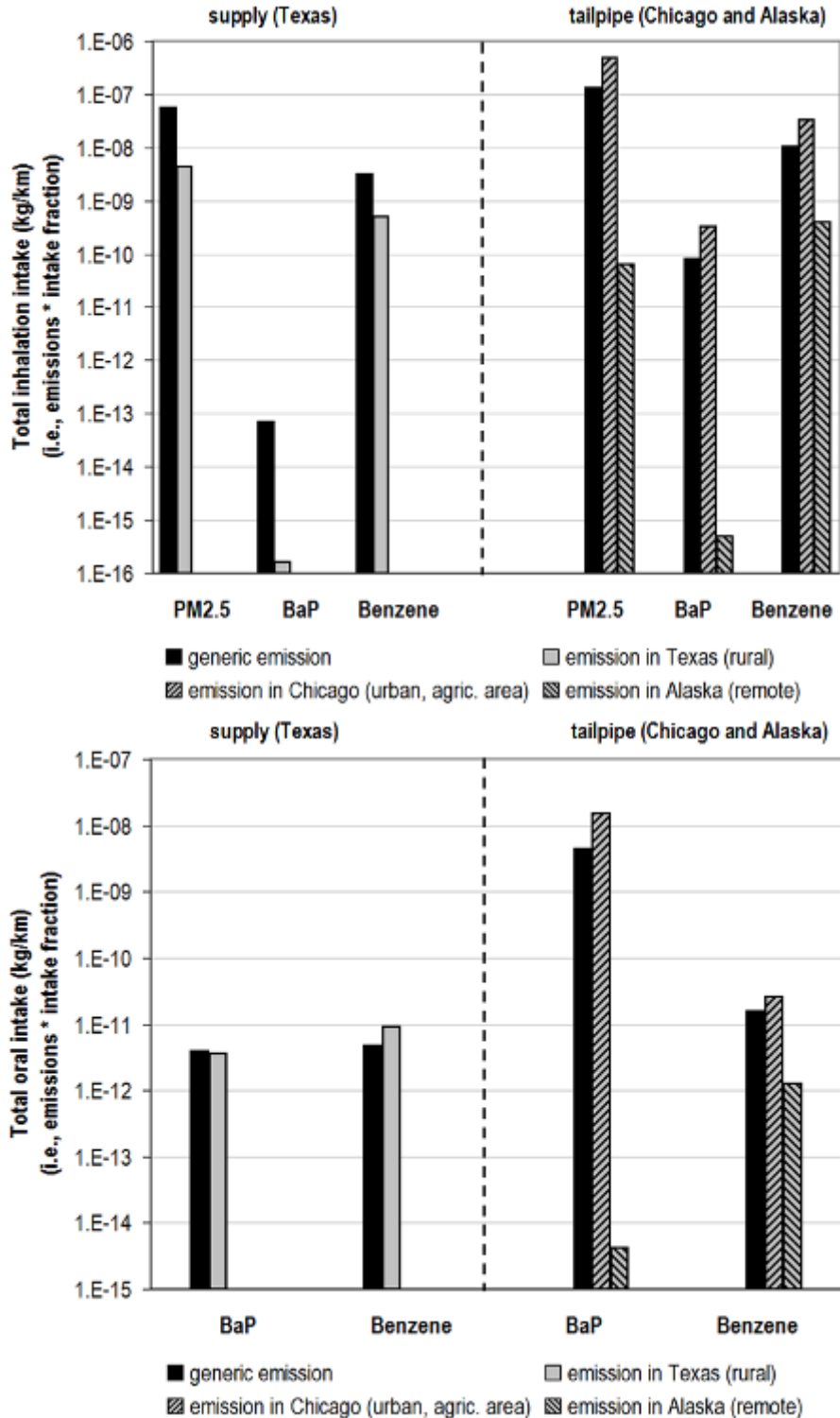


Figure 2-6: Total (i.e., the entire exposed population) inhalation and oral intake (in kg per km) for PM_{2.5}, benzo(a)pyrene (BaP), and benzene emissions from a diesel car, using generic intake fractions (in black) as well as regional intake fractions (in grey) for emissions in a rural area (Texas) in an urban area (Chicago) and in a remote area (Alaska). Note that PM_{2.5} is not considered toxic through oral ingestion and therefore is not included in the oral part of the figure.

For emissions in rural areas with medium population densities and medium agricultural intensities, such as Texas, most inhalation and oral intakes evaluated with the generic intake fraction are close to those calculated with a region-specific intake fraction (within an order of magnitude). For urban emissions, in a high population density and within a high agricultural intensity region (such as Chicago), inhalation intakes evaluated with a region-specific intake fraction are systematically higher than those evaluated with a generic intake fraction. The same observation can be made for oral intake. For emissions in remote areas, with low population densities and low agricultural intensities (such as Alaska), the intake can be overestimated by up to six orders of magnitude when using a generic intake fraction rather than a region-specific intake fraction (as demonstrated by benzo(a)pyrene tailpipe emissions). This example demonstrates the importance of regionalizing intake fractions when working with population densities and agricultural intensities far below or above average conditions.

Indoor versus outdoor

For comparison purposes, indoor emissions can also occur if the car is started or stopped inside a house-attached garage.

Batterman et al. (2007) reported that the average house-to-garage flows are $2.6 \text{ m}^3/\text{h}$, which is 4.9% of the garage's total air-exchange rate. This indicates an average ventilation rate for the garages of $53 \text{ m}^3/\text{h}$. The authors also report that the air flows from the garage to the house average $9.3 \text{ m}^3/\text{h}$. Therefore the fraction of air in the garage that flows into the house is, on average, $9.3 \text{ m}^3/\text{h}$ divided by $53 \text{ m}^3/\text{h}$, representing 18%.

Assuming a transfer factor of 0.18 from the garage to the house (Batterman et al. 2007), and that half of the household members are staying inside the house after the car started, the “adapted” household intake fraction is $4.2 \cdot 10^{-4}$. Considering that the population-weighted average intra-urban intake fraction for $\text{PM}_{2.5}$ is $1.3 \cdot 10^{-5}$ (see section 2.8), this suggests that if 3% of the emissions of $\text{PM}_{2.5}$ occur inside the garage (when departing or arriving), the total intake of $\text{PM}_{2.5}$ by the persons in the household is as high as the total intake of $\text{PM}_{2.5}$ by the rest of the urban area. Assuming constant emissions, the damage due to 28 seconds of the engine running inside the garage is equivalent to that from a 15 minutes car ride in the urban area, demonstrating that even limited indoor emissions can lead to significant intakes compared to outdoor emissions.

Finally, the phenomenon of self pollution (i.e., the exposure of the driver and passengers to their own vehicle’s emissions — “on-road amplification” of intake fraction) can also increase the total intake fraction of diesel emissions. Marshall and Behrentz (2005) showed that intake fraction from self pollution can be higher than total intake fraction. Marshall (2005) shows that the difference in individual intake fraction values between self-pollution and non-self-pollution is between five and six orders of magnitude. This phenomenon of self pollution requires more attention and should be considered in life-cycle assessment to better capture the total damage from diesel emissions.

2.5. Conclusions

Intake fractions in North America vary greatly depending on the emission location. To accurately assess these variations, the three original features proposed in the present chapter are essential: (i) the inclusion of the continental model within a world box to assess the full extent of persistent and long-range transport pollutants, (ii) the addition of an urban archetype for short- and medium-lived substances (if the grid size is larger than 100 km), and (iii) the option to assess intake associated with indoor emissions. The North American spatially resolved, multimedia, multipathway, steady-state model proposed by this chapter includes these three features and

provides results comparable in magnitude to monitored concentrations in different media and intake fractions predicted by other evaluated models. It can be used as a screening tool to estimate intake fractions and human damage factors for toxic emissions in North America. As a multiscale model, including indoor versus outdoor, urban versus non-urban, and regional scales, it can also be adapted to assess specific emission scenarios for processes situated in a known location in North America. These specific emission scenarios can be classified as situation-dependent (e.g., urban areas) (Sedlbauer et al. 2007), site-dependent (e.g., Iowa) or site-specific (e.g., Ames, IA) (Potting and Hauschild 2006). I suggest adopting the population-weighted average intake fraction as a default value for emissions occurring in unknown locations, based on the assumption that, at the resolution of this model (a few thousand to a few tens of thousands km² zones), emissions of many pollutants are correlated to population. This model can therefore be used to explore questions associated with regionalization within North America such as the following. (i) What is the variability of fate factors, intake fractions and, therefore, human damage factors among the different emission locations in North America? (ii) What level of spatial resolution is needed to be environmentally relevant, yet still affordable in terms of the amount of input data to collect and manage in practical applications? (iii) What parameters really matter, i.e., what is the influence on inhalation and oral intake fractions of various parameters, such as population density, food production intensity, residence time of water within the different watersheds, wind patterns, or pollutant half-lives?

In addition to regional exposure assessment or hazard screening studies, this model and its results are also intended to help the life-cycle assessment community address the issue of regionalization to keep life-cycle assessment as simple as possible, but as complex as necessary. Characterization factors estimated with this model can be directly used for human toxicity (carcinogenic, non-carcinogenic and respiratory effects) and ecotoxicity impact categories from damage-oriented life-cycle impact assessment methods such as IMPACT 2002+ (Jolliet et al. 2003), TRACI (Bare et al. 2003), Hofstetter (1998) or ReCiPe (Goedkoop et al. 2008).

Limitations. This model also presents several limitations. The model is not suited to assess impacts from a localized source (within a few kilometers from the source). To support more detailed and localized risk assessment purposes, there is a need for the development of an extended multiscale model that could work at a km scale around a plant, and be embedded in the present model to account for long-range transport. At the other extreme, advection outside North America needs to be better considered by complementing it with intercontinental transport. Finally, further research is needed to account for the food trade within North America and among continents to assess “embedded” transfer of pollutant emissions through food exports and imports (Jolliet et al. 2008).

The IMPACT North America model, along with a complete list of the characterization factors modeled using the default parameters indicated in the present chapter can be downloaded from <http://www.impactmodeling.org> or obtained by contacting me at sebastien.humbert@cal.berkeley.edu.

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2.8. Supporting information for this chapter

The “IMPACT North America” model, the values, the sources, as well as the updates can be found at <http://www.impactmodeling.org>, or obtained by contacting sebastien.humbert@cal.berkeley.edu.

2.8.1. Model framework

The model framework is based on the spatial version of IMPACT 2002 developed for Western Europe (Pennington et al. 2005).

2.8.2. Model parameterization

The model parameters that can be adapted to regional or local conditions are presented in Table 2-4. The numerical values are presented directly in the IMPACT North America model as Excel tables.

Table 2-4: Model parameters that can be adapted to regional or local conditions.

Watershed zone modeling:	Coastal/ocean zone modeling:	Air zone modeling:
<p>soil module: rainfall infiltration fraction (-) runoff rate (fraction of rainfall rate) (-) ppm of solids in runoff water (ppm) solid phase soil density (kg/m³) fraction organic carbon in solid phase of soil (-) area soil (m²) temp (K) air boundary layer thickness (m) rainfall rate (m/h)</p> <p>surface layer: depth (m) volumetric water fraction (-) volumetric air fraction (-) volumetric solids fraction (-)</p> <p>agricultural root zone: depth (m) volumetric water fraction (-) volumetric air fraction (-) volumetric solids fraction (-)</p> <p>vadose layer: depth (m) volumetric water fraction (-) volumetric air fraction (-) volumetric solids fraction (-)</p> <p>bulk vegetation module: dry plant mass (kg/m² soil) volume fraction leaf to aerial plant part (-) volume fraction root to aerial plant part (-) transpiration coefficient (l/kg) leaf area index (-) type of land cover (-) diffusion length stomata (m) diffusion length boundary layer (m) density root (bulk) (kg/l) correction plant lipid-n-octanol (barley) (-) root water content (g/g) root lipid content (g/g) density stem (bulk) (kg/l) correction plant lipid-n-octanol (barley) (-) stem water content (g/g) stem lipid content (g/g) density leaf (bulk) (g/g)</p>	<p>oceanic water module: depth of upper layer (m) suspended sediment phase fraction - top depth of lower layer (m) suspended sediment phase fraction–bottom (-) water pH (-) suspended sediment density (kg/m³) fraction of organic carbon in suspended sediment (-) vertical mixing velocity (m/h) area oceanic water (m²) temp (K) sediment deposition rate constant (m/h) sediment to water mass transfer coefficient (m/h)</p> <p>sediment module: depth of sediment (m) area sediment (m²) solids fraction (-) solid phase sediment density (kg/m³) fraction of organic carbon in solid phase of sediment (-) sediment to water mass transfer coefficient (m/h) sediment deposition rate constant (m/h) fraction of sediment deposition buried (-) fraction of sediment deposition resuspended (-)</p> <p>exposure module (usable production in considered zone): sea fish (kg/y)</p>	<p>air module: dry deposition velocity (m/h) wet deposition scavenging volume (m³ air/m³ rain) rain air boundary layer thickness (m) rainfall rate (m/h) average dry period (hours) lower to upper atmospheric loss rate const. (m/h) temp (K) aerosol solid density (kg/m³) height of lower atmospheric boundary layer (m) aerosol phase fraction (m³/m³ air) paved area with drainage systems (m²) area water (m²) area soil (m²) fraction of area soil covered by agri. vegetation (-) capture coefficient (klai) leaf area index (-) diffusion length stomata (m) diffusion length stomatal boundary layer (m)</p> <p>surface soil layer module: volumetric water fraction (-) volumetric air fraction (-) volumetric solids fraction (-)</p> <p>exposure module: population (number of persons) usable production in considered zone: sum unexposed produce (kg/y) sum exposed produce (kg/y) pigs (kg/y) beef (kg/y) broilers (kg/y) goat and Sheep meat (kg/y) eggs (kg/y) dairy products (cow milk) (kg/y)</p> <p>number of head: pigs (number (nb)) beef+veal (nb) broilers (nb of utility chicks of table strains hatched) goat and sheep (nb)</p>

Watershed zone modeling:	Coastal/ocean zone modeling:	Air zone modeling:
<p>correction plant lipid-n-octanol (barley) (-) leaves water content (g/g) leaves lipid content (g/g) temp (K) fraction of area soil covered by agri. vegetation (g) water module: mean depth (m) surface area (m²) water pH (-) temperature (annual average) (K) mean wind speed 10 m over surface (m/s) diffusion constant of oxygen in water at the temperature T (cm²/s) diffusion constant of water in air at the temperature T (cm²/s) sediment to water diffusion mass transfer coefficient (m/h) mass fraction of organic matter in suspended solids (-) concentration of colloidal organic matter per unit bulk volume (kg/m³) solid-to-water phase ratio (-) mean sediment accumulation (kg-s/(m-y)) sediment-land module: sediment to water mass transfer coefficient (m/h) fraction of sediment deposition buried (-) fraction of sediment deposition resuspended (-) solid phase sediment density (kg/m³) fraction of organic carbon in solid phase of sediment (-) mean sediment accumulation sediment depth (m) area sediment (m²) solids volume fraction (-) exposure module: population (number of persons) usable production in considered zone: sum unexposed produce (kg/y) sum exposed produce (kg/y) fresh water fish (kg/y) pigs (kg/y) beef+veal (kg/y) broilers (kg/y) goat and sheep (kg/y) eggs (kg/y) dairy products (cow milk) (kg/y) fraction of surface drinking water (-) number of head: pigs (number (nb)) beef+veal (nb) broilers (nb of utility chicks of table strains hatched) goat and sheep (nb) laying hens (nb) dairy cattle (nb) specific to emission modeling: fraction of burnable area (-) burnable area (km²)</p>		<p>laying hens (nb) dairy cattle (nb) specific to emission modeling: fraction of burnable area (-) burnable area (km²)</p>
<p>Finally, "Air advection among air cells" and "Water advection among watersheds" can be parameterized to adapt to regional or local climatic conditions, but by being careful to always conserve mass balance.</p>		

Other important parameters integrated in the model but not geographically dependent

The breathing rate used in IMPACT North America is 13 m³/(pers·d) (US EPA 1997). Note that MacLeod et al. (2004) (in the BETR model), Rosenbaum et al. (2008) and Van Zelm et

al. (2008) also use a breathing rate of 13 m³/(pers·d), whereas TRACI (Bare et al. 2003) and Greco et al. (2007) use 20 m³/(pers·d). The earlier versions of IMPACT models (Pennington et al. 2005) used a breathing rate value of 20 m³/(pers·d). Note that the value of 13 m³/(pers·d) that I use in the IMPACT North America model (US EPA 1997) might be slightly underestimating average population breathing rate — Stifelman (2007) reports mean values of 16 m³/(pers·d) for male and 13 m³/(pers·d) for female, giving an average of 14.5 m³/(pers·d).

2.8.3. Indoor box

The default indoor intake fraction (iF) is adapted from Hellweg et al. (2009) and evaluated as:

$$iF = f_{TE} \times N \times BR / (V \times m \times k_{ex}) \quad (2-2)$$

where *BR* is an individual's daily breathing rate of air (m³/(pers·d)), *N* is the number of people exposed (unitless), *V* is the volume of the exposure area (m³), *k_{ex}* is the air exchange rate of the volume in the exposure area (h⁻¹) and *m* is the mixing factor (unitless). To account for the time people spend inside, a corrective parameter can be applied, which is the fraction of the time exposed, *f_{TE}*. This correction factor is evaluated to be 0.7 for household, 0.3 for office and 0.6 (because of assumed two shifts) for industrial situations.

The following parameters are suggested by default (extrapolated from Hellweg et al. (2009)):

Household: *V/N* = 160 m³/pers, *m* = 1, *k_{ex}* = 0.5 per hour = 12 per day. Therefore intake fraction = 0.0068 = 6,800 ppm for full time exposure. This corresponds to 4,700 ppm with correction factor *f_{TE}*.

Office: *V/N* = 100 m³/pers (assumption), *m* = 1, *k_{ex}* = 1 per hour = 24 per day. Therefore intake fraction = 0.0054 = 5,400 ppm for full time exposure. This corresponds to 1,600 ppm with correction factor *f_{TE}*.

Industrial: *V/N* = 1,000 m³/pers, *m* = 1, *k_{ex}* = 10 per hour = 240 per day. Therefore intake fraction = 0.000054 = 54 ppm for full time exposure. This corresponds to 33 ppm with correction factor *f_{TE}*.

For indoor intake fraction estimates, Equation 2-2 is implicitly assuming that emissions are not temporally correlated with occupancy. This assumption might need further research to evaluate whether it is acceptable or not in a life-cycle impact assessment context.

For indoor industrial intake fraction estimates, Equation 2-2 is likely to underestimate the true intake fraction owing to the importance of near-field exposures in that setting. The value for indoor industrial intake fraction needs further research to increase its robustness in a life-cycle impact assessment context.

2.8.4. Urban box

The default urban box can be parameterized as follows:

Size: 49 km × 49 km (population-weighted average value for United States urban areas - USDOT 2006)

Population of the urban box: the population of the urban box is calculated by multiplying the size of the urban box (49 km × 49 km = 2401 km² — USDOT 2006) by the United States average urban population density (210,421,000 pers / 279,300 km² = 753 persons/km² — USDOT 2006).

Note that all these parameters can be adjusted by the user.

Intra-urban intake fraction: The intra-urban intake fraction (iF) of a pollutant emitted in an urban area i is computed as:

$$iF_i = \frac{a \times BR \times N_i}{u_i \times H_i \times W_i} = \frac{a \times BR}{u_i \times H_i} \times d_i \times L_i, \text{ with } d_i = \frac{N_i}{L_i \times W_i} \quad (2-3)$$

where N_i (in pers) is the number of persons in the urban area i , BR (in $\text{m}^3/(\text{pers}\cdot\text{d})$) is the breathing rate, a (unitless) is the correction factor to account for the facts that (1) a pollutant can be emitted anywhere in the urban area i and not only at the edge, and (2) the air that left the urban area i can come back with some of the pollutant (i.e., a back-and-forth movement of air) (this factor varies between 0.5 and 1, and can be approximated to 0.75 – Benarie (1980)), u (m/d) is the dominant wind speed of the urban area i , H (m) is the mixing height of the urban area i , L (m) is the length of the urban area i (measured in the direction of the dominant wind u), W (m) is the width of the urban area i (in general, in modeling, the urban area i is assumed square, thus $L = W$), and d (pers/m^2) is the population density of the urban area i . Equation 2-3 assumes that deposition and degradation rates within the urban area are negligible.

The North American population-weighted average intra-urban intake fraction, based on the 292 urban areas presented in version 1.0 of the model, is 13 ppm. This finding is coherent with the finding of Marshall et al. (2005), who found a population-weighted mean intra-urban intake fraction of 14 ppm for nonreactive gaseous vehicle emissions in U.S. urban areas.

2.8.5. Air zones

Figure 2-7 presents the layout of the air zones of IMPACT North America (version 1.0).

IMPACT North America Air, coastal and oceanic zones

Version 1 (April 2009)

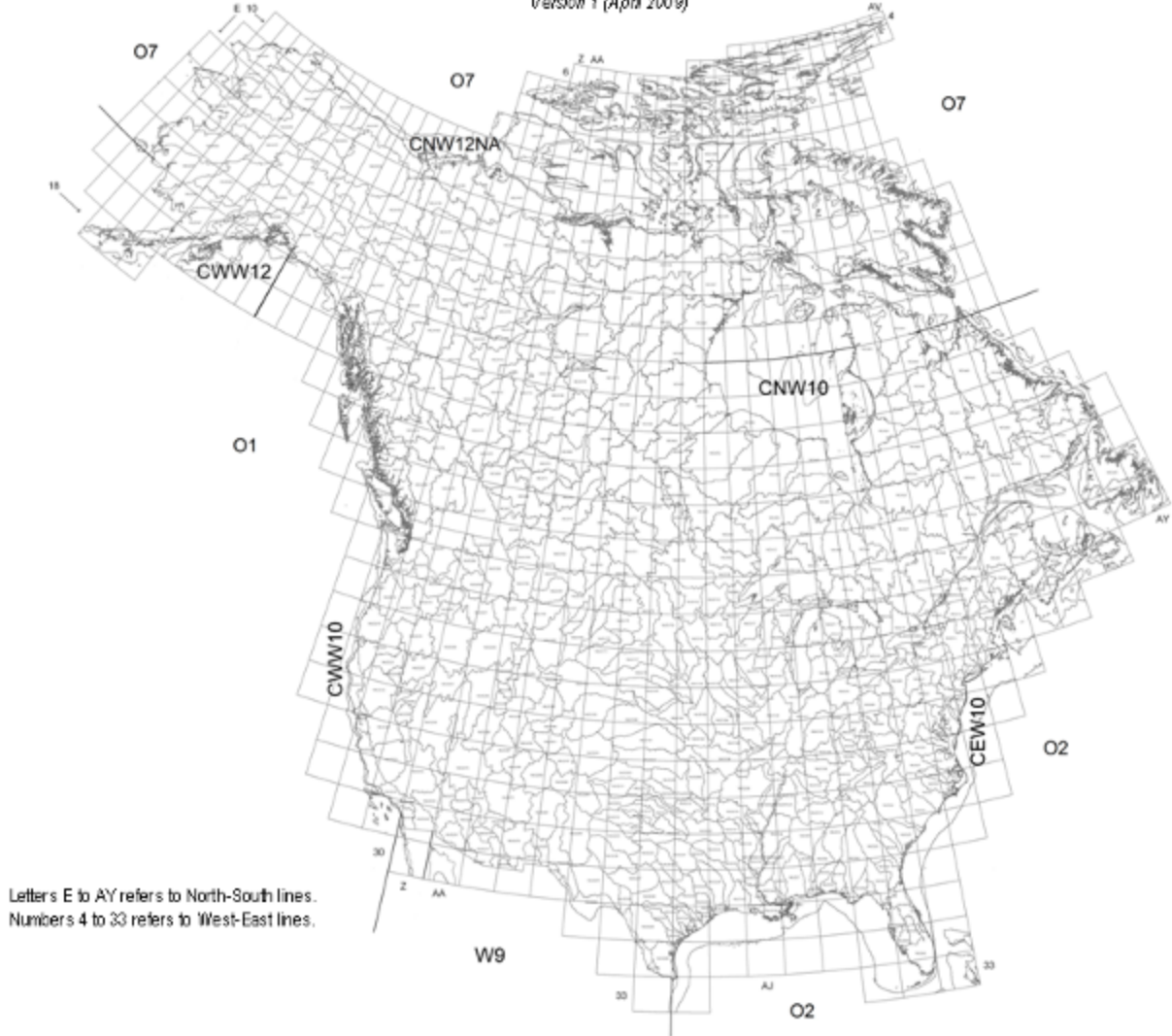


Figure 2-7: IMPACT North America: Air zones (version 1.0). Note that this figure aims at depicting only the big zones (e.g., O1, O2, ...) as well as the air zones numbering (e.g., Z, AA, ..., 30, ...). It is not the aim of this figure to show the watershed codes that are actually too small to be read on this figure. Numerical values for total and land area, population, population density and agricultural intensity can be found in Table 3-4.

2.8.6. Watershed zones

Figure 2-8 to Figure 2-11 present the layout of the watershed zones of IMPACT North America (version 1.0).

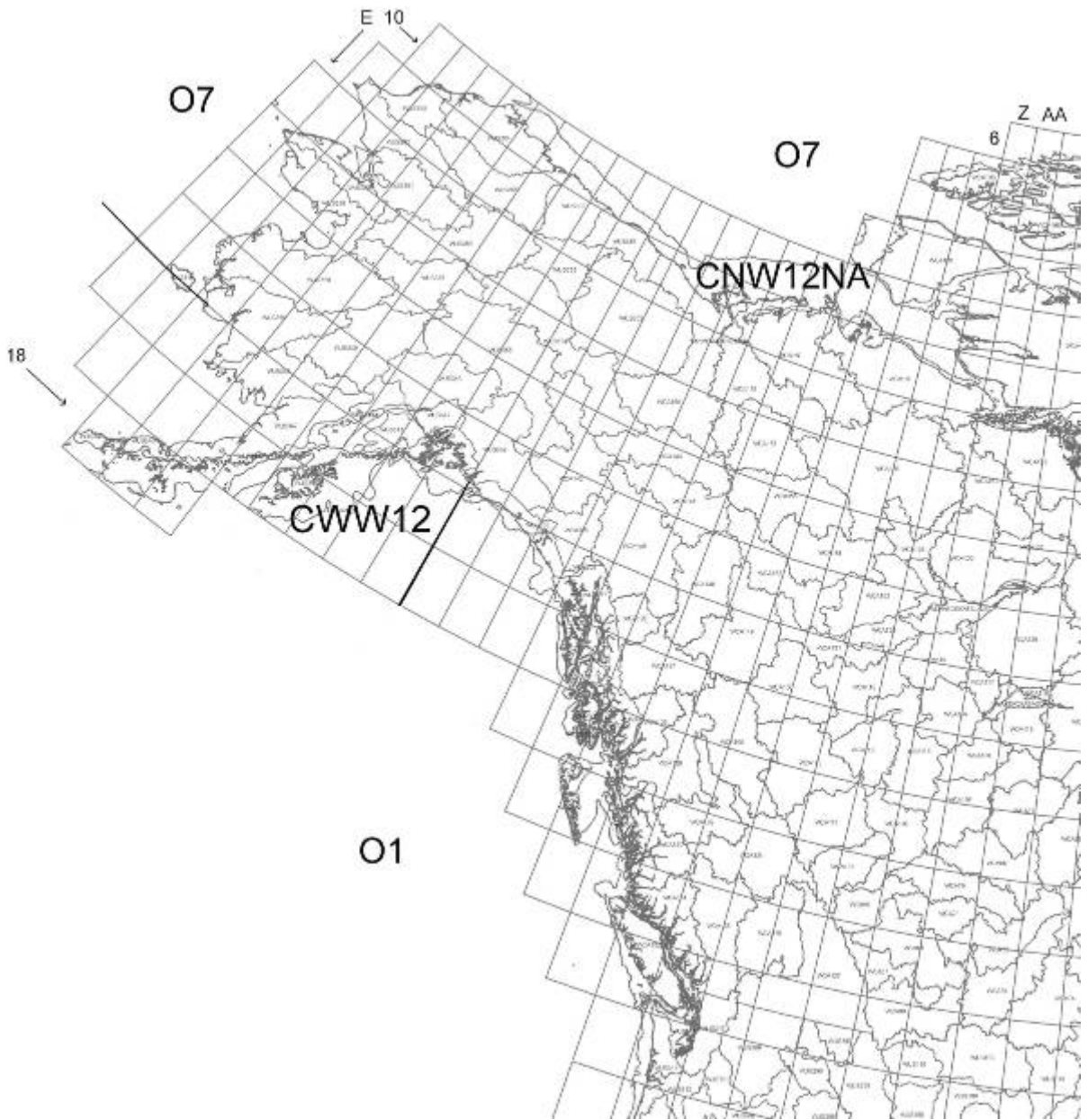


Figure 2-8: IMPACT North America: Watershed zones (version 1.0). North West side of North America.

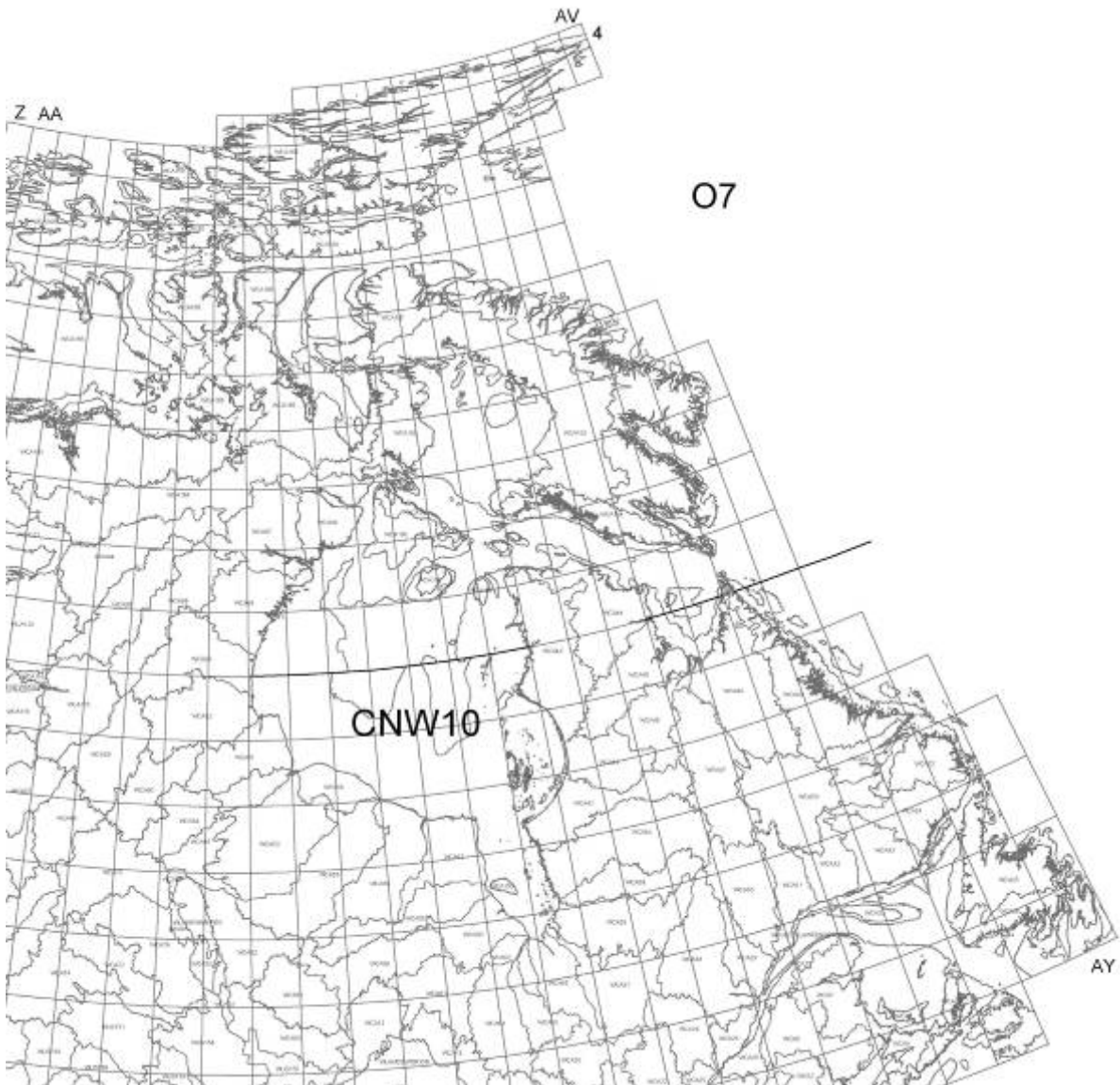


Figure 2-9: IMPACT North America: Watershed zones (version 1.0). North East side of North America.

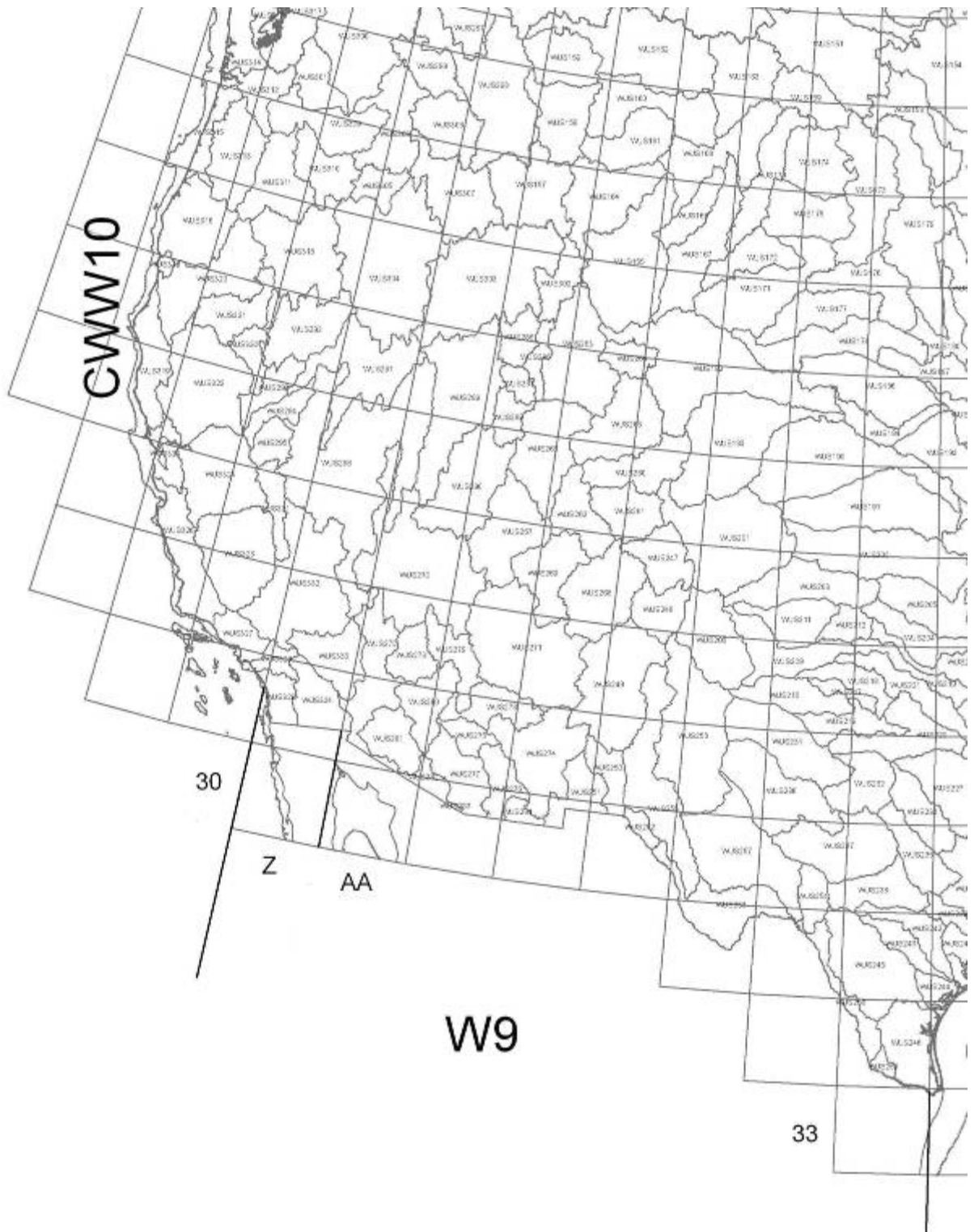


Figure 2-10: IMPACT North America: Watershed zones (version 1.0). South West side of North America.

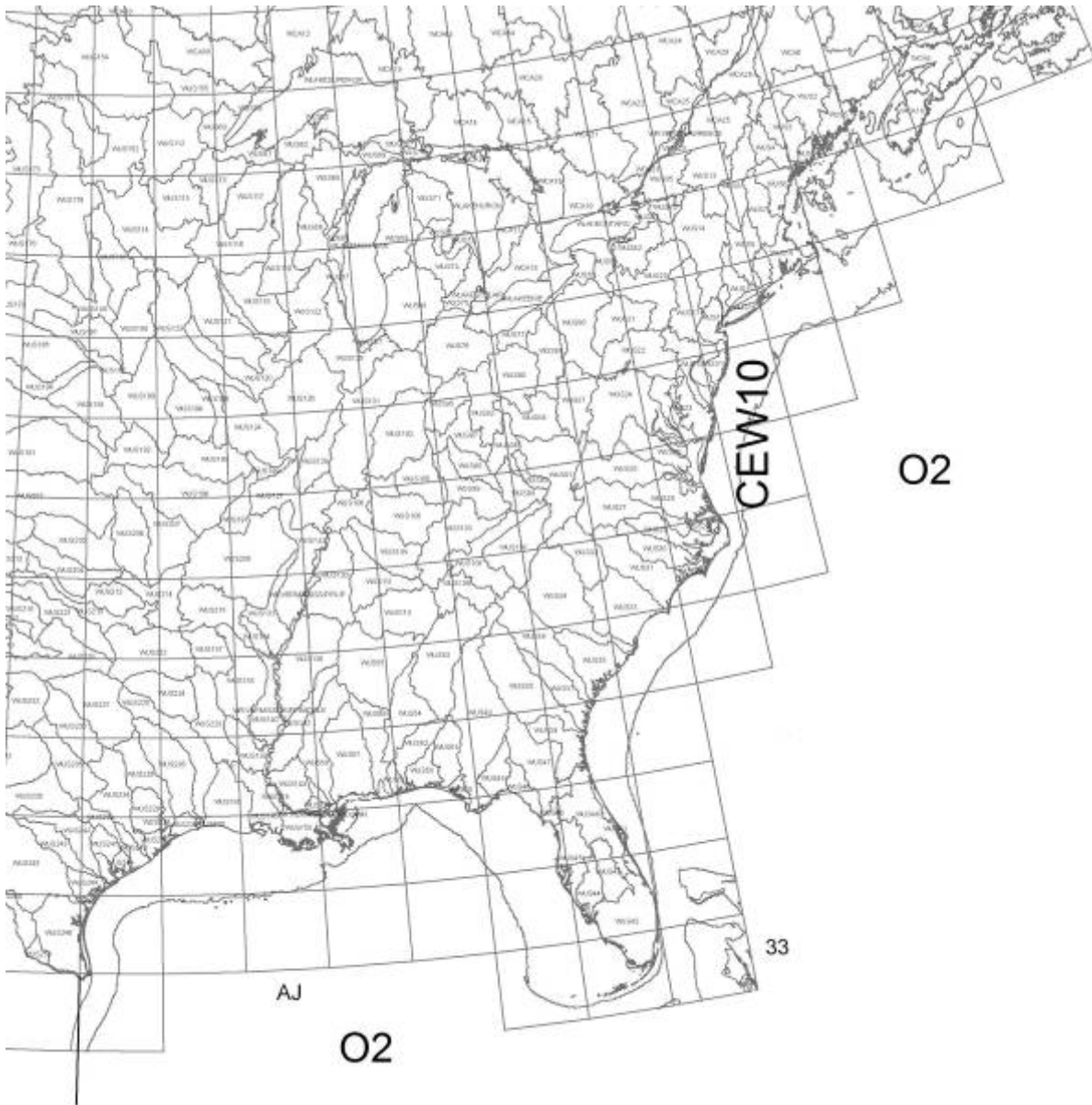


Figure 2-11: IMPACT North America: Watershed zones (version 1.0). Watershed zones (version 1.0). South East side of North America.

2.8.7. Comparison with monitored data

Monitored emissions and concentrations of benzo(a)pyrene (BaP), 2,3,7,8-TCDD, and mercury (Hg) are used to evaluate the model. In the case of benzo(a)pyrene and 2,3,7,8-TCDD, emissions have significantly decreased in the last 10 years because of improved emissions controls, therefore monitored data are separated between those before 2000 and those after. A summary of the physico-chemical properties of these substances is provided in Table 2-5.

Table 2-5: Physico-chemical properties of the substances ^a used to evaluate the model.

Sub-stance	CAS#	Henry's Constant (Pa·m ³ /mol)	Log Kow	T _{1/2} (air) (h)	T _{1/2} (soil) (h)	T _{1/2} (water) (h)	T _{1/2} (sediment) (h)	T _{1/2} (vegetation) (h)	BCF (bioconcentration factor) (kg _{water} /kg _{fish})	
Benzo(a)pyrene	50-32-8	0.045	6.0	170	17,000	1,700	62,000	17,000	10,000	
2,3,7,8-TCDD	1746-01-6	3.3	6.8	200	900,000	4,000	900,000	17,000	34,000	
Sub-stance	CAS#	Suspended solids - water part coeff (L _{water} /kg _{solids})	Sediment solids - water part coeff (L _{water} /kg _{solids})	Soil solids - water part coeff (L _{water} /kg _{solids})	Particle - gas part coeff (dimensionless)	BCF (bioconcentration factor) (kg _{water} /kg _{fish})	B_milk (bio-transfer factor in milk) [d/L]	B_meat (bio-transfer factor in meat) [d/kg]	B_eggs (bio-transfer factor in eggs) [d/kg]	Bulk plant-soil concentration ratio [m ³ _{soil} /kg _{plant}]
Mercury	7439-97-6	170,000	110,000	170	2.5E+9	3,000	4.6E-4	1.6E-3	4.6E-4	1.2E-6

^a Values are taken directly from Pennington et al. (2005) and used in Jolliet et al. (2003) (unpublished supporting information for both publications). The complete list of properties and sources can be found in the "Chem data" sheet of the model.

Monitored data

I converted each monitored data in the same unit as the modeled concentrations to perform the analysis presented in Figure 2-4. To convert mass un volume, for sediment, I used a density of 2,400 kg/m³, for surface sediment, I used a density of 2,000 kg/m³, and for soil, I used a density of 1,500 kg/m³.

Benzo(a)pyrene

Table 2-6 presents a summary of the amount and distribution of emissions of benzo(a)pyrene, along with reported concentrations and their location. Annual anthropogenic emissions of benzo(a)pyrene in North America are ~1000 tonnes for the 1990s and ~100 tonnes after 2000. Annual natural benzo(a)pyrene emissions in North America (from forest fires and agricultural burning) are ~100 tonnes². Worldwide (outside of North America) annual anthropogenic benzo(a)pyrene emissions are estimated to be ~3600 tonnes (Korte 1999). Anthropogenic emissions are assumed to be distributed proportionally to population in the model. Natural emissions are assumed to be distributed proportionally to the burnable biomass (outside of backyard burning) (using the burnable area as a proxy in the model).

² Benzo(a)pyrene emissions from forest and agricultural fires throughout the United States was originally estimated to be 127 t/y (NAS 1972), and later revised to 9.5 t/y (Sullivan and Mix 1983). However Baek et al. (1991) report values of 400 (Ramdahl et al. 1983) to 1190 (Peters et al. 1981) t/y for agriculture fires and 600 (Ramdahl et al. 1983) to 1478 (Peters et al. 1981) t/y for forest fires. An average value of 100 t/y is taken and distributed proportionally to burnable area in North America.

Table 2-6: Monitored concentrations of benzo(a)pyrene and their respective location (all monitored concentration are for emissions occurring before 2000).

Monitored concentration (with exact value reported and original unit given in the reference) (ppt means 10^{-12} kg/kg _{water})	Location of the monitored concentration	Source for the monitored concentration	Zone/Cell in IMPACT North America	Modeled concentration (kg/m ³)
1,500 µg/kg (mean concentrations from 1990 to 2000)	Sediment of lakes situated in "dense urban" watershed.	van Metre and Mahler (2005)	In Van Metre and Mahler (2005), land use in the watersheds was categorized as "dense urban" (>52% urban land use; 14 lakes), "light urban" (5-42% urban; 17 lakes), or "reference" (<1.5% urban; 7 lakes). Assumption in the model: urban land use = 25% paved/75% unpaved. Dense urban: >50% paved area; Light urban: 1-50% paved area; Reference: <1% paved area	4E-5
120 µg/kg (mean concentrations from 1990 to 2000)	Sediment of lakes situated in "light urban" watershed.			4E-5
20 µg/kg (mean concentrations from 1990 to 2000)	Sediment of lakes situated in "reference" watershed.			5E-6
8 pg/m ³	Air, in Alert, Canada	Macdonald et al. (2000)	AV4	3E-15
3 pg/m ³	Air, in Tagish, Canada	Macdonald et al. (2000)	S15	8E-15
0.11-0.18 ng/m ³	Air, Chula Vista/South California	ARB 1994	Z29	6E-13
1.4-1.5 ng/m ³	Air, Sacramento/Fresno California	ARB 1994	X27, Y28	4-5E-13
5.1-430 ng/g	Sediment, Lake Erie	GLC 2007	WLAKEERIE	3E-6
5-2,200 ng/g	Sediment, Michigan inland lakes	GLC 2007	WUS68, WUS75, WUS73, WUS72, WUS69, WUS71, WUS63, WUS65	1E-5, 2E-5, 2E-5, 1E-5, 5E-6, 4E-6, 2E-6, 5E-6
1,500-1,700 ng/g	Sediment, urbanized Minnesota inland lakes	GLC 2007	WUS116	5E-6
150-460 ng/g	Sediment, rural IL inland lakes	GLC 2007	WUS126, WIS125, WUS122, WUS101	1E-5, 1E-5, 9E-6, 3E-5
480-1,900 ng/g	Sediment, Foy River, WI	GLC 2007	WUS117, WUS119, WUS66	5E-6, 6E-6, 6E-6
11-43 ng/g	Sediment, Lac St Louis, QC	GLC 2007	WCA23, WCA25	4E-6, 5E-6
1,100 ng/g	Sediment, Ashtabula River, OH	GLC 2007	WUS77	7E-4
130-270 ng/g	Sediment, Lake Michigan	GLC 2007	WLAKEMICHIGAN	5E-7
9-22,000 ng/g	Sediment, St Lawrence River, international section	GLC 2007	WRIVERSTLAWRENCE, WUS85	2E-6, 4E-5
80-610 ng/g	Sediment, Sheboygan River, WI	GLC 2007	WUS65	5E-6
0.48 ng/l	Water, Lac St Louis, QC	GLC 2007	WCA23, WCA25	4E-9, 4E-9
0.4 ng/l	Water, Southern Lake Michigan	GLC 2007	WLAKEMICHIGAN	4E-10
2-6,500 ng/g	Soil	GLC 2007 / Wolfgang 2000	AVERAGE	3E-7
0.03-0.7 ppt	Water, Great Lakes, Canada	Wang et al. (1997)	WLAKESUPERIOR, WLAKEMICHIGAN, WLAKEHURON, WLAKESTCLAIR, WLAKEERIE, WLAKEONTARIO	8E-11, 4E-10, 4E-10, 4E-10, 3E-9, 4E-10
5.6 ng/g	Soil, Long Island	Shantakumar et al. (2005)	WUS16	4E-5
40 µg/kg	Soil, Massachusetts	Nelson (1983)	WUS7	2E-5
240 µg/kg	Soil, Connecticut	Nelson (1983)	WUS11	3E-5
87 µg/kg	Soil, Canada near Toronto	Nelson (1983)	WCA19	4E-5

2,3,7,8-TCDD

Table 2-7 presents a summary of the amount and distribution of emissions of 2,3,7,8-TCDD, along with reported concentrations and their location. Annual anthropogenic emissions

of 2,3,7,8-TCDD in North America are estimated, on average, to be ~10 kg for the period 1970-2000 and ~1 kg after 2000 (USEPA 2006). Annual natural emissions (from forest fires and agriculture burning) of 2,3,7,8-TCDD in North America are assumed to be ~0.1 kg. Anthropogenic emissions are assumed to be distributed proportionally to population in the model. Natural emissions are assumed to be distributed proportionally to the burnable biomass (outside of backyard burning) (using the burnable area as a proxy in the model). Sources related to dioxin include <http://www.umdioxin.org> and <http://cfpub.epa.gov/ncea/cfm/recorddisplay.cfm?deid=55264>.

Table 2-7: Monitored concentrations of 2,3,7,8-TCDD and their respective location (all monitored concentration are for emissions occurring before 2000).

Monitored concentration (with exact value reported and original unit given in the reference) (ppt means 10^{-12} kg/kg _{water})	Location of the monitored concentration	Source for the monitored concentration	Zone/Cell in IMPACT North America	Concentration modeled (kg/kg for fish, eggs and meat, kg/m ³ for sediment, water and soil)
~0.8 pg/g (0-1,500 pg/g)	Marine (coastal) sediment, in Maine	Wade et al. (1997)	CEW10	4E-11
23.1 pg/g (1985) and 5.3 pg/g (1991)	Sediment, Androscoggin river, Maine	Wade et al. (1997)	WUS6	3E-9
0.84 µg/kg	Sediment, Passaic River NJ	Scott et al. (2000)	WUS15	5E-9
1 pg/g	Fish, Pond Inlet (Canadian Arctic)	Sanderson et al. (1997)	CNW12NA	9E-14
2 pg/g	Fish, Somerset Island (Canadian Arctic)	Sanderson et al. (1997)	CNW12NA	9E-14
1 pg/g	Fish, Spence Bay, (Canadian Arctic)	Sanderson et al. (1997)	CNW12NA	9E-14
0.1-2 pg/g	Fish, Lake Laberge	Sanderson et al. (1997)	WCA140	1E-14
0.1-0.2 pg/g	Fish, Great Slave Lake	Sanderson et al. (1997)	WLAKEGREATSLAVE	1E-13
0.06-16 pg/g	Fish, Slave River	Sanderson et al. (1997)	WCA120	5E-14
2 pg/g	Fish, Kusawa Lake	Sanderson et al. (1997)	WCA86	2E-13
1 pg/g	Fish, central Arctic archipelago	Braune et al. (1999)	CNW12NA	9E-14
0.1 pg/g	Caribou in Yukon and NWT	Braune et al. (1999)	AVERAGE	2E-16
0.3-1.4 ng/kg	Fish, Saguenay Fjord, QC	Brochu et al. (1995)	WCA28, WCA25	7E-13, 2E-12
0.2-1.2 ng/kg	Fish, St Lawrence Estuary, QC	Brochu et al. (1995)	WCA25	2E-12
8.8-24 ppt	Fish, Saginaw Bay MI	Fehring et al. (1985a, 1985b)	WLAKEHURON	2E-13
11 ppt	Fish, Muskegon Lake MI	Fehring et al. (1985a, 1985b)	WUS68	2E-12
~0.01 ng/kg	Rural soil, WH	Rogowski and Yake (2005)	WUS300, WUS301	9E-12, 1E-11
~1 ng/kg	Urban soil, WH	Rogowski and Yake (2005)	WUS317, WUS312	n/a
410 ppt	Fish, Newark Bay NJ urban	USEPA 2004	WUS15	8E-12
1 ppt	Fish, Lake Superior	USEPA 2004	WLAKESUPERIOR	5E-14
8.6 ppt	Fish, Lake Huron	USEPA 2004	WLAKEHURON	2E-13
4.4 ppt	Fish, Lake Michigan	USEPA 2004	WLAKEMICHIGAN	2E-13
1.8 ppt	Fish, Lake Erie	USEPA 2004	WLAKEERIE	9E-13
6.6 ppt	Fish, Lake St Clair	USEPA 2004	WLAKESTCLAIR	2E-13
15 ppt	Fish, Lake Ontario	USEPA 2004	WLAKEONTARIO	2E-13
1 ppt	Soil, Henry IL, residential	USEPA 2004	WUS125	3E-11
1 ppt	Soil, Middletown OH, residential	USEPA 2004	WUS97	3E-11
0.39 ppt	Soil, Ohio, background	USEPA 2004	WUS92	3E-11
2.3 ppt	Soil, Ohio, urban	USEPA 2004	WUS92	3E-11
0.61 ppt	Soil, Connecticut, urban	USEPA 2004	WUS11	6E-11
0.28 ppt	Soil, Yarmouth, Maine, background	USEPA 2004	WUS6	7E-11

Monitored concentration (with exact value reported and original unit given in the reference) (ppt means 10 ⁻¹² kg/kg _{water})	Location of the monitored concentration	Source for the monitored concentration	Zone/Cell in IMPACT North America	Concentration modeled (kg/kg for fish, eggs and meat, kg/m ³ for sediment, water and soil)
0.61 ppt	Soil, Denver Front Range, various land use	USEPA 2004	WUS183	2E-11
2.7 ppt	Water, Ontario, Canada	USEPA 2004	WCA61	1E-14
1.7 ppt	Water, Lockport, New York	USEPA 2004	WUS15	2E-13
110 ppt	Fish, Passaic River, New Jersey, urban	USEPA 2004	WUS15	8E-12
0.26 ppt	Beef, Los Angeles	USEPA 2004	WUS327	2E-14
0.28 ppt	Beef, San Francisco	USEPA 2004	WUS325	2E-14
0.017 ppt	Beef, New York	USEPA 2004	WUS15	3E-13
0.04 ppt	Ground Beef, South Mississippi	USEPA 2004	WUS142	1E-14
0.30 ppt	Pork, Los Angeles	USEPA 2004	WUS327	2E-14
0.44 ppt	Pork, San Francisco	USEPA 2004	WUS325	2E-14
0.013 ppt	Pork, New York	USEPA 2004	WUS15	3E-13
0.23 ppt	Chicken, Los Angeles	USEPA 2004	WUS327	2E-14
0.70 ppt	Chicken, San Francisco	USEPA 2004	WUS325	2E-14
0.011 ppt	Chicken, New York	USEPA 2004	WUS15	3E-13
0.16 ppt	Chicken, South Mississippi	USEPA 2004	WUS142	1E-14
0.02 ppt	Eggs, Los Angeles	USEPA 2004	WUS327	1E-15
0.02 ppt	Eggs, San Francisco	USEPA 2004	WUS325	2E-15
0.04 ppt	Eggs, South Mississippi	USEPA 2004	WUS142	3E-15

Mercury

Table 2-8 presents a summary of the amount and distribution of emissions of mercury, along with reported concentrations and their location. Annual anthropogenic emissions of mercury in North America are approximately 100 t/y.³ Annual natural mercury emissions in North America (from forest fires and agriculture burning) are ~31 t/y for the 48 lower states and 12 t/y for Alaska (Wiedinmyer and Friedli 2007). No data are available for natural emissions for Canada. They are estimated to be 30 t/y, one third from anthropogenic sources and two thirds from forest fires and agricultural burning. An estimated 2,000 t/y of mercury are emitted worldwide in the atmosphere by coal-burning power plants. Anthropogenic emissions are assumed to be distributed proportionally to population in the model. Natural emissions are assumed to be distributed proportionally to burnable biomass (other than backyard burning) (using the burnable area as a proxy in the model).

Table 2-8: Monitored concentrations of mercury and their respective location.

Monitored concentration (with exact value reported and original unit given in the reference)	Location of the monitored concentration	Source for the monitored concentration	Zone/Cell in IMPACT North America	Concentration modeled (kg/kg for fish, kg/m ³ for water and sediment)
3.7 ng/l	Arctic Ocean water	Schmidt and Freimann (1984)	CNW12NA	1E-11
1-15 ng/l	Water, Beaufort Sea	Thomas (1983)	CNW12NA	1E-11
60 ng/g, but in constant increase	Sediment, Arctic Ocean	Gobeil et al. (1999)	CNW12NA	4E-7
0.044 (0.017-0.15) µg/l	Suspended sediments in Slave River at Fort Smith (considered "in bulk water")	McCarthy et al. (1997)	WLAKEGREATSLAVE	3E-9

³ Source: Environment News Service (2009) and National Emissions Inventory (NEI). Available at <http://www.epa.gov/ttn/chief/einformation.html>.

Monitored concentration (with exact value reported and original unit given in the reference)	Location of the monitored concentration	Source for the monitored concentration	Zone/Cell in IMPACT North America	Concentration modeled (kg/kg for fish, kg/m ³ for water and sediment)
30-100 ng/g	Surface sediment, average NWT lakes	Macdonald et al. (2000)	AVERAGE	2E-4
23-88 ng/g	Surface sediment, average Yukon lakes	Macdonald et al. (2000)	AVERAGE	2E-4
0.45 (year 1975) - 0.15 (year 2000) mg/kg	Sediment, Tennessee rivers and lakes	TVA (2002), http://www.tva.gov/environment/air/ontheair/merc_emis.htm	AVERAGE	3E-8
0.031-0.15 mg/kg	Sediment, North Mississippi Lakes	Huggett et al. (2001)	WUS136	8E-4
0.035 mg/kg	Sediment, Carson City NV	Davis et al. (1997)	WUS293	3E-4
mean ~12 (0-797) µg/100g	Seal/fish		AVERAGE	7E-9
~ 0.3 (0.02-1.5) mg/kg	Freshwater fish in Alaska	Jewett and Duffy (2007)	WUS365	7E-9
~ 0.05 (0.02-0.36) mg/kg	Marine water fish in Alaska	Jewett and Duffy (2007)	CNW12NA	8E-11
0.63-1.9 mg/kg	Fish, Enid Lake, North Mississippi Lakes	Huggett et al. (2001)	WUS136	4E-8

2.8.8. Comparison with other models

Table 2-9 provides a summary of the physico-chemical properties of the substances used to evaluate the differences among models. Some properties differ from those presented in Table 2-5 (properties that changed are italicized in Table 2-9) to match those of BETR (MacLeod et al. 2004).

Table 2-9: Physico-chemical properties of the substances used to evaluate the model against BETR.

Substance	CAS#	Henry's Constant (Pa·m ³ /mol)	Log Kow	T _{1/2} (air) (h)	T _{1/2} (soil) (h)	T _{1/2} (water) (h)	T _{1/2} (sediment) (h)	T _{1/2} (vegetation) (h)	BCF (kg _{water} /kg _{fish})
Benzo(a)pyrene	50-32-8	0.046	6.3	<i>1.5</i>	<i>5,500</i>	56	28,000	1.5	10,000
2,3,7,8-TCDD	1746-01-6	2.5	6.7	720	<i>160,000</i>	<i>10,000</i>	<i>49,000</i>	<i>52,000</i>	34,000
Benzene	71-43-2	550	2.2	140	4,600	270	540	4,600	8.7
Carbon tetra chloride	56-23-5	3,300	2.7	87,000	4,700	6,500	4,500	87,000	n/a

The complete list of properties and sources can be found in the "Chem data" sheet of the model.

2.8.9. References used in the supporting information of this chapter

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3. Evaluating the meaningful level of resolution for regional intake fractions

Performing a complete regionalized life-cycle impact assessment study based on geographically differentiated information remains essentially impossible with current life-cycle assessment software because of the significant amount of data to be treated. This problem is addressed in this chapter. Therefore, one can ask whether regionalization in life-cycle assessment makes sense, and if so, how to perform it. This chapter interprets the previous chapter to consider in more detail the issue of regionalization in fate and exposure of pollutants emitted in North America. This evaluation is based on the information that the IMPACT North America model is able to provide at the level of intake fractions.

3.1. Summary

Results of life-cycle assessments can be significantly improved by performing archetype-based regionalization. The regionalization of inventory and impact assessment is recognized as an important step towards improving accuracy, precision and confidence in life-cycle assessment results, as well as its discriminatory power. Two approaches can be used to perform regionalization in life-cycle assessment: the geographically differentiated approach and the archetype approach. The geographic approach uses information on where the process is situated in the world (e.g., downtown Chicago), and considers local conditions to estimate the impacts of direct emissions. For the archetype approach, however, the exact location of emissions is not needed, as this approach uses the “representative” characteristics of the emission location to evaluate the subsequent impacts. The geographic approach is based on the actual location of emission whereas the archetype approach is based on a representative set of characteristics of the location of emission. I have analyzed the variation in intake fractions of a selected set of pollutants in North America and found that geographic intake fractions can vary by eight orders of magnitude depending on the location of emission. Inhalation and ingestion are generally correlated to the characteristics of population density and agricultural intensity, respectively. I found that when generic intake fraction is evaluated as the emission-weighted average intake fraction, emissions are better correlated with population than with land area or agricultural production intensity. I compiled a list of suggested archetypes. The archetypes approach can provide the same accuracy as the geographic approach with significantly less information. Since there is currently no tool for gathering large sets of detailed geographic data on inventory and impact assessment in an efficient way, I suggest using the archetype approach as a realistic implementation of regionalization in life-cycle assessment. Using the archetype approach will significantly improve the confidence in results while making the regionalization process practical. I also suggest expanding this work to other regions of the world, other media of emissions such as water or soil, as well as other impact categories such as ecotoxicity, land use, or water use.

3.2. Introduction

Regionalization in life-cycle impact assessment

In this chapter I address the issue of regionalization of intake fractions in life-cycle impact assessment of damage to human health. Regionalization is recognized as an important step towards improving accuracy, precision and confidence in life-cycle assessment results, as

well as its discriminatory power (i.e., its capacity to discriminate with confidence between two alternatives), especially for comparative assessments (Potting and Hauschild 2006, Sedlbauer et al. 2007, Reap et al. 2008, Margni et al. 2008). Life-cycle assessment evaluates the environmental consequences of a product or service by modeling its entire value chain from resource extraction to the end of the product's life (ISO 2006a, 2006b). Life-cycle assessment addresses the environmental consequences over a set of impact categories, by combining a life-cycle inventory with a life-cycle impact assessment method. These consequences can be global, regional or local. Global warming or ozone layer depletion are global impact categories because their consequences are independent of the emission location. Other outcomes such as toxicological and eco-toxicological impact to both humans and ecosystems or acidification, often occur as regional or local impacts (Potting and Hauschild 2006, Sedlbauer et al. 2007, Reap et al. 2008, Margni et al. 2008, Manneh et al. 2009), making it important to evaluate them in relation to where the emission takes place.

Definition of generic, regionalization, geographic and archetype

Generic information does not account for where the emission occurs, and therefore represents “world average” conditions.

When addressing regionalization, Potting and Hauschild (2006) propose three distinctions – site-generic (no spatial differentiation), site-dependent (some spatial differentiation), or site-specific (a very detailed spatial differentiation). Sedlbauer et al. (2007) differentiate between the approaches of geographically differentiated solutions and situation-dependent solutions. Geographically differentiated refers to the differences among geographic locations such as continents, countries, or regions throughout the world. Situation-dependent refers to archetypical situations leading to important variations in the characterization modeling and its results, and therefore justifying a differentiation. Based on the same approach, Margni et al. (2008) differentiated among archetype differentiation, geographic differentiation, and combined archetype-geographic approaches.

In this dissertation, regionalization is used in the sense of evaluating non-global impacts by considering local specificities. Following the above suggestions, the two main approaches identified to perform regionalization in life-cycle assessment are the geographic approach and the archetype approach. The geographic approach (the geographically differentiated or site-specific solutions) uses information on where the process is situated in the world (e.g., downtown Chicago), and considers local conditions to estimate the impacts of direct emissions. The precision of the estimated impacts increases as the scale of the regionalization decreases (e.g., continental scale models are less precise than urban scale models), but data needs also increase with decreasing scale (increased spatial resolution). For the archetype approach (i.e., the situation-dependent or site-dependent solutions), the exact location of emissions is not needed, since this approach uses information on the main characteristics of the emission location (e.g., population density, agricultural intensity) to evaluate the subsequent impacts. Each archetype can, however, contain a certain amount of geographic information (e.g., developed versus developing country). The data needs increase with an increasing number of archetypes (e.g., how many different levels of population density should be included).

Furthermore, in this chapter I suggest using the following definitions based on the distance from the location of emission: “local” means within a few ten kilometers (e.g., the urban area), “regional” means within a few hundred kilometers (e.g., California, the Central Valley), “continental” means within the continent (e.g., North America), and “global” means worldwide.

Summary of existing archetypes

This section reviews the different archetypes that have been explored in the life-cycle assessment literature.

Sedlbauer et al. (2007) suggest the following archetypes for human toxicity. For air emission: high versus average versus low population density (for pollutants dominated by the inhalation pathway); intensive versus extensive versus nonagricultural region (for pollutants dominated by the food pathway); and off-shore. For water emission: upstream versus downstream of a lake; ocean versus lake versus river. For soil emission: agricultural versus non-agricultural soil. For respiratory effects caused by inorganic air emissions, only high versus average versus low population density, and off-shore counts. Margni et al. (2008) suggest the following list of archetypes, independent of the media of emission: high population density (urban), medium population density, low population density (rural), and indoor emissions, as well as the height of emission (tailpipe, stack, or plane). Note that Margni et al. (2008) used the term “low” population density for “rural,” whereas in the present chapter “medium” population density is used for “rural” and “low” population density is used for remote areas such as Alaska or the ocean. Ecoinvent (Frischknecht 2005) uses the following archetypes: for air emissions: high population density, low population density, low population density long-term, lower stratosphere and upper troposphere, and unspecified; for water emissions: fossil water, groundwater, groundwater long-term, lake, ocean, river, river long-term, and unspecified; for soil emissions: agricultural, forestry, industrial, and unspecified.

Studies have also examined the different characteristics that can significantly influence intake fractions (and therefore should be divided into archetypes). For air emissions, the population density and the agricultural intensity are found to be significant parameters influencing the intake fraction (MacLeod et al. 2004, and see Chapter 2). Intake of primary particulate matter is mainly influenced by local population density (Levy et al. 2002, Heath et al. 2006, Greco et al. 2007, and see Chapter 2), whereas intake of secondary particulate matter is mainly influenced by regional population density (and see Chapter 4). More generally, for short-range to medium-range pollutants that have inhalation as their main intake pathway, there is a need to consider whether or not they are emitted in an urban area (Rosenbaum et al. 2008, and see Chapter 2). For air emissions, Goedkoop et al. (2008) distinguish between urban (default being the urban compartment) and non-urban emissions for human toxicity, but do not make any distinctions in the location of particulate matter emissions. For water emissions, they distinguish between freshwater (default being the freshwater compartment) and ocean. Pennington et al. (2005) find that for water emissions, whether the emission occurs upstream or downstream of a lake is a significant parameter. For soil emissions, Goedkoop and Spriensma (2000) distinguish between agricultural and industrial land, whereas Goedkoop et al. (2008) have added a distinction between forestry soil and industrial soil (the latter being considered the same as urban soil). Rochat et al. (2006) showed, on a continental basis, that the oral intake fraction from air, water and soil emissions is correlated with the food production intensity (i.e., the agricultural yield, in, e.g., tonnes per ha per year). For indoor emissions, there is clearly a need to model the intake using an indoor box (Nazaroff 2008, Margni et al. 2008, Hellweg et al. 2009).

Objectives

In this chapter, I aim to explore whether regionalization based on the archetype approach can improve the ability of life-cycle assessment to better address human health damage from alternatives scenarios, while reducing the significant data requirements needed by the geographically differentiated approach. This question is addressed through the following steps:

(i) review the different archetypes that have been suggested in the literature, (ii) evaluate how population density and agricultural production intensity, two major archetypes, influence the regional intake fractions for air emissions, (iii) generate a suggested set of archetypes, (iv) evaluate the different ways to calculate generic intake fractions, (v) compare the magnitude and the variability of geographically differentiated intake fractions versus generic intake fractions for air emissions, and (vi) compare and discuss the advantages and disadvantage of the “archetype” and “geographic” approaches when performing regionalization of intake fractions in life-cycle assessment.

3.3. Method

Modeling framework

Assessing the toxicological effects on human health of a chemical emitted into the environment requires a cause-and-effect chain assessment linking the emission source to the damage through four intermediary parameters, as mathematically expressed by Equation 2-1 in Chapter 2 and depicted in Figure 2-1.

Within this overall framework, we assumed that fate, exposure, dose-response, and severity are not functions of time, and that dose-response and severity are not functions of space. These common assumptions in life-cycle impact assessment reflect the limited availability of temporally and spatially dependent data (Pennington et al. 2006, Rosenbaum et al. 2007). In the present chapter, regionalization is therefore analyzed using the intake fraction, because the characterization factor is proportional to the intake fraction.

Calculating generic versus regional intake fractions

Multimedia and multipathways models are recognized as a well-suited modeling approach for assessing the fate and exposure in life-cycle impact assessment (Hertwich 2002). To perform the present analysis, one needs to use a model that can provide both generic and geographic regional characterization factors. Several models are available to calculate geographic fate and exposure for hundreds of pollutants, such as BETR North America (MacLeod et al. 2001), BETR World (Toose et al. 2004), IMPACT 2002 Western Europe (Pennington et al. 2005), IMPACT 2002 Continental (Rochat et al. 2006, GLOBOX (Sleeswijk 2006), and IMPACT North America (Chapter 2). The IMPACT North America model introduced in Chapter 2 (see Figure 2-3) is chosen here because it allows one to calculate both intake fractions and characterization factors at a high geographic resolution and includes urban boxes. In addition to the air cells presented in Figure 2-3, 292 urban areas are nested in the respective air cells, and these urban boxes allow one to better capture the influence of urban emissions. Indeed, Rosenbaum et al. (2008) and Chapter 2 have shown that considering urban compartments in impact modeling is important for low- to medium-persistent pollutants with inhalation as a dominant intake pathway. Finally, generic intake fractions are defined and calculated as the emissions-weighted average intake fractions for the emissions in the different regions of North America. Since the spatial distribution of most emissions is unknown, proxies for evaluating distribution of emissions are identified. Differences among the different proxies are evaluated.

Test set

Physical-chemical properties for the pollutants considered in the test set are provided in Table 3-1.

The pollutants used as test set are presented in Table 3-1. This test set was designed by the OMNITOX team and for the OMNITOX project (Molander et al. 2004), which aimed at

evaluating different multimedia models. This test set results from an international consensus and aims at representing pollutants with a wide range of properties. I added benzene, benzo(a)pyrene, dioxin, mercury, and primary PM_{2.5} to the original test set. The rationale for the added pollutants is that they represent pollutants that are often found to be dominant in life-cycle assessment results (for PM_{2.5}, benzene, benzo(a)pyrene, and dioxin – see Chapter 5) or representative of an inorganic emission for which the model IMPACT North America was tested (for mercury – see Chapter 2).

Table 3-1: Test set used in the present chapter.

CAS#	Name	Log Kow (octanol-water partitioning coefficient)	Tropospheric degradation half life (h) ^a	Water degradation half life (h) ^a	Vegetation degradation half life (h) ^a	Soil degradation half life (h) ^a	Source
127-18-4	Tetrachloroethylene	2.6	550	550	550	1,700	Based on the unpublished supporting information for Molander et al. (2004) and Rosenbaum et al. (2008) (Ralph Rosenbaum, personal communication, fall 2009)
56-23-5	Carbon tetrachloride	2.6	17,000	1,700	17,000	5,500	
106-99-0	1,3-Butadiene	2.0	5	170	5	550	
16752-77-5	Methomyl	0.60	550	5,500	550	550	
30560-19-1	Acephate	-1.0	8	1,300	8	53	
50-00-0	Formaldehyde	0.35	5	55	5	55	
1336-36-3	PCBs _s	6.3	385	900	390	900	
117-84-0	Di(n-octyl) phthalate	8.1	27	340	27	340	
87-82-1	Benzene, hexabromo-	6.1	22,000	1,440	22,000	1,400	
52315-07-8	Cypermethrin	6.6	10	120	10	1,200	
2385-85-5	Mirex	5.3	170	170	170	55,000	
1582-09-8	Trifluralin	5.3	170	1,700	170	1,700	
115-32-2	Dicofol	5.0	70	900	70	1,500	
106-46-7	p-Dichlorobenzene	3.5	550	1,700	550	5,500	
309-00-2	Aldrin	3.0	5	17,000	5	17,000	
79-34-5	1,1,2,2-Tetrachloroethane	2.4	17,000	1,700	17,000	5,500	
133-06-2	Captan	2.3	17	17	17	550	
23950-58-5	Pronamide	3.5	1,400	980	1,400	1,900	
120-12-7	Anthracene	4.5	55	550	55	5,500	
58-89-9	gamma-Hexachlorocyclohexane	3.7	170	17,000	170	17,000	
131-11-3	Dimethyl phthalate	1.7	170	170	170	550	
67-56-1	Methanol	-0.8	170	55	170	55	
107-06-2	1,2-Dichloroethane	1.4	1,700	1,700	1,700	5,500	
141-78-6	Ethyl acetate	0.69	55	55	55	170	
55-18-5	N-Nitrosodiethylamine	0.5	5	17	5	1,700	
137-26-8	Thioperoxydicarbonic diamide, tetramethyl-	1.7	170	170	170	550	
114-26-1	Propoxur	1.5	5	550	5	550	
133-07-3	1H-Isoindole-1,3(2H)-dione, 2-(trichloromethyl)thio -	3.6	27	14,000	27	14,000	
50-32-8	Benzo(a)pyrene	6.0	170	1,700	17,000	17,000	Chapter 2
1746-01-6	2,3,7,8-TCDD	6.9	170	550	170	17,000	Chapter 2
	Inorganics	Suspended solids - water part coefficient (L/kg)	Sediment solids - water part coefficient (L/kg)	Soil solids - water part coefficient (L/g)	Particle - gas part coefficient (-)	BCF (bioconcentration factor) (L/kg fish)	
	Primary particulate matter (PM)	5.E+04	3.E+04	2.E+02	1.E+15		Chapter 2

^a The same values (or a value ten times greater or smaller) appear for different media, especially for half-lives constants. This comes from the fact that often physico-chemical parameters are missing and are extrapolated from values for other media using simple relations such as “ten times longer” or “ten times shorter”.

3.4. Results and discussion

Regional factors influencing intake fractions

Here, I evaluate how population density and agricultural production intensity, two major archetypes, influence the regional intake fractions for air emissions. I modeled the intake fraction for an emission of particulate matter smaller than 2.5 μm ($\text{PM}_{2.5}$) and benzo(a)pyrene in each of 1,121 zones in the IMPACT North America model using the physico-chemical parameters described in Table 3-1. I therefore obtain 2,242 intake fraction values. These values are plotted in Figure 3-1, once as a function of population density (a), and once as a function of agricultural intensity (b).

Figure 3-1 illustrates the relationship between the total intake fraction of $\text{PM}_{2.5}$ and benzo(a)pyrene for different emission zones as a function of the population density and the agricultural intensity of that zone, calculated with IMPACT North America, using. I selected $\text{PM}_{2.5}$, which is a typical inhalation-dominated pollutant, because it is taken in entirely by inhalation and benzo(a)pyrene, a typical ingestion-dominant chemical, because more than 99% of its intake fraction is via agricultural produce (MacLeod et al. 2004). The agricultural intensity is measured as the mass (in metric tonnes of wet matter) of agricultural products (including cereals, vegetables, fruits and animal products such as milk and meat) produced over a certain surface (in km^2) during a certain time (in years).

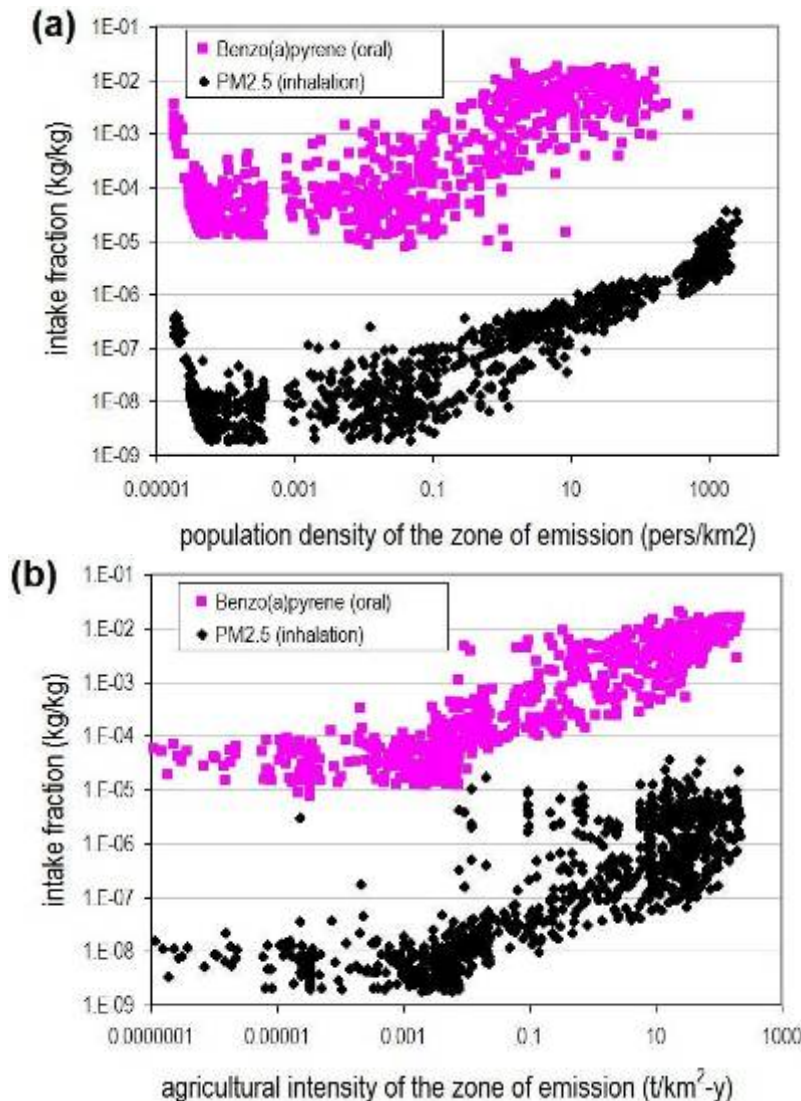


Figure 3-1: Total intake fractions of PM_{2.5} and benzo(a)pyrene, for different zones of emissions, as a function of (a) the population density and (b) the agricultural intensity of the zone of emission. Each dot represents one of the 1,121 zones, in the IMPACT North America model, for which an intake fraction has been modeled. The same zones and the same intake fraction associated with each zone is reported in both parts (a) and (b), but once as a function of (a) population density and (b) agricultural intensity.

The results from Figure 3-1 are aligned with the findings of Rochat et al. (2006) at a much lower resolution (continental scale) and show that for pollutants that have inhalation as a main intake pathway, the population density surrounding the zone of emission appears indeed to be an important parameter to be considered in the archetype. The same observation holds that pollutants dominated by the oral pathway have intake fractions that are influenced by agricultural intensity. However, Figure 3-1 shows that there is some correlation between intake fractions of benzo(a)pyrene and population density. This observation comes from the fact that over the entire range of air cells studied, there is no uninhabited region with substantial agricultural production (see Figure 3-2). At low population density levels, both population density and agricultural intensity result in low intake fractions for benzo(a)pyrene. Therefore, if there is a need to avoid multiplying the amount of information related to the location of emission, providing the

population density could be, depending on the question raised, a satisfactory proxy for agricultural production intensity.

Correlation between population density and agricultural production intensity

Agricultural production intensity (in tonnes of wet matter produced — grain, vegetables, fruits, roots, milk, meat, etc. — per km² per year) is plotted against population density (in person per km²) in Figure 3-2. Section 3.8 lists the numerical values for population density (persons/km²) and agricultural production intensity (t/(km²·y)) for each air grid cell in the North America model.

Values below 0.001 pers/km² or below 0.001 t/(km²·y) (hatched zones in Figure 3-2) should be evaluated with caution. In these ranges, absolute values can be uncertain by one or two orders of magnitude. For regions of low population or agriculture (approximately below 0.001 pers/km² or below 0.001 t/(km²·y)), low values were often given “by default” in GIS maps (so-called “border effects”). Analysis should be performed for values above 0.001 pers/km² and above 0.001 t/(km²·y). Figure 3-2 shows that population density and agricultural production intensity are moderately correlated (R² of 0.65). There are cells with medium to high population density but with low agricultural intensity. There are, however, no cells with high agricultural production intensity and low population density.

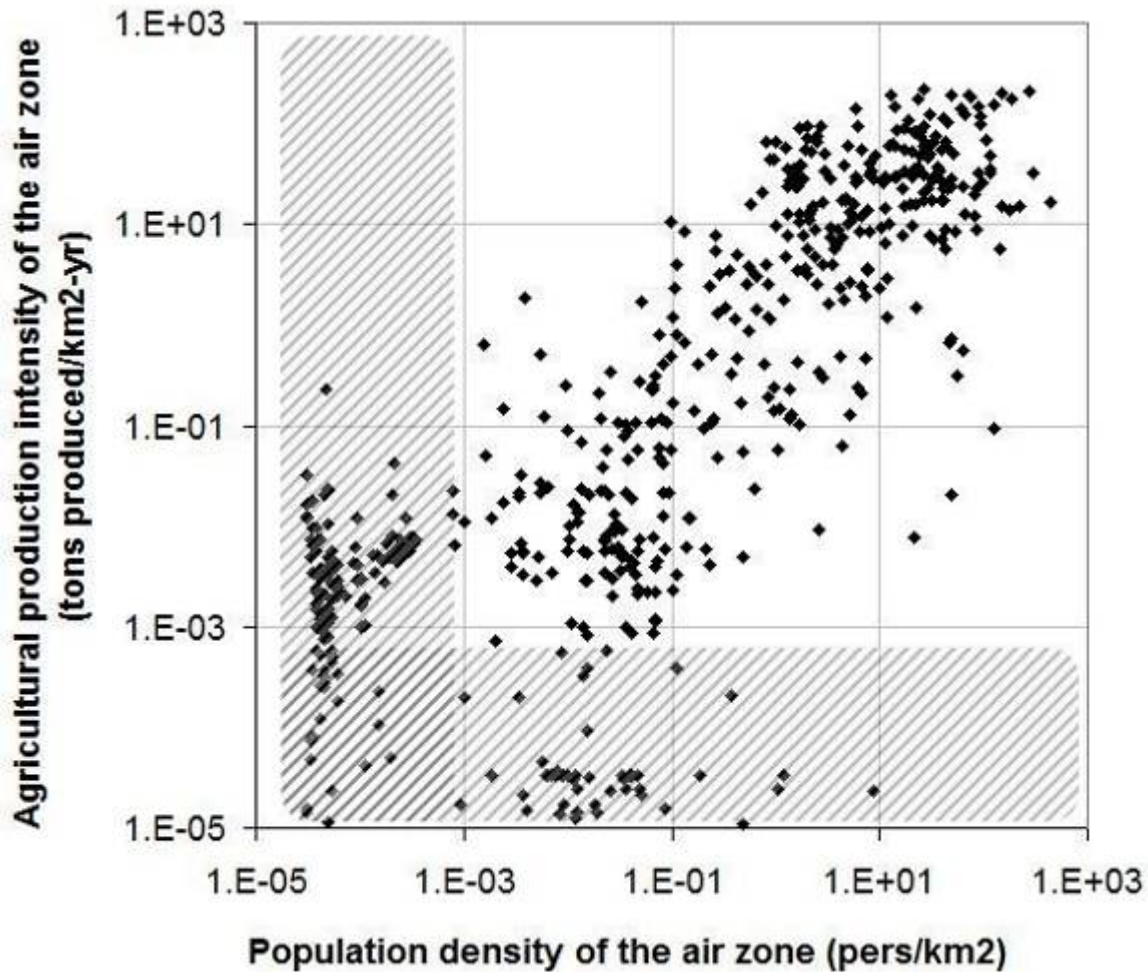


Figure 3-2: Population density versus agricultural production intensity in the different air cells defined in the model IMPACT North America (Chapter 2). Zones hatched should be interpreted with care as they represent “border effects” for zones with very low population density or agricultural intensity.

Lessons about archetypes related to population and agricultural intensity

In light of the above results, it is important to have archetypes that can both indicate the population density and the agricultural intensity of the emission zone. However, for remote regions such as Alaska, where both population and agricultural intensity is low, the archetype “remote” would be sufficient information for both pollutants that have inhalation or oral consumption as their main damaging pathway.

What are the archetypes that can be defined?

Considering the observations above, I suggest using the archetypes presented in Table 3-2 to implement regionalization in life-cycle assessment.

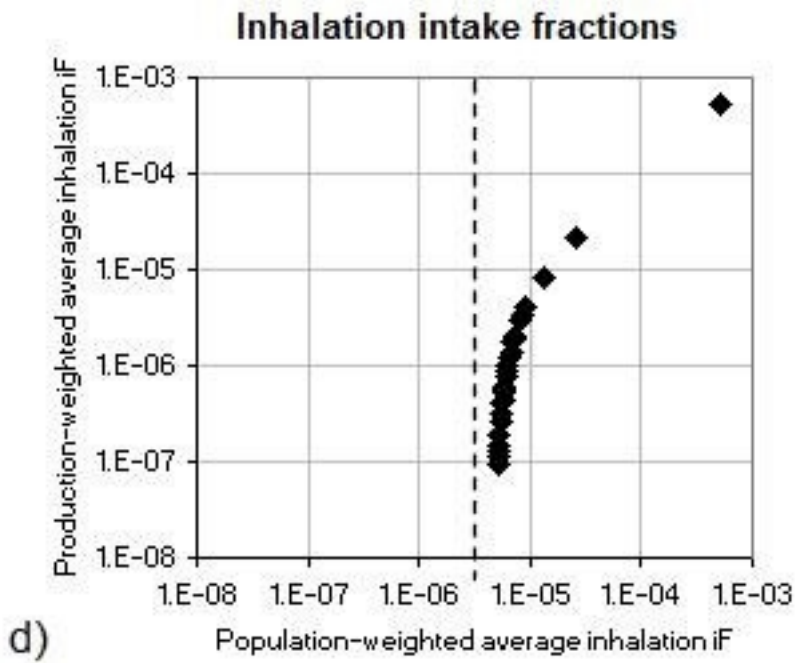
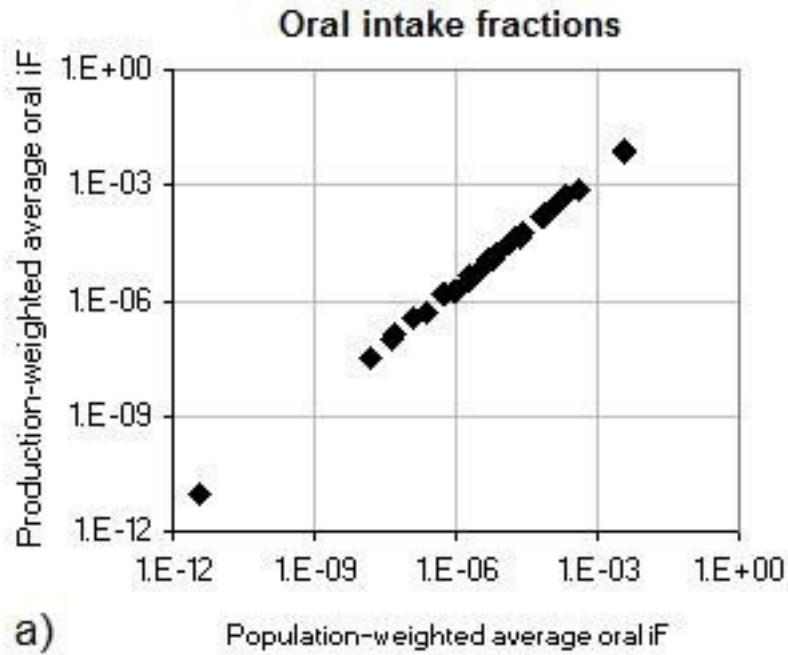
Table 3-2: Suggested archetypes to be used when performing regionalization for intake fractions of pollutants emitted into the air.

Archetype for outdoor, troposphere air emissions	Explanation
unspecified	emission weighted average
high population density, high agricultural intensity	e.g., Chicago
high population density, low agricultural intensity	e.g., Phoenix
high population density, non-agricultural	e.g., Las Vegas
medium population density, high agricultural intensity	e.g., Small town or countryside in Iowa
medium population density, low agricultural intensity	e.g., Small town or countryside in Montana
medium population density, non-agricultural	e.g., Small town in Nevada or Alaska
low population density (remote)	e.g., Oil platform North of Alaska – Note that low population density is always correlated with non-agricultural intensity (see SI). Can be used as a proxy for air emissions in coastal zones.
Archetypes for air emissions but not evaluated in this chapter	Explanation
coastal zones	
oceanic zones	
indoor	
high altitude	e.g., airplanes

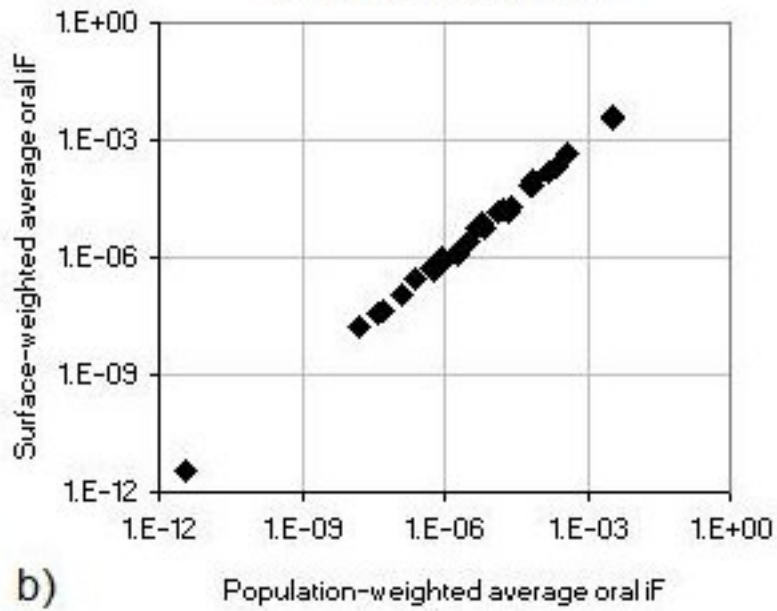
Generic intake fractions

Most life-cycle inventories do not contain any regional information. Only a generic characterization can be associated with life-cycle inventories that do not contain any regional information. A generic characterization factor is based on a generic intake fraction. Here I explore how to evaluate a generic intake fraction.

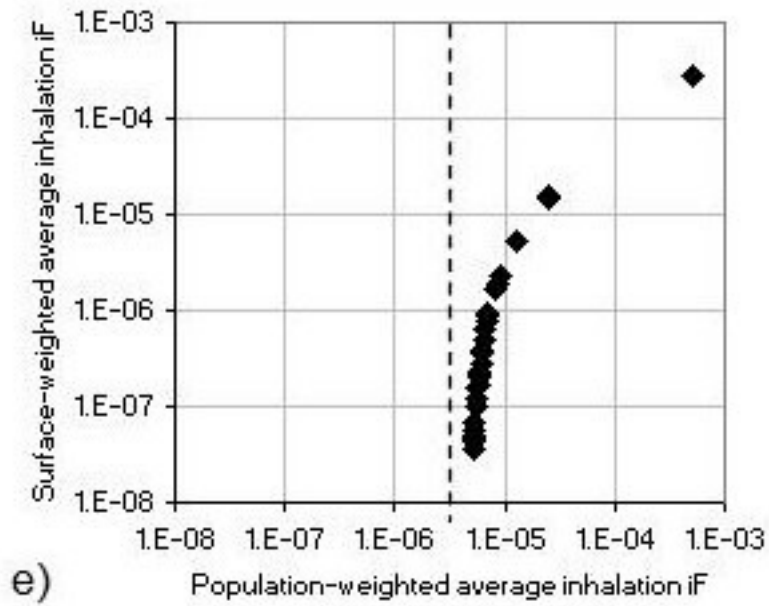
The generic intake fraction can be computed using a non-spatial model or calculated as a weighted average from the intake fractions computed using a spatial model. Three proxies are evaluated for the test set described above to calculate emissions-weighted average intake fractions: (i) population, (ii) agricultural production and (iii) surface (i.e., land area) (Figure 3-3). It means that an emission of 1 kg/h is distributed proportionally to (i) the population in North America (population in each air grid cell), (ii) agricultural production in North America (agricultural production, in kg of wet matter of all agricultural products in each air grid cell), and (iii) land area in North America (land area of each air grid cell). Section 3.8 lists the oral and inhalation intake fractions used to generate Figure 3-3. Furthermore, section 3.8 lists the population (persons), agricultural production intensity ($t/(km^2 \cdot y)$) and land area (km^2) for each air grid cell in the North America model used to distribute the emissions when generating the average intake fractions depicted in Figure 3-3.



Oral intake fractions



Inhalation intake fractions



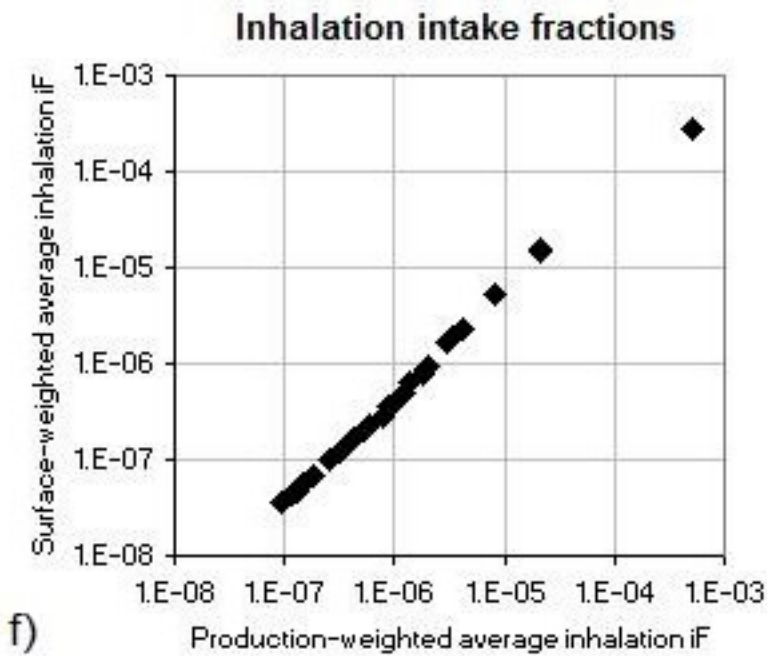
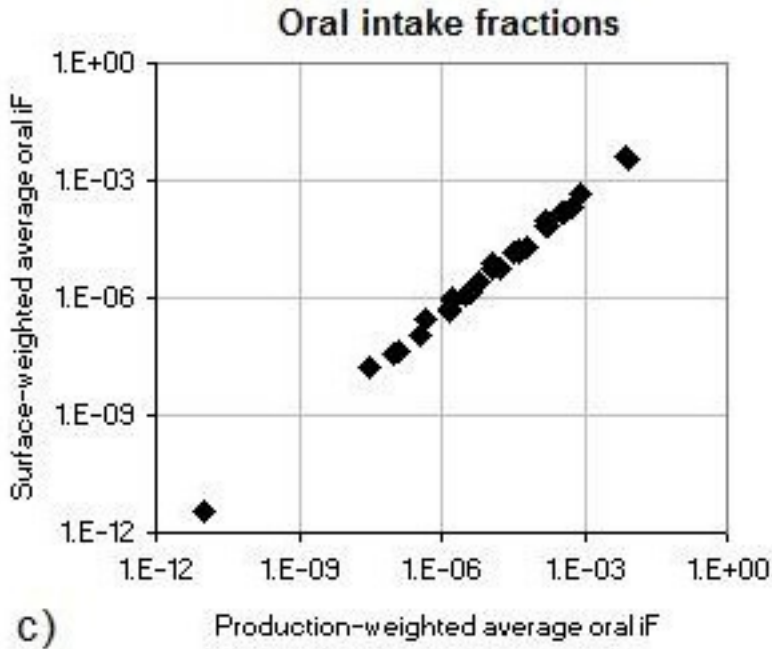


Figure 3-3: Population-weighted, production-weighted, and surface-weighted average oral and inhalation intake fractions (in kg taken in per kg emitted) for the substances presented in Table 3-1 (see Table 3-5 in supporting information for the intake fraction values).

An inhalation population-weighted average intake fraction is driven by urban emissions (see asymptote at approximately 5×10^{-6} kg inhaled per kg emitted in parts d) and e) of Figure 3-3). Therefore, even for short-lived substances, the total population-weighted average intake

fraction has a lower boundary corresponding to the urban intake fraction. For short lived-pollutants, the surface-weighted average intake fraction is slightly lower than production-weighted or population-weighted average intake fraction because of the amount of emissions assumed in the north of the continent where only reduced populations or agricultural production is present. For long-lived pollutants, the three proxies (i.e., population, agricultural production and surface area) give similar weighted average intake fractions. Therefore, it is short-lived substances that have their intake fractions mostly influenced by the type of proxy used for emissions.

The aim of generic intake fractions is to accurately evaluate the human health damage score for the emissions throughout the life cycle when no information is available on where the pollutants are emitted. At the scale of North America, emissions are assumed to be better represented by population distribution than by land surface (see section 2.4 in Chapter 2). For example, though Alaska and northern Canada represent up to a third to a half of North America, emissions occurring in these regions represent only a small fraction of total North American emissions. Therefore, as a first approximation, for air emissions, generic intake fractions are calculated as the population-weighted average intake fraction of an emission in the different zones of North America.

Geographically differentiated intake fractions versus generic intake fractions for air emissions

In this section I compare the magnitude and the variability of geographically differentiated intake fractions versus generic intake fractions for air emissions.

The variation in intake fraction as a function of the location of emission in North America is depicted in Figure 3-4 and Figure 3-5. Intake fractions are presented for, respectively, inhalation and oral intake fractions of the representative test set of pollutants described above, calculated with the IMPACT North America model (Chapter 2). Generic intake fractions are calculated as the population-weighted average intake fractions of an emission in the different zones of North America. For each pollutant, Figure 3-4 and Figure 3-5 also show the minimum and the maximum intake fraction as well as the 5th, 25th, 50th, 75th and 95th percentile of the distribution of intake fractions.

Intake fractions can vary by more than eight orders of magnitude depending on the location of emission. When 90% of the 831 air zones defined in the IMPACT North America model are considered, the variation can be up to six orders of magnitude. Actual intake fraction can be up to one order of magnitude higher and seven orders of magnitude lower than the generic intake fraction.

The complete list of numerical values is too large to be reported in this document and can be downloaded as an Excel table from <http://www.impactmodeling.org>.

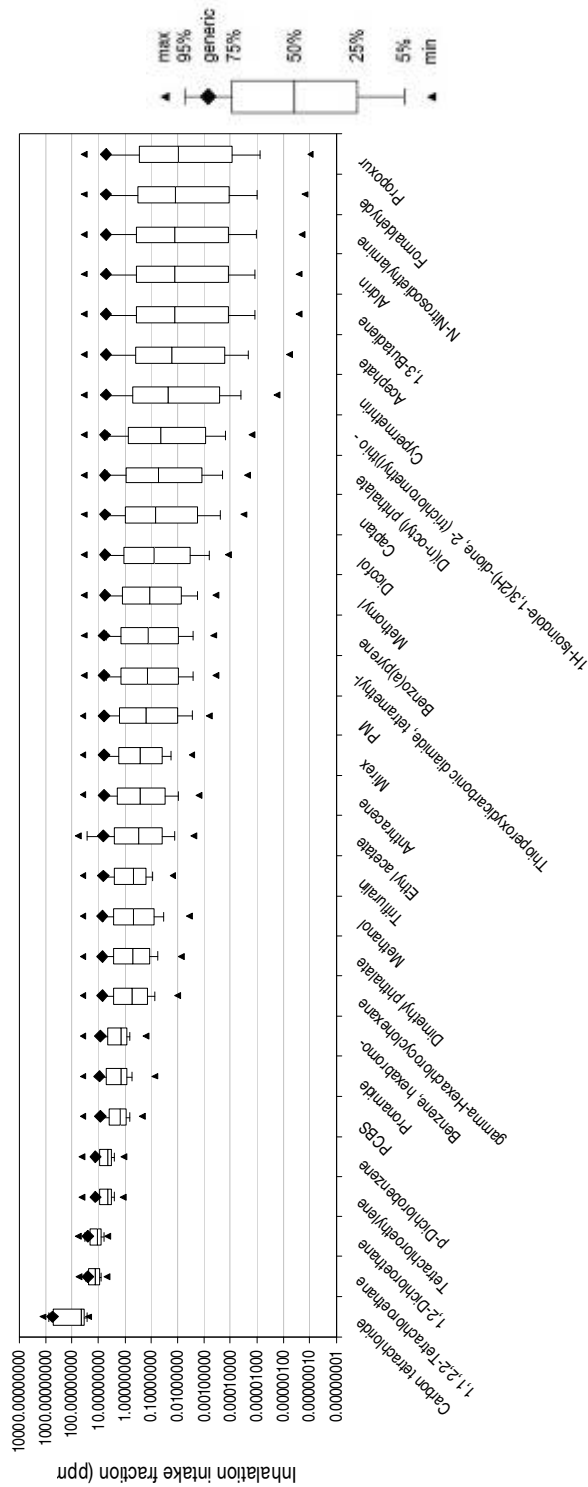


Figure 3-4: Variation in inhalation intake fractions, depending on the location of emission in North America. The dot represents the generic intake fraction of each pollutant, calculated as the population-weighted average intake fraction for each zone of emission.

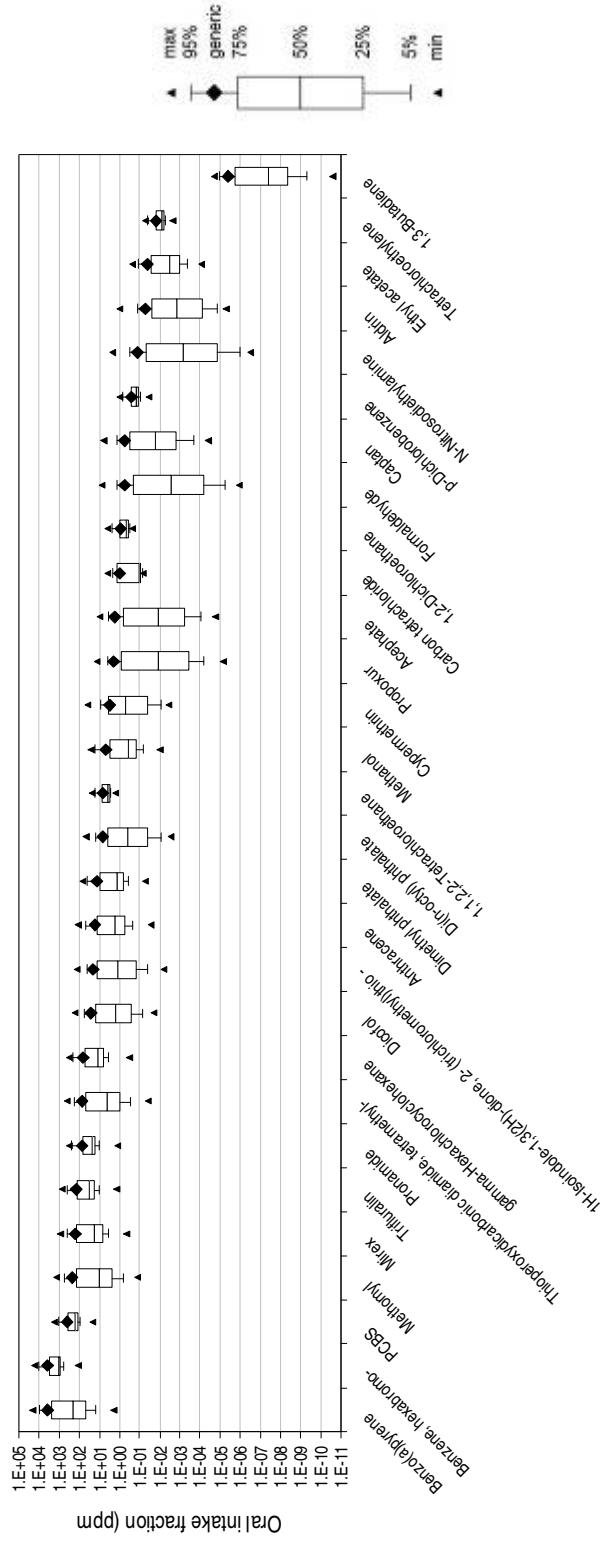


Figure 3-5: Variation in oral intake fraction depending on the location of emission in North America. The dot represents the generic intake fraction of each pollutant, calculated as the population-weighted average intake fraction for each zone of emission.

Comparison of geographic-based intake fractions and archetype-based intake fractions

In this section I compare “geographic” and “archetype” approaches when performing regionalization of intake fractions. The intake fractions are calculated with the IMPACT North America model, for a selected numbers of locations in the United States. These locations were selected following the need to have some locations representing urban, rural and remote regions (in terms of population density) throughout the United States (West, Rockies, Midwest and Alaska).

Table 3-3 shows the numerical values for the different air grid cells considered to model the intake fractions for **Error! Reference source not found.**

Table 3-3: Numerical values for the different air grid cells considered to model the intake fractions for Figure 3-6.

Cell	Region	Population density (pers/km ²)	Population archetype	Total intake fraction
X26	West	65	rural	7.9E-07
X27	West	100	rural	1.0E-06
X28	West	18	rural	4.8E-07
Y26	West	7.0	rural	5.3E-07
Y27	West	30	rural	1.0E-06
Y28	West	110	rural	1.5E-06
Y29	West	66	rural	7.4E-07
Z26	West	1.1	rural	3.9E-07
Z27	West	20	rural	8.5E-07
Z28	West	130	rural	1.6E-06
Z29	West	66	rural	1.1E-06
San Francisco	West	1,700	urban	1.4E-05
San Diego	West	1,300	urban	1.2E-05
AD24	Rockies	2.5	rural	2.9E-07
AD25	Rockies	11	rural	4.0E-07
AD26	Rockies	16	rural	4.0E-07
AE24	Rockies	2.4	rural	3.1E-07
AE26	Rockies	19	rural	4.3E-07
AF24	Rockies	1.6	rural	3.5E-07
AF25	Rockies	6.5	rural	4.0E-07
AF26	Rockies	6.7	rural	4.2E-07
Denver	Rockies	1,900	urban	8.9E-06
AJ24	Midwest	23	rural	9.8E-07
AJ25	Midwest	29	rural	1.2E-06
AJ26	Midwest	45	rural	1.2E-06
AK24	Midwest	110	rural	1.3E-06
AK26	Midwest	37	rural	1.4E-06
AL25	Midwest	110	rural	1.4E-06
AL26	Midwest	72	rural	1.5E-06
Chicago	Midwest	2,400	urban	2.4E-05
J11	Alaska	0.034	remote	2.3E-08
J12	Alaska	0.054	remote	2.2E-08
J13	Alaska	0.021	remote	1.9E-08
K11	Alaska	0.032	remote	1.8E-08
K12	Alaska	0.035	remote	1.7E-08
K13	Alaska	0.031	remote	1.5E-08
L11	Alaska	0.031	remote	1.4E-08
L12	Alaska	0.019	remote	1.4E-08
L13	Alaska	0.42	remote	1.6E-08
Fairbanks	Alaska	410	urban	9.0E-07

Figure 3-6 presents the inhalation intake fractions of particulate matter for different locations in the United States. The left side of Figure 3-6 groups these intake fractions in the different regions of United States. The right side groups the same intake fractions not by region but whether they occur in an urban, a rural or a remote location.

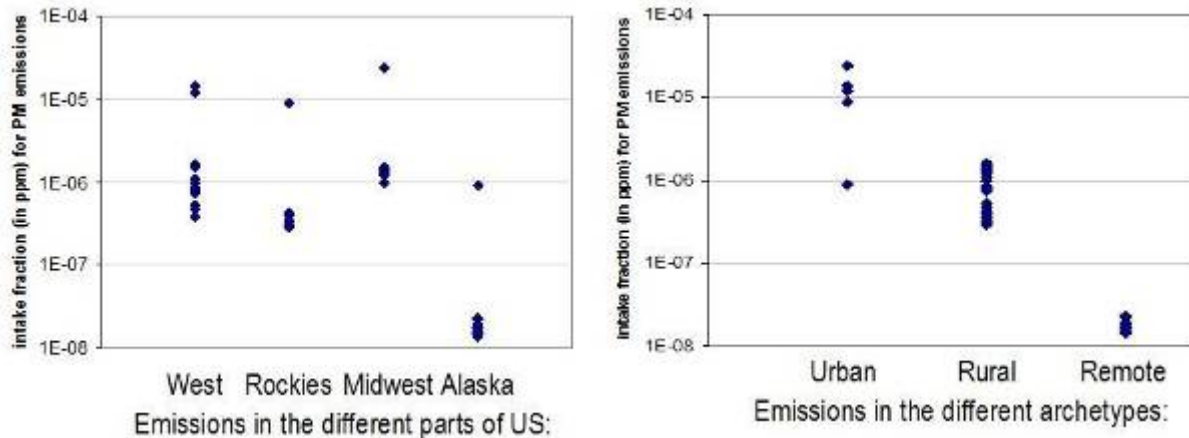


Figure 3-6: Intake fraction of particulate matter grouped per region (left) and per characteristics of population density where the emission occurred (right).

Figure 3-6 shows that intake fractions are more influenced by the archetype function of the population density (e.g., urban, rural, etc.) than by the actual location in North America because the range of intake fractions within one region of North America is larger than the range among the different regions of North America. With the archetype approach, fewer intake fraction values are necessary than with the geographic approach to get an accurate intake fraction, rendering the application of regionalization in life-cycle assessment more practical.

3.5. Conclusions

This chapter shows that regionalization is important, and that the archetype-based approach is a practical way to perform regionalization as it is as accurate as the geographically based regionalization, but more practical to implement due to significantly less required data. Furthermore, archetype-based regionalization can be more easily implemented in life-cycle inventory databases because the information regarding the archetype can be directly attached to the elementary flows. For example, inventory databases such as ecoinvent (Frischknecht 2005) can have a label (so-called “sub-compartment” in life-cycle assessment software such as SimaPro – PRÉ 2006) attached to each elementary flow (or pollutant) that gives some indications on where the pollutant is emitted (e.g., “high population density,” “low population density,” etc.). This chapter shows that for outdoor air emissions, archetypes should not only be based on population density but also on agricultural production intensity. It shows that urban and rural archetypes are important, but also the remote archetype is important as emissions in remote areas can have significantly lower intake fractions than when emitted in urban or rural areas or in high or low agricultural production intensity regions, both for inhalation and oral intake fractions.

Future research needs

In this chapter I explore the question of regionalization of intake fractions of air pollutants emitted outdoors in North America. This type of work should be expanded to other media of emission such as high altitude, water or soil, other regions of the world as well as other types of impact categories such as damage related to tropospheric ozone formation, ecotoxicity, acidification, eutrophication, land use or water use. Further research should be conducted to better assess and validate the optimum archetypes for the different impact categories, media of emissions, world regions, and types of pollutants. Furthermore, because medium-lived pollutants such as primary PM_{2.5} are both influenced by local and continental parameters, the possibility of

multiscale modeling should be explored as it could be an approach to capture both local and continental intake fractions.

Outlook

As long as no detailed geographically differentiated inventories are available, matched with impact assessments, I suggest using the archetype approach to make the regionalization practical in life-cycle assessment, which is important, needed and demanded (see Chapter 5 for an application). Indeed, the lack of regionalization in life-cycle assessment has been discussed since the 1990s and has been considered a limitation to the use of human health damage life-cycle assessment-based results in decision making (Potting and Hauschild 2006, Sedlbauer et al. 2007, Reap et al. 2008, Margni et al. 2008). However, up to this point, life-cycle assessment studies that have integrated regionalization have been rare, mainly because of the data intensiveness and many computations needed to perform geographically differentiated regionalization. The archetypes-based regionalization approach can significantly contribute to building confidence in decisions based on life-cycle assessments evaluating human health damage from air pollution by improving accuracy and precision.

In Chapter 5, I will provide a further evaluation of the issue of regionalization in life-cycle assessment with a focus on PM. However, because PM is such a dominant contributors to human health damage, further analysis of this pollutant is done in Chapter 4 prior to evaluating total human health damage from different processes in Chapter 5.

3.6. Acknowledgments

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3.8. Supporting information for this chapter

Table 3-4 presents the total area (“Area”), land area (soil and paved area), population, population density and agricultural intensity of the different air cells defined in the model IMPACT North America used to plot Figure 3-2.

Table 3-4: Total area (“Area”), Land area (soil and paved area), Population, Population density and Agricultural intensity of the different air cells defined in the model IMPACT North America.

Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))	Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))
AA10	1.8.E+04	1.6.E+04	1.0.E+00	5.5.E-05	4.5.E-03	AP12	2.2.E+04	6.4.E+03	2.2.E+00	1.0.E-04	1.6.E-03
AA11	2.0.E+04	1.3.E+04	1.0.E+00	4.9.E-05	3.0.E-03	AP13	2.4.E+04	1.2.E+04	2.7.E+03	1.1.E-01	3.4.E-03
AA12	2.2.E+04	1.9.E+04	7.7.E+01	3.5.E-03	6.8.E-03	AP14	2.6.E+04	5.9.E+03	9.6.E+02	3.7.E-02	9.9.E-04
AA13	2.4.E+04	2.2.E+04	1.3.E+02	5.3.E-03	2.1.E-02	AP15	2.8.E+04	2.6.E+04	1.2.E+03	4.3.E-02	4.4.E-03
AA14	2.6.E+04	2.6.E+04	1.7.E+04	6.3.E-01	2.3.E-02	AP16	3.0.E+04	2.7.E+04	2.6.E+00	8.5.E-05	4.4.E-03
AA15	4.0.E+04	3.1.E+04	5.1.E+02	1.3.E-02	1.3.E-02	AP17	3.2.E+04	2.0.E+04	3.3.E+02	1.0.E-02	7.5.E-03
AA16	3.0.E+04	2.4.E+04	3.4.E+03	1.1.E-01	8.1.E-01	AP18	3.4.E+04	2.9.E+04	5.4.E+02	1.6.E-02	2.0.E-02
AA17	3.2.E+04	3.1.E+04	9.1.E+03	2.9.E-01	3.2.E+00	AP19	3.6.E+04	3.2.E+04	2.0.E+02	5.5.E-03	2.6.E-02
AA18	3.4.E+04	3.4.E+04	5.4.E+04	1.6.E+00	3.1.E+01	AP20	3.7.E+04	3.4.E+04	7.4.E+02	2.0.E-02	2.1.E-01
AA19	3.8.E+04	3.7.E+04	1.3.E+06	3.4.E+01	7.4.E+01	AP21	3.9.E+04	3.7.E+04	1.3.E+04	3.3.E-01	1.5.E+00
AA20	3.9.E+04	3.9.E+04	1.3.E+06	3.2.E+01	6.3.E+01	AP22	4.1.E+04	3.9.E+04	5.2.E+04	1.3.E+00	1.2.E+01
AA21	3.9.E+04	3.5.E+04	2.3.E+05	6.0.E+00	2.9.E+01	AP23	4.2.E+04	3.5.E+04	1.9.E+06	4.4.E+01	2.5.E+01
AA22	4.1.E+04	3.4.E+04	2.1.E+05	5.3.E+00	2.7.E+00	AP24	4.6.E+04	1.2.E+03	2.2.E+06	4.8.E+01	6.8.E-01
AA23	4.2.E+04	3.2.E+03	4.0.E+04	9.4.E-01	2.4.E-01	AP25	5.1.E+04	4.8.E+04	5.8.E+06	1.1.E+02	4.8.E+01
AA24	5.0.E+04	5.0.E+04	2.1.E+05	4.2.E+00	4.8.E-01	AP26	5.1.E+04	4.2.E+04	1.1.E+07	2.2.E+02	1.5.E+01
AA25	5.0.E+04	4.9.E+04	1.8.E+04	3.6.E-01	3.4.E+00	AP27	5.0.E+04	2.8.E+04	2.6.E+06	5.3.E+01	8.6.E+00
AA26	5.0.E+04	4.9.E+04	2.7.E+04	5.4.E-01	3.8.E+00	AP28	5.0.E+04	1.6.E+04	5.8.E+05	1.1.E+01	6.5.E+00
AA27	5.0.E+04	5.0.E+04	2.3.E+05	4.5.E+00	1.8.E+00	AP29	5.2.E+04	3.0.E-04	1.0.E+00	1.9.E-05	0.0.E+00
AA28	5.0.E+04	5.0.E+04	2.5.E+05	5.1.E+00	1.3.E+01	AP4	3.8.E+03	3.8.E+03	1.0.E+00	2.6.E-04	0.0.E+00
AA29	5.1.E+04	4.4.E+04	3.9.E+05	7.7.E+00	1.4.E+01	AP5	9.6.E+03	9.5.E+03	2.8.E+00	2.9.E-04	0.0.E+00
AA30	5.3.E+04	8.3.E+02	2.8.E+05	5.2.E+00	1.3.E-01	AP6	9.8.E+03	9.2.E+03	2.9.E+00	3.0.E-04	0.0.E+00
AA6	9.8.E+03	3.5.E+02	1.0.E+00	1.0.E-04	0.0.E+00	AP7	1.2.E+04	2.4.E+02	1.0.E+00	8.4.E-05	0.0.E+00
AA7	1.2.E+04	4.6.E+03	1.0.E+00	8.4.E-05	0.0.E+00	AP8	1.4.E+04	3.0.E-04	1.0.E+00	7.1.E-05	0.0.E+00
AA8	1.4.E+04	9.2.E+03	2.5.E+00	1.8.E-04	0.0.E+00	AP9	1.6.E+04	7.1.E+03	1.5.E+00	9.6.E-05	4.0.E-03
AA9	1.6.E+04	9.0.E+03	1.0.E+00	6.2.E-05	2.9.E-03	AQ10	1.8.E+04	1.5.E+04	3.3.E+00	1.8.E-04	6.8.E-03
AB10	2.0.E+04	1.9.E+04	1.0.E+00	5.1.E-05	5.0.E-03	AQ11	2.0.E+04	1.6.E+04	4.9.E+00	2.4.E-04	4.6.E-03
AB11	2.0.E+04	1.5.E+04	1.0.E+00	4.9.E-05	4.0.E-03	AQ12	2.2.E+04	6.7.E+03	2.3.E+00	1.0.E-04	1.7.E-03
AB12	2.2.E+04	2.0.E+04	1.0.E+00	4.5.E-05	7.7.E-04	AQ13	2.4.E+04	1.7.E+04	5.6.E+00	2.3.E-04	4.5.E-03
AB13	2.4.E+04	2.2.E+04	2.3.E+00	9.4.E-05	1.2.E-02	AQ14	2.6.E+04	4.1.E+03	1.0.E+00	3.8.E-05	5.9.E-04
AB14	2.6.E+04	2.5.E+04	2.9.E+02	1.1.E-02	1.6.E-02	AQ15	2.8.E+04	2.7.E+04	1.0.E+00	3.5.E-05	3.4.E-03
AB15	2.8.E+04	1.8.E+04	2.3.E+03	8.2.E-02	1.2.E-02	AQ16	3.0.E+04	2.9.E+04	1.0.E+00	3.3.E-05	4.9.E-03
AB16	3.0.E+04	2.7.E+04	1.3.E+04	4.2.E-01	4.7.E-01	AQ17	3.2.E+04	2.9.E+04	1.0.E+00	3.1.E-05	1.3.E-02
AB17	3.2.E+04	3.2.E+04	2.6.E+04	8.0.E-01	3.9.E+00	AQ18	3.4.E+04	3.0.E+04	1.1.E+02	3.3.E-03	2.0.E-02
AB18	3.4.E+04	3.3.E+04	5.5.E+04	1.6.E+00	2.4.E+01	AQ19	3.6.E+04	3.2.E+04	2.3.E+02	6.5.E-03	2.5.E-02
AB19	3.7.E+04	3.6.E+04	1.0.E+05	2.7.E+00	9.5.E+01	AQ20	3.7.E+04	3.5.E+04	3.6.E+03	9.7.E-02	4.8.E-01

Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))	Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))
AB20	3.9.E+04	3.8.E+04	9.8.E+04	2.5.E+00	6.5.E+01	AQ21	3.9.E+04	3.8.E+04	2.5.E+04	6.5.E-01	3.1.E+00
AB21	3.9.E+04	2.6.E+04	5.7.E+04	1.5.E+00	3.5.E+01	AQ22	4.1.E+04	3.8.E+04	4.6.E+05	1.1.E+01	1.4.E+01
AB22	4.1.E+04	8.8.E+02	1.8.E+05	4.4.E+00	6.4.E-02	AQ23	4.3.E+04	3.4.E+04	5.0.E+06	1.1.E+02	3.2.E+01
AB23	4.2.E+04	1.6.E+02	1.1.E+05	2.6.E+00	9.2.E-03	AQ24	4.6.E+04	1.3.E+03	2.2.E+06	4.9.E+01	7.2.E-01
AB24	5.0.E+04	4.9.E+04	3.4.E+05	6.7.E+00	1.2.E+01	AQ25	5.3.E+04	3.9.E+04	2.3.E+07	4.4.E+02	1.6.E+01
AB25	5.1.E+04	4.9.E+04	2.1.E+06	4.2.E+01	8.9.E+00	AQ26	4.8.E+04	5.5.E+03	1.1.E+06	2.2.E+01	1.5.E+00
AB26	5.0.E+04	4.8.E+04	1.2.E+05	2.3.E+00	4.8.E+00	AQ27	4.9.E+04	3.0.E-04	1.0.E+00	2.0.E-05	0.0.E+00
AB27	5.0.E+04	4.9.E+04	6.3.E+04	1.3.E+00	4.8.E+00	AQ28	5.1.E+04	3.0.E-04	1.0.E+00	2.0.E-05	0.0.E+00
AB28	5.1.E+04	5.0.E+04	3.3.E+05	6.5.E+00	8.3.E+00	AQ4	4.0.E+03	4.0.E+03	1.0.E+00	2.5.E-04	0.0.E+00
AB29	5.2.E+04	5.1.E+04	4.4.E+06	8.5.E+01	8.9.E+00	AQ5	9.8.E+03	9.6.E+03	2.8.E+00	2.9.E-04	0.0.E+00
AB30	5.3.E+04	1.3.E+04	5.2.E+05	9.9.E+00	2.3.E+00	AQ6	9.8.E+03	2.8.E+03	2.3.E+00	2.4.E-04	0.0.E+00
AB6	9.8.E+03	2.4.E+03	1.0.E+00	1.0.E-04	0.0.E+00	AQ7	1.2.E+04	3.0.E-04	1.0.E+00	8.4.E-05	0.0.E+00
AB7	1.2.E+04	3.8.E+03	1.0.E+00	8.4.E-05	0.0.E+00	AQ8	1.4.E+04	3.0.E-04	1.0.E+00	7.1.E-05	0.0.E+00
AB8	1.4.E+04	8.1.E+03	2.2.E+00	1.6.E-04	0.0.E+00	AQ9	1.6.E+04	3.1.E+02	1.0.E+00	6.2.E-05	1.8.E-04
AB9	1.6.E+04	6.3.E+03	1.0.E+00	6.2.E-05	2.0.E-03	AR10	1.8.E+04	1.0.E+04	2.6.E+00	1.4.E-04	5.2.E-03
AC10	1.9.E+04	1.8.E+04	1.0.E+00	5.2.E-05	5.0.E-03	AR11	2.0.E+04	2.0.E+04	6.6.E+00	3.3.E-04	6.6.E-03
AC11	2.0.E+04	1.4.E+04	1.0.E+00	4.9.E-05	3.2.E-03	AR12	2.2.E+04	2.1.E+04	6.2.E+00	2.8.E-04	5.4.E-03
AC12	2.2.E+04	1.7.E+04	7.6.E+01	3.4.E-03	2.0.E-04	AR13	2.4.E+04	2.4.E+04	7.1.E+00	2.9.E-04	5.7.E-03
AC13	2.4.E+04	2.2.E+04	2.3.E+00	9.5.E-05	2.9.E-03	AR14	2.6.E+04	1.1.E+04	3.6.E+00	1.4.E-04	3.4.E-03
AC14	2.6.E+04	2.0.E+04	7.2.E+00	2.7.E-04	1.2.E-02	AR15	2.8.E+04	1.8.E+04	7.7.E+02	2.7.E-02	2.1.E-03
AC15	2.8.E+04	1.8.E+04	5.9.E+00	2.1.E-04	7.9.E-03	AR16	3.0.E+04	2.8.E+04	1.0.E+00	3.3.E-05	7.5.E-03
AC16	5.7.E+04	4.7.E+04	1.4.E+02	2.4.E-03	1.5.E-01	AR17	3.2.E+04	3.1.E+04	1.0.E+00	3.1.E-05	1.2.E-02
AC17	3.2.E+04	3.0.E+04	3.5.E+03	1.1.E-01	4.0.E+00	AR18	3.4.E+04	3.1.E+04	1.1.E+02	3.4.E-03	2.1.E-02
AC18	3.4.E+04	3.2.E+04	1.9.E+04	5.7.E-01	1.6.E+01	AR19	3.6.E+04	3.2.E+04	2.2.E+02	6.1.E-03	1.2.E-01
AC19	3.7.E+04	3.6.E+04	7.4.E+04	2.0.E+00	7.1.E+01	AR20	3.7.E+04	3.6.E+04	1.4.E+02	3.8.E-03	1.9.E+00
AC20	3.9.E+04	3.8.E+04	4.9.E+04	1.2.E+00	5.8.E+01	AR21	3.9.E+04	3.8.E+04	2.7.E+05	6.8.E+00	2.5.E+00
AC21	3.9.E+04	3.9.E+04	3.8.E+04	9.8.E-01	4.5.E+01	AR22	4.1.E+04	9.2.E+03	1.2.E+06	2.9.E+01	2.3.E+01
AC22	4.1.E+04	1.2.E+03	3.4.E+04	8.3.E-01	1.2.E+00	AR23	4.2.E+04	1.4.E+04	7.5.E+05	1.8.E+01	1.5.E+01
AC23	5.0.E+04	4.9.E+04	2.0.E+05	4.0.E+00	8.8.E+00	AR24	4.7.E+04	3.5.E+04	6.7.E+06	1.4.E+02	5.6.E+00
AC24	5.0.E+04	5.0.E+04	5.1.E+04	1.0.E+00	9.6.E+00	AR25	4.7.E+04	1.0.E+04	2.6.E+06	5.4.E+01	3.1.E-01
AC25	5.0.E+04	5.0.E+04	8.2.E+04	1.6.E+00	3.5.E+00	AR26	4.8.E+04	3.0.E-04	1.0.E+00	2.1.E-05	0.0.E+00
AC26	5.0.E+04	5.0.E+04	2.4.E+05	4.8.E+00	1.8.E+01	AR4	4.3.E+03	4.3.E+03	1.0.E+00	2.3.E-04	0.0.E+00
AC27	5.0.E+04	5.0.E+04	2.4.E+05	4.9.E+00	1.0.E+01	AR5	9.2.E+03	9.1.E+03	2.6.E+00	2.9.E-04	0.0.E+00
AC28	5.1.E+04	5.0.E+04	1.7.E+05	3.4.E+00	9.2.E+00	AR6	9.8.E+03	9.1.E+02	1.7.E+01	1.7.E-03	0.0.E+00
AC29	5.1.E+04	5.0.E+04	1.2.E+05	2.4.E+00	8.7.E+00	AR7	1.2.E+04	3.5.E-03	1.2.E+01	9.8.E-04	0.0.E+00
AC30	5.3.E+04	1.4.E+04	2.2.E+05	4.2.E+00	2.3.E+00	AR8	1.4.E+04	3.0.E-04	1.0.E+00	7.1.E-05	0.0.E+00
AC6	9.8.E+03	6.3.E+02	1.0.E+00	1.0.E-04	0.0.E+00	AR9	1.6.E+04	3.0.E-04	1.0.E+00	6.2.E-05	0.0.E+00
AC7	1.2.E+04	3.2.E+03	1.0.E+00	8.4.E-05	0.0.E+00	AS10	1.8.E+04	4.2.E+03	8.5.E+02	4.7.E-02	2.1.E-03
AC8	1.4.E+04	7.0.E+03	1.9.E+00	1.4.E-04	0.0.E+00	AS11	2.0.E+04	1.7.E+04	4.9.E+00	2.4.E-04	7.2.E-03
AC9	1.6.E+04	6.1.E+03	1.0.E+00	6.2.E-05	2.0.E-03	AS12	2.2.E+04	2.1.E+04	7.0.E+00	3.1.E-04	6.4.E-03
AD10	2.0.E+04	1.9.E+04	1.0.E+00	5.0.E-05	5.0.E-03	AS13	2.4.E+04	2.3.E+04	8.0.E+00	3.3.E-04	7.7.E-03
AD11	2.0.E+04	1.5.E+04	1.4.E+03	6.8.E-02	2.2.E-03	AS14	2.6.E+04	1.8.E+04	5.5.E+03	2.1.E-01	5.9.E-03
AD12	2.2.E+04	2.2.E+04	1.0.E+00	4.5.E-05	2.5.E-04	AS15	2.8.E+04	2.8.E+03	4.0.E+02	1.4.E-02	3.2.E-04

Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))	Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))
AD13	2.4.E+04	2.3.E+04	2.5.E+00	1.0.E-04	1.0.E-03	AS16	3.0.E+04	1.3.E+04	2.2.E+03	7.2.E-02	4.5.E-03
AD14	2.6.E+04	2.3.E+04	5.2.E+00	2.0.E-04	4.8.E-03	AS17	3.2.E+04	2.9.E+04	1.0.E+00	3.1.E-05	3.2.E-02
AD15	2.8.E+04	2.1.E+04	5.8.E+00	2.0.E-04	2.1.E-02	AS18	3.4.E+04	3.0.E+04	7.1.E+00	2.1.E-04	4.2.E-02
AD16	3.0.E+04	2.5.E+04	2.3.E+03	7.8.E-02	1.2.E-01	AS19	3.6.E+04	3.2.E+04	1.7.E+00	4.7.E-05	2.3.E-01
AD17	3.2.E+04	2.7.E+04	1.7.E+02	5.4.E-03	5.1.E-01	AS20	3.7.E+04	3.4.E+04	5.8.E+01	1.6.E-03	6.3.E-01
AD18	3.4.E+04	3.0.E+04	9.0.E+03	2.6.E-01	5.4.E+00	AS21	3.9.E+04	3.0.E+04	1.5.E+05	3.8.E+00	6.1.E+00
AD19	3.6.E+04	3.3.E+04	3.2.E+05	9.1.E+00	4.9.E+01	AS22	4.0.E+04	3.4.E+04	2.3.E+05	5.6.E+00	1.6.E+01
AD20	3.9.E+04	3.8.E+04	7.5.E+04	1.9.E+00	5.5.E+01	AS23	4.2.E+04	3.3.E+03	5.1.E+05	1.2.E+01	1.2.E+00
AD21	3.9.E+04	3.7.E+04	3.4.E+04	8.7.E-01	4.4.E+01	AS24	4.5.E+04	8.2.E+02	6.4.E+04	1.4.E+00	1.3.E-01
AD22	4.1.E+04	8.5.E+02	2.3.E+04	5.6.E-01	8.6.E-01	AS25	4.7.E+04	1.2.E+02	1.8.E+04	3.8.E-01	2.1.E-04
AD23	5.0.E+04	5.0.E+04	8.5.E+04	1.7.E+00	1.1.E+01	AS4	4.0.E+03	3.9.E+03	1.0.E+00	2.5.E-04	0.0.E+00
AD24	5.0.E+04	5.0.E+04	9.3.E+04	1.8.E+00	2.8.E+01	AS5	7.6.E+03	6.1.E+03	1.8.E+00	2.4.E-04	0.0.E+00
AD25	5.0.E+04	5.0.E+04	5.7.E+05	1.1.E+01	2.7.E+01	AS6	9.8.E+03	8.6.E-03	2.9.E+01	2.9.E-03	0.0.E+00
AD26	5.0.E+04	5.0.E+04	1.4.E+06	2.7.E+01	2.0.E+01	AS7	1.2.E+04	9.5.E-03	3.2.E+01	2.7.E-03	0.0.E+00
AD27	5.0.E+04	5.0.E+04	1.4.E+05	2.8.E+00	1.6.E+01	AS8	1.4.E+04	3.0.E-04	1.0.E+00	7.1.E-05	0.0.E+00
AD28	5.0.E+04	5.0.E+04	1.0.E+06	2.0.E+01	9.8.E+00	AS9	1.6.E+04	3.0.E-04	1.0.E+00	6.2.E-05	0.0.E+00
AD29	5.1.E+04	5.0.E+04	2.9.E+05	5.6.E+00	1.1.E+01	AT10	1.8.E+04	4.7.E+01	1.0.E+00	5.5.E-05	2.4.E-05
AD30	5.3.E+04	1.5.E+04	2.3.E+06	4.4.E+01	5.6.E+00	AT11	2.0.E+04	2.9.E+03	1.0.E+00	4.9.E-05	1.3.E-03
AD6	9.8.E+03	3.9.E+02	1.0.E+00	1.0.E-04	0.0.E+00	AT12	2.2.E+04	1.9.E+04	1.5.E+03	6.7.E-02	7.7.E-03
AD7	1.2.E+04	9.9.E+02	1.0.E+00	8.4.E-05	0.0.E+00	AT13	2.4.E+04	1.2.E+04	4.0.E+00	1.6.E-04	4.6.E-03
AD8	1.4.E+04	5.6.E+03	1.5.E+00	1.1.E-04	0.0.E+00	AT14	2.6.E+04	1.7.E+04	5.6.E+00	2.1.E-04	5.7.E-03
AD9	1.6.E+04	1.3.E+04	1.0.E+00	6.2.E-05	4.1.E-03	AT15	2.8.E+04	1.2.E+03	1.0.E+00	3.5.E-05	3.7.E-04
AE10	1.8.E+04	9.3.E+03	1.0.E+00	5.5.E-05	2.7.E-03	AT16	3.0.E+04	1.3.E+04	6.7.E+02	2.2.E-02	7.4.E-03
AE11	2.0.E+04	1.2.E+04	1.0.E+02	5.1.E-03	2.9.E-03	AT17	3.2.E+04	3.1.E+04	1.0.E+00	3.1.E-05	1.6.E-02
AE12	2.2.E+04	2.1.E+04	1.0.E+00	4.5.E-05	2.8.E-04	AT18	3.4.E+04	3.0.E+04	1.3.E+03	3.8.E-02	4.5.E-02
AE13	2.4.E+04	2.3.E+04	1.0.E+00	4.1.E-05	2.0.E-03	AT19	3.6.E+04	3.1.E+04	1.3.E+04	3.7.E-01	3.3.E-01
AE14	2.6.E+04	2.3.E+04	1.0.E+00	3.8.E-05	6.9.E-03	AT20	3.7.E+04	3.4.E+04	3.4.E+04	9.0.E-01	1.1.E+00
AE15	2.8.E+04	2.3.E+04	1.0.E+00	3.5.E-05	1.8.E-02	AT21	3.9.E+04	2.4.E+04	8.0.E+04	2.1.E+00	1.2.E+01
AE16	3.0.E+04	2.4.E+04	1.1.E+03	3.6.E-02	7.7.E-02	AT22	4.0.E+04	3.9.E+04	1.9.E+05	4.7.E+00	1.8.E+01
AE17	3.2.E+04	2.6.E+04	1.1.E+03	3.6.E-02	1.0.E-01	AT23	4.2.E+04	2.5.E+04	3.4.E+05	8.1.E+00	8.4.E+00
AE18	3.4.E+04	2.4.E+04	4.5.E+03	1.3.E-01	8.4.E+00	AT24	4.5.E+04	3.5.E+03	3.5.E+04	7.8.E-01	4.0.E-01
AE19	3.6.E+04	2.5.E+04	4.7.E+04	1.3.E+00	2.3.E+01	AT4	3.4.E+03	3.4.E+03	1.0.E+00	2.9.E-04	0.0.E+00
AE20	3.9.E+04	3.8.E+04	2.6.E+05	6.7.E+00	5.6.E+01	AT5	7.6.E+03	3.2.E+03	2.2.E+01	3.0.E-03	0.0.E+00
AE21	3.9.E+04	3.0.E+04	6.9.E+04	1.8.E+00	3.9.E+01	AT6	9.8.E+03	3.6.E-03	1.2.E+01	1.2.E-03	0.0.E+00
AE22	5.0.E+04	4.9.E+04	6.4.E+04	1.3.E+00	3.5.E+01	AU11	2.0.E+04	2.5.E+01	1.0.E+00	4.9.E-05	1.2.E-05
AE23	5.0.E+04	4.9.E+04	1.5.E+05	3.0.E+00	3.4.E+01	AU12	2.2.E+04	1.4.E+04	5.9.E+02	2.6.E-02	6.0.E-03
AE24	5.0.E+04	5.0.E+04	8.4.E+04	1.7.E+00	3.8.E+01	AU13	2.4.E+04	7.8.E+03	2.5.E+00	1.0.E-04	3.0.E-03
AE25	5.0.E+04	5.0.E+04	4.1.E+05	8.2.E+00	3.3.E+01	AU14	2.6.E+04	2.8.E+03	1.0.E+00	3.8.E-05	1.0.E-03
AE26	5.1.E+04	5.0.E+04	2.1.E+06	4.1.E+01	2.2.E+01	AU15	2.8.E+04	1.9.E+03	3.2.E+00	1.1.E-04	1.0.E-03
AE27	5.0.E+04	5.0.E+04	6.4.E+04	1.3.E+00	7.7.E+00	AU16	3.0.E+04	2.0.E+04	5.7.E+01	1.9.E-03	1.2.E-02
AE28	5.0.E+04	5.0.E+04	1.1.E+05	2.3.E+00	1.5.E+01	AU17	3.2.E+04	3.1.E+04	7.6.E+01	2.4.E-03	1.7.E-02
AE29	5.1.E+04	5.0.E+04	2.2.E+05	4.4.E+00	1.7.E+01	AU18	3.4.E+04	3.2.E+04	1.2.E+02	3.5.E-03	3.2.E-02
AE30	5.2.E+04	4.8.E+04	8.1.E+04	1.6.E+00	2.4.E+01	AU19	3.6.E+04	3.2.E+04	3.3.E+02	9.3.E-03	2.5.E-01

Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))	Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))
AE31	5.4.E+04	1.4.E+04	9.9.E+04	1.8.E+00	7.7.E+00	AU20	3.7.E+04	3.2.E+04	5.0.E+03	1.3.E-01	6.6.E-01
AE6	9.8.E+03	4.7.E+03	1.0.E+00	1.0.E-04	0.0.E+00	AU21	3.9.E+04	1.0.E+04	3.2.E+04	8.3.E-01	2.5.E+00
AE7	1.2.E+04	2.9.E+03	1.0.E+00	8.4.E-05	0.0.E+00	AU22	4.1.E+04	9.6.E+03	3.2.E+05	7.8.E+00	3.1.E+01
AE8	1.4.E+04	1.9.E+03	1.0.E+00	7.1.E-05	0.0.E+00	AU23	4.2.E+04	2.7.E+04	6.9.E+05	1.6.E+01	7.7.E+00
AE9	1.6.E+04	1.0.E+03	1.0.E+00	6.2.E-05	3.3.E-04	AU24	4.5.E+04	3.4.E+02	9.6.E+02	2.1.E-02	3.9.E-02
AF10	1.8.E+04	2.0.E+03	1.0.E+00	5.5.E-05	5.9.E-04	AU4	2.9.E+03	2.9.E+03	1.0.E+00	3.4.E-04	0.0.E+00
AF11	2.0.E+04	4.2.E+03	1.0.E+00	4.9.E-05	1.1.E-03	AU5	7.6.E+03	9.0.E+02	3.1.E+01	4.1.E-03	0.0.E+00
AF12	2.2.E+04	1.9.E+04	1.0.E+00	4.5.E-05	3.3.E-04	AV12	2.2.E+04	3.1.E+03	1.0.E+00	4.5.E-05	1.3.E-03
AF13	2.4.E+04	2.3.E+04	1.0.E+00	4.1.E-05	3.0.E-03	AV13	2.4.E+04	3.2.E+02	1.0.E+00	4.1.E-05	1.2.E-04
AF14	2.6.E+04	2.2.E+04	1.0.E+00	3.8.E-05	9.5.E-03	AV14	2.6.E+04	3.0.E-04	1.0.E+00	3.8.E-05	0.0.E+00
AF15	2.8.E+04	2.4.E+04	1.0.E+00	3.5.E-05	7.0.E-03	AV15	2.8.E+04	3.0.E-04	1.0.E+00	3.5.E-05	0.0.E+00
AF16	3.0.E+04	2.6.E+04	6.1.E+02	2.0.E-02	2.2.E-02	AV16	3.0.E+04	1.2.E+02	1.0.E+00	3.3.E-05	7.3.E-05
AF17	3.2.E+04	2.7.E+04	1.5.E+03	4.8.E-02	5.8.E-02	AV17	3.2.E+04	1.1.E+04	9.9.E+02	3.1.E-02	6.1.E-03
AF18	3.4.E+04	2.6.E+04	1.4.E+04	4.2.E-01	5.0.E+00	AV18	3.4.E+04	3.1.E+04	7.5.E+02	2.2.E-02	2.3.E-02
AF19	3.6.E+04	2.7.E+04	2.6.E+04	7.4.E-01	2.1.E+01	AV19	3.6.E+04	3.3.E+04	9.2.E+03	2.6.E-01	1.2.E-01
AF20	3.7.E+04	3.7.E+04	8.3.E+04	2.2.E+00	5.2.E+01	AV20	3.7.E+04	3.3.E+04	2.9.E+03	7.6.E-02	7.8.E-01
AF21	3.9.E+04	3.7.E+04	1.2.E+05	3.0.E+00	5.1.E+01	AV21	3.9.E+04	1.9.E+03	6.5.E+01	1.7.E-03	4.9.E-02
AF22	4.1.E+04	3.8.E+03	1.4.E+05	3.5.E+00	4.0.E+00	AV22	4.1.E+04	7.3.E+03	8.1.E+04	2.0.E+00	5.7.E+00
AF23	5.0.E+04	4.9.E+04	5.0.E+04	1.0.E+00	6.5.E+01	AV23	4.2.E+04	1.0.E+04	1.1.E+05	2.5.E+00	2.5.E+00
AF24	5.0.E+04	4.9.E+04	4.1.E+04	8.2.E-01	6.6.E+01	AV4	2.9.E+03	7.9.E+02	1.0.E+00	3.5.E-04	0.0.E+00
AF25	5.0.E+04	5.0.E+04	1.0.E+05	2.0.E+00	9.5.E+01	AV5	7.6.E+03	8.7.E-02	4.3.E+01	5.7.E-03	0.0.E+00
AF26	5.0.E+04	5.0.E+04	8.6.E+04	1.7.E+00	9.0.E+01	AW17	3.2.E+04	3.0.E-04	1.0.E+00	3.1.E-05	0.0.E+00
AF27	5.0.E+04	4.9.E+04	1.7.E+05	3.3.E+00	2.8.E+01	AW18	3.4.E+04	1.4.E+04	9.3.E+02	2.8.E-02	1.3.E-02
AF28	5.0.E+04	4.9.E+04	4.3.E+05	8.7.E+00	2.8.E+01	AW19	3.6.E+04	3.5.E+04	4.8.E+02	1.4.E-02	2.4.E-02
AF29	5.1.E+04	4.9.E+04	5.4.E+05	1.1.E+01	2.6.E+01	AW20	3.7.E+04	2.0.E+04	3.7.E+03	9.8.E-02	5.8.E-02
AF30	5.2.E+04	4.9.E+04	3.2.E+05	6.2.E+00	2.7.E+01	AW21	3.9.E+04	1.4.E+04	5.4.E+04	1.4.E+00	1.2.E-01
AF31	5.4.E+04	1.7.E+04	5.6.E+05	1.0.E+01	9.2.E+00	AW22	4.1.E+04	5.5.E+03	1.9.E+04	4.6.E-01	1.7.E-01
AF32	5.5.E+04	1.6.E+02	5.3.E+05	9.6.E+00	0.0.E+00	AW23	4.1.E+04	5.5.E+03	4.2.E+03	1.0.E-01	1.7.E-01
AF6	9.8.E+03	4.8.E+03	1.0.E+00	1.0.E-04	0.0.E+00	AX18	3.4.E+04	4.4.E+02	5.2.E+00	1.5.E-04	2.2.E-04
AF7	1.2.E+04	3.5.E+03	1.0.E+00	8.4.E-05	0.0.E+00	AX19	3.6.E+04	2.0.E+04	2.5.E+03	7.0.E-02	4.0.E-03
AF8	1.4.E+04	3.4.E+03	1.0.E+00	7.1.E-05	0.0.E+00	AX20	3.7.E+04	1.7.E+04	1.8.E+04	4.8.E-01	5.4.E-02
AF9	1.6.E+04	7.7.E+03	1.0.E+00	6.2.E-05	2.5.E-03	AX21	3.9.E+04	3.4.E+04	5.3.E+04	1.4.E+00	2.3.E-01
AG10	1.8.E+04	4.2.E+03	1.0.E+00	5.5.E-05	1.2.E-03	AX22	4.1.E+04	1.1.E+04	3.4.E+04	8.3.E-01	1.9.E-01
AG11	2.0.E+04	7.8.E+03	1.0.E+00	4.9.E-05	1.5.E-03	AY19	3.6.E+04	3.0.E-04	1.0.E+00	2.8.E-05	0.0.E+00
AG12	2.2.E+04	2.0.E+04	1.0.E+00	4.5.E-05	3.8.E-04	AY20	3.7.E+04	3.0.E-04	1.0.E+00	2.7.E-05	0.0.E+00
AG13	2.4.E+04	2.2.E+04	1.0.E+00	4.1.E-05	3.7.E-03	AY21	3.9.E+04	1.6.E+04	6.6.E+04	1.7.E+00	1.0.E-01
AG14	2.6.E+04	2.2.E+04	1.0.E+00	3.8.E-05	5.7.E-03	AY22	4.1.E+04	1.3.E+04	2.5.E+05	6.0.E+00	2.3.E-01
AG15	2.8.E+04	2.5.E+04	1.0.E+00	3.5.E-05	9.7.E-03	E12	2.2.E+04	7.2.E-03	2.4.E+01	1.1.E-03	0.0.E+00
AG16	3.0.E+04	2.8.E+04	3.2.E+02	1.1.E-02	1.0.E-02	E13	2.4.E+04	2.0.E-02	6.7.E+01	2.7.E-03	0.0.E+00
AG17	3.2.E+04	2.9.E+04	2.0.E+03	6.2.E-02	2.3.E-01	E14	2.6.E+04	5.8.E-02	1.9.E+02	7.4.E-03	0.0.E+00
AG18	3.4.E+04	3.1.E+04	2.2.E+04	6.6.E-01	1.4.E+00	E15	2.8.E+04	3.0.E-04	1.0.E+00	3.5.E-05	0.0.E+00
AG19	5.0.E+04	4.5.E+04	4.9.E+03	9.8.E-02	1.0.E+01	E16	3.0.E+04	3.0.E-04	1.0.E+00	3.3.E-05	0.0.E+00
AG20	3.7.E+04	3.4.E+04	5.2.E+04	1.4.E+00	2.7.E+01	F11	2.0.E+04	8.3.E+03	8.1.E+01	4.0.E-03	1.5.E-05

Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))	Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))
AG21	4.1.E+04	4.0.E+04	1.9.E+05	4.6.E+00	3.8.E+01	F12	2.2.E+04	3.7.E+03	2.3.E+02	1.0.E-02	2.3.E-07
AG22	4.1.E+04	1.6.E+04	8.8.E+04	2.2.E+00	1.4.E+01	F13	2.4.E+04	2.3.E+04	4.9.E+03	2.1.E-01	2.2.E-06
AG23	4.2.E+04	4.6.E+02	1.1.E+05	2.6.E+00	3.5.E-01	F14	2.6.E+04	2.2.E+03	2.5.E+02	9.4.E-03	6.7.E-10
AG24	5.0.E+04	5.0.E+04	1.2.E+05	2.5.E+00	7.7.E+01	F15	2.8.E+04	1.1.E+04	1.3.E+03	4.7.E-02	3.7.E-07
AG25	5.0.E+04	5.0.E+04	3.0.E+05	5.9.E+00	1.4.E+02	F16	3.0.E+04	1.6.E+03	1.9.E+02	6.3.E-03	1.1.E-07
AG26	5.0.E+04	5.0.E+04	3.1.E+05	6.3.E+00	9.6.E+01	F17	3.2.E+04	3.0.E-04	1.0.E+00	3.1.E-05	0.0.E+00
AG27	5.0.E+04	4.9.E+04	2.5.E+05	5.0.E+00	5.9.E+01	G10	1.8.E+04	1.5.E+01	1.0.E+00	5.5.E-05	2.3.E-11
AG28	5.0.E+04	4.9.E+04	1.1.E+06	2.1.E+01	3.5.E+01	G11	3.0.E+04	2.6.E+04	3.8.E+02	1.2.E-02	1.4.E-05
AG29	5.1.E+04	4.9.E+04	7.0.E+05	1.4.E+01	2.9.E+01	G12	2.7.E+04	2.3.E+04	9.8.E+02	3.6.E-02	2.4.E-05
AG30	5.2.E+04	4.9.E+04	1.7.E+06	3.2.E+01	2.8.E+01	G13	3.6.E+04	3.1.E+04	2.2.E+03	6.0.E-02	2.2.E-06
AG31	5.4.E+04	4.9.E+04	2.1.E+06	3.9.E+01	2.7.E+01	G14	2.6.E+04	2.4.E+04	2.6.E+03	1.0.E-01	2.7.E-07
AG32	5.5.E+04	3.5.E+04	1.8.E+06	3.4.E+01	7.1.E+00	G15	4.6.E+04	4.0.E+04	4.4.E+03	9.5.E-02	1.1.E-06
AG33	5.6.E+04	4.0.E+02	1.3.E+06	2.4.E+01	0.0.E+00	G16	3.0.E+04	2.1.E+03	2.4.E+02	8.1.E-03	1.5.E-07
AG6	9.8.E+03	2.5.E+03	1.0.E+00	1.0.E-04	0.0.E+00	G17	3.2.E+04	3.0.E-04	1.0.E+00	3.1.E-05	0.0.E+00
AG7	1.2.E+04	4.4.E+03	1.0.E+00	8.4.E-05	0.0.E+00	G18	3.4.E+04	2.5.E-01	8.4.E+02	2.5.E-02	0.0.E+00
AG8	1.4.E+04	7.3.E+03	2.1.E+00	1.5.E-04	1.0.E-04	H10	1.8.E+04	6.6.E+03	5.8.E+01	3.2.E-03	6.8.E-06
AG9	1.6.E+04	1.6.E+04	2.1.E+00	1.3.E-04	5.1.E-03	H11	5.0.E+04	4.3.E+04	5.8.E+02	1.2.E-02	1.2.E-05
AH10	1.8.E+04	9.7.E+03	1.0.E+00	5.5.E-05	3.6.E-03	H12	4.5.E+04	3.9.E+04	1.6.E+03	3.5.E-02	3.1.E-05
AH11	2.0.E+04	1.1.E+04	9.7.E+02	4.8.E-02	2.5.E-03	H13	3.9.E+04	3.3.E+04	2.0.E+03	5.1.E-02	1.8.E-06
AH12	2.2.E+04	1.9.E+04	1.0.E+00	4.5.E-05	9.8.E-04	H14	4.2.E+04	3.6.E+04	2.3.E+03	5.4.E-02	6.9.E-07
AH13	2.4.E+04	2.3.E+04	1.7.E+03	7.0.E-02	1.2.E-03	H15	4.9.E+04	4.2.E+04	1.1.E+04	2.2.E-01	1.5.E-06
AH14	2.6.E+04	2.3.E+04	1.0.E+00	3.8.E-05	2.4.E-03	H16	3.0.E+04	2.0.E+04	2.0.E+03	6.8.E-02	1.0.E-06
AH15	2.8.E+04	2.5.E+04	1.0.E+00	3.5.E-05	6.4.E-03	H17	3.2.E+04	1.5.E+03	1.8.E+02	5.6.E-03	1.7.E-06
AH16	3.0.E+04	2.8.E+04	3.0.E+01	9.9.E-04	1.1.E-02	H18	3.4.E+04	4.5.E-01	1.5.E+03	4.4.E-02	0.0.E+00
AH17	3.2.E+04	2.9.E+04	2.1.E+03	6.6.E-02	2.5.E-01	I10	1.8.E+04	1.7.E+04	1.5.E+02	8.2.E-03	3.6.E-05
AH18	3.4.E+04	3.1.E+04	2.4.E+03	7.0.E-02	3.1.E-01	I11	5.0.E+04	4.3.E+04	5.6.E+02	1.1.E-02	3.1.E-05
AH19	3.9.E+04	3.6.E+04	4.2.E+03	1.1.E-01	2.3.E+00	I12	4.7.E+04	4.0.E+04	1.9.E+03	4.0.E-02	3.3.E-05
AH20	4.4.E+04	4.1.E+04	9.1.E+04	2.1.E+00	1.1.E+01	I13	5.0.E+04	4.4.E+04	1.3.E+03	2.5.E-02	2.4.E-05
AH21	3.9.E+04	3.9.E+04	8.0.E+05	2.0.E+01	3.0.E+01	I14	4.9.E+04	4.2.E+04	4.9.E+02	1.0.E-02	1.9.E-07
AH22	4.1.E+04	3.5.E+04	4.6.E+05	1.1.E+01	3.0.E+01	I15	5.0.E+04	4.3.E+04	1.6.E+03	3.3.E-02	8.0.E-06
AH23	4.2.E+04	4.4.E+03	3.3.E+05	7.7.E+00	3.5.E+00	I16	3.0.E+04	2.8.E+04	3.8.E+03	1.3.E-01	3.1.E-07
AH24	5.0.E+04	4.9.E+04	7.0.E+05	1.4.E+01	6.1.E+01	I17	3.2.E+04	1.8.E+04	5.8.E+02	1.8.E-02	1.7.E-05
AH25	5.0.E+04	5.0.E+04	1.4.E+06	2.7.E+01	9.4.E+01	I18	3.4.E+04	4.9.E-02	1.6.E+02	4.9.E-03	0.0.E+00
AH26	5.0.E+04	5.0.E+04	7.2.E+05	1.4.E+01	8.5.E+01	J10	2.7.E+04	2.3.E+04	5.2.E+03	1.9.E-01	3.3.E-05
AH27	5.1.E+04	5.0.E+04	1.7.E+06	3.3.E+01	6.5.E+01	J11	5.1.E+04	4.4.E+04	4.6.E+02	9.1.E-03	3.3.E-05
AH28	5.0.E+04	5.0.E+04	1.2.E+06	2.3.E+01	3.6.E+01	J12	4.8.E+04	4.1.E+04	1.6.E+03	3.4.E-02	3.3.E-05
AH29	5.3.E+04	5.0.E+04	6.2.E+06	1.2.E+02	3.5.E+01	J13	5.1.E+04	4.4.E+04	8.0.E+02	1.6.E-02	3.2.E-05
AH30	5.2.E+04	4.9.E+04	1.8.E+06	3.4.E+01	2.8.E+01	J14	4.9.E+04	4.2.E+04	4.4.E+02	9.1.E-03	1.7.E-05
AH31	5.5.E+04	3.7.E+04	4.4.E+06	8.1.E+01	2.0.E+01	J15	5.0.E+04	4.3.E+04	1.3.E+03	2.6.E-02	2.3.E-05
AH32	5.5.E+04	2.7.E+03	3.6.E+05	6.6.E+00	2.1.E-01	J16	4.8.E+04	4.2.E+04	2.3.E+03	4.8.E-02	2.4.E-05
AH33	5.6.E+04	9.1.E+01	3.0.E+05	5.4.E+00	0.0.E+00	J17	3.2.E+04	1.5.E+04	2.7.E+02	8.5.E-03	1.4.E-05
AH5	7.6.E+03	1.5.E+03	1.0.E+00	1.3.E-04	0.0.E+00	J18	3.4.E+04	3.0.E-04	1.0.E+00	3.0.E-05	0.0.E+00
AH6	9.8.E+03	4.9.E+03	1.0.E+00	1.0.E-04	0.0.E+00	K10	2.2.E+04	1.9.E+04	1.5.E+02	6.9.E-03	3.3.E-05

Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))	Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))
AH7	1.2.E+04	4.5.E+03	1.0.E+00	8.4.E-05	0.0.E+00	K11	5.1.E+04	4.4.E+04	4.0.E+02	7.8.E-03	3.3.E-05
AH8	1.4.E+04	3.8.E+03	2.8.E+01	2.0.E-03	6.3.E-06	K12	4.9.E+04	4.2.E+04	5.0.E+02	1.0.E-02	3.3.E-05
AH9	1.6.E+04	7.1.E+03	1.0.E+00	6.2.E-05	2.3.E-03	K13	5.0.E+04	4.3.E+04	3.3.E+02	6.6.E-03	3.3.E-05
AI10	1.8.E+04	1.5.E+04	1.0.E+00	5.5.E-05	5.7.E-03	K14	4.9.E+04	4.2.E+04	4.2.E+02	8.6.E-03	3.3.E-05
AI11	2.0.E+04	1.5.E+04	7.1.E+02	3.5.E-02	5.1.E-03	K15	4.6.E+04	3.9.E+04	5.5.E+02	1.2.E-02	2.5.E-05
AI12	2.2.E+04	2.2.E+04	1.0.E+00	4.5.E-05	2.1.E-03	K16	3.0.E+04	2.7.E+04	5.6.E+02	1.9.E-02	1.4.E-05
AI13	2.4.E+04	2.3.E+04	1.0.E+00	4.1.E-05	2.6.E-04	K17	3.2.E+04	1.3.E+04	3.4.E+03	1.1.E-01	3.0.E-08
AI14	2.6.E+04	2.2.E+04	3.7.E+02	1.4.E-02	9.9.E-04	L10	1.8.E+04	1.0.E+04	6.7.E+01	3.7.E-03	2.1.E-05
AI15	2.8.E+04	8.3.E+03	1.9.E+03	6.8.E-02	8.7.E-04	L11	5.0.E+04	4.3.E+04	3.5.E+02	7.0.E-03	3.3.E-05
AI16	3.0.E+04	1.0.E+04	9.9.E+02	3.3.E-02	9.1.E-03	L12	4.9.E+04	4.2.E+04	3.0.E+02	6.2.E-03	3.3.E-05
AI17	3.2.E+04	2.8.E+04	1.5.E+03	4.8.E-02	2.8.E-01	L13	4.9.E+04	4.2.E+04	5.8.E+02	1.2.E-02	3.3.E-05
AI18	3.4.E+04	3.2.E+04	2.6.E+03	7.7.E-02	6.0.E-02	L14	5.0.E+04	4.3.E+04	2.3.E+03	4.7.E-02	3.3.E-05
AI19	3.6.E+04	3.4.E+04	8.5.E+03	2.4.E-01	5.2.E-01	L15	3.8.E+04	3.3.E+04	4.0.E+04	1.1.E+00	2.4.E-05
AI20	3.7.E+04	3.4.E+04	8.7.E+03	2.3.E-01	2.4.E+00	L16	3.0.E+04	1.3.E+04	1.4.E+04	4.8.E-01	1.1.E-05
AI21	3.9.E+04	3.4.E+04	6.5.E+04	1.7.E+00	1.3.E+01	L17	3.2.E+04	5.9.E+02	1.1.E+04	3.5.E-01	1.4.E-09
AI22	4.1.E+04	7.1.E+03	3.1.E+05	7.5.E+00	3.6.E+00	M10	1.8.E+04	8.1.E+03	1.7.E+01	9.5.E-04	1.7.E-05
AI23	5.1.E+04	4.6.E+04	3.8.E+06	7.4.E+01	1.8.E+02	M11	5.0.E+04	4.3.E+04	9.4.E+01	1.9.E-03	3.3.E-05
AI24	5.0.E+04	4.9.E+04	6.4.E+05	1.3.E+01	1.9.E+02	M12	4.9.E+04	4.2.E+04	3.2.E+02	6.6.E-03	3.3.E-05
AI25	5.0.E+04	5.0.E+04	8.6.E+05	1.7.E+01	9.1.E+01	M13	4.9.E+04	4.2.E+04	5.9.E+04	1.2.E+00	3.3.E-05
AI26	5.1.E+04	5.0.E+04	2.3.E+06	4.6.E+01	1.0.E+02	M14	5.0.E+04	4.3.E+04	2.0.E+03	4.1.E-02	3.3.E-05
AI27	5.1.E+04	5.0.E+04	1.4.E+06	2.8.E+01	6.4.E+01	M15	4.0.E+04	3.5.E+04	3.4.E+05	8.6.E+00	2.3.E-05
AI28	5.0.E+04	4.9.E+04	8.5.E+05	1.7.E+01	5.5.E+01	M16	3.0.E+04	1.9.E+03	5.3.E+02	1.8.E-02	3.2.E-09
AI29	5.1.E+04	4.7.E+04	1.3.E+06	2.4.E+01	7.6.E+01	M17	3.2.E+04	3.0.E-04	1.0.E+00	3.1.E-05	0.0.E+00
AI30	5.2.E+04	4.6.E+04	1.1.E+06	2.1.E+01	8.1.E+01	N10	1.8.E+04	2.8.E+03	5.8.E+00	3.2.E-04	5.8.E-06
AI31	5.4.E+04	7.9.E+03	3.9.E+05	7.3.E+00	1.3.E+01	N11	2.0.E+04	1.6.E+02	1.1.E+02	5.6.E-03	4.6.E-05
AI32	5.5.E+04	3.0.E-04	1.0.E+00	1.8.E-05	0.0.E+00	N12	2.2.E+04	3.6.E+02	3.4.E+02	1.5.E-02	9.3.E-05
AI5	7.6.E+03	5.6.E+03	1.6.E+00	2.1.E-04	0.0.E+00	N13	4.9.E+04	4.3.E+04	#####	#####	3.3.E-05
AI6	9.8.E+03	5.3.E+03	1.5.E+00	1.6.E-04	0.0.E+00	N14	4.9.E+04	4.3.E+04	2.5.E+03	5.1.E-02	2.2.E-05
AI7	1.2.E+04	5.3.E+03	1.6.E+00	1.3.E-04	0.0.E+00	N15	3.4.E+04	2.9.E+04	3.7.E+03	1.1.E-01	1.5.E-06
AI8	1.4.E+04	5.5.E+03	2.1.E+02	1.5.E-02	3.8.E-04	N16	3.0.E+04	7.9.E+01	2.7.E+01	9.0.E-04	1.3.E-10
AI9	1.6.E+04	1.4.E+04	3.7.E+00	2.3.E-04	4.4.E-03	N17	3.2.E+04	3.0.E-04	1.0.E+00	3.1.E-05	0.0.E+00
AJ10	1.8.E+04	1.3.E+03	1.0.E+00	5.5.E-05	5.1.E-04	O10	1.8.E+04	1.4.E+02	2.0.E+00	1.1.E-04	4.1.E-05
AJ11	2.0.E+04	1.5.E+04	1.0.E+00	4.9.E-05	4.9.E-03	O11	2.0.E+04	1.2.E+04	1.4.E+02	7.1.E-03	3.5.E-03
AJ12	2.4.E+04	2.3.E+04	1.0.E+00	4.1.E-05	3.6.E-03	O12	2.2.E+04	2.1.E+04	3.5.E+02	1.6.E-02	5.3.E-03
AJ13	2.4.E+04	2.4.E+04	1.0.E+00	4.1.E-05	9.3.E-04	O13	2.4.E+04	1.4.E+04	3.3.E+03	1.4.E-01	6.1.E-03
AJ14	2.6.E+04	1.1.E+04	2.9.E+03	1.1.E-01	3.8.E-04	O14	2.6.E+04	6.1.E+03	1.8.E+03	7.0.E-02	1.1.E-03
AJ15	2.8.E+04	8.5.E-01	1.0.E+00	3.5.E-05	4.8.E-08	O15	2.8.E+04	1.1.E+04	2.5.E+03	8.8.E-02	1.5.E-05
AJ16	3.0.E+04	3.0.E-04	1.0.E+00	3.3.E-05	0.0.E+00	O16	3.0.E+04	1.2.E+02	1.1.E+01	3.7.E-04	4.9.E-10
AJ17	3.2.E+04	1.9.E+04	2.5.E+01	7.8.E-04	2.3.E-02	O17	3.2.E+04	3.0.E-04	1.0.E+00	3.1.E-05	0.0.E+00
AJ18	3.4.E+04	3.2.E+04	1.3.E+03	3.9.E-02	9.0.E-02	P10	1.8.E+04	3.0.E-04	1.0.E+00	5.5.E-05	0.0.E+00
AJ19	3.6.E+04	3.4.E+04	2.2.E+03	6.3.E-02	1.1.E-01	P11	2.0.E+04	1.8.E+04	1.1.E+02	5.3.E-03	5.0.E-03
AJ20	3.7.E+04	3.4.E+04	6.7.E+03	1.8.E-01	4.1.E-01	P12	2.4.E+04	2.3.E+04	2.5.E+02	1.0.E-02	5.6.E-03
AJ21	3.9.E+04	3.4.E+04	1.0.E+04	2.7.E-01	7.9.E+00	P13	2.6.E+04	2.5.E+04	7.7.E+02	2.9.E-02	1.0.E-02

Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))	Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))
AJ22	4.1.E+04	7.9.E+03	2.9.E+05	7.2.E+00	2.0.E+00	P14	2.6.E+04	2.6.E+04	1.1.E+03	4.2.E-02	4.5.E-03
AJ23	5.0.E+04	4.3.E+04	7.0.E+05	1.4.E+01	1.4.E+02	P15	2.8.E+04	2.6.E+04	1.6.E+03	5.8.E-02	2.2.E-03
AJ24	5.0.E+04	4.7.E+04	9.6.E+05	1.9.E+01	1.1.E+02	P16	3.0.E+04	2.1.E+03	2.6.E+02	8.6.E-03	5.5.E-04
AJ25	5.1.E+04	4.9.E+04	1.2.E+06	2.4.E+01	1.7.E+02	P17	3.2.E+04	3.0.E-04	1.0.E+00	3.1.E-05	0.0.E+00
AJ26	5.1.E+04	5.0.E+04	3.1.E+06	6.1.E+01	1.4.E+02	Q10	1.8.E+04	3.0.E-04	1.0.E+00	5.5.E-05	0.0.E+00
AJ27	5.0.E+04	4.9.E+04	6.2.E+05	1.2.E+01	6.0.E+01	Q11	2.0.E+04	1.4.E+04	5.7.E+01	2.8.E-03	4.0.E-03
AJ28	5.0.E+04	4.9.E+04	1.5.E+06	3.1.E+01	5.0.E+01	Q12	2.4.E+04	2.3.E+04	3.4.E+02	1.4.E-02	5.6.E-03
AJ29	5.1.E+04	4.7.E+04	1.1.E+06	2.2.E+01	8.1.E+01	Q13	2.6.E+04	2.5.E+04	1.3.E+03	4.8.E-02	7.4.E-03
AJ30	5.3.E+04	4.5.E+04	2.1.E+06	4.0.E+01	1.1.E+02	Q14	2.8.E+04	2.7.E+04	1.1.E+02	3.8.E-03	5.7.E-03
AJ31	5.4.E+04	1.2.E+04	1.1.E+06	2.0.E+01	1.6.E+01	Q15	3.0.E+04	2.9.E+04	6.7.E+02	2.2.E-02	5.6.E-03
AJ32	5.5.E+04	3.0.E-04	1.0.E+00	1.8.E-05	0.0.E+00	Q16	3.0.E+04	1.2.E+04	9.4.E+02	3.1.E-02	3.5.E-03
AJ5	7.6.E+03	5.4.E+03	1.6.E+00	2.1.E-04	0.0.E+00	Q17	3.2.E+04	3.0.E-04	1.0.E+00	3.1.E-05	0.0.E+00
AJ6	1.0.E+04	1.0.E+04	2.9.E+00	2.8.E-04	0.0.E+00	R10	1.8.E+04	3.0.E-04	1.0.E+00	5.5.E-05	0.0.E+00
AJ7	1.2.E+04	5.5.E+03	1.6.E+00	1.3.E-04	0.0.E+00	R11	2.0.E+04	1.2.E+04	4.5.E+02	2.2.E-02	3.4.E-03
AJ8	1.4.E+04	9.3.E+03	2.7.E+00	1.9.E-04	5.0.E-05	R12	2.4.E+04	2.3.E+04	6.8.E+01	2.8.E-03	5.4.E-03
AJ9	1.6.E+04	5.9.E+03	1.8.E+00	1.1.E-04	1.9.E-03	R13	2.6.E+04	2.5.E+04	9.9.E+01	3.7.E-03	5.5.E-03
AK10	1.8.E+04	9.9.E+03	3.5.E+00	1.9.E-04	5.0.E-03	R14	2.8.E+04	2.7.E+04	1.1.E+03	3.9.E-02	4.0.E-03
AK11	2.0.E+04	8.9.E+03	5.5.E+02	2.7.E-02	3.0.E-03	R15	3.0.E+04	2.8.E+04	1.4.E+04	4.8.E-01	5.0.E-03
AK12	2.2.E+04	2.1.E+04	2.0.E+00	8.9.E-05	6.2.E-03	R16	3.0.E+04	2.5.E+04	4.9.E+03	1.6.E-01	1.4.E-01
AK13	2.4.E+04	2.0.E+04	1.0.E+00	4.1.E-05	1.2.E-03	R17	3.2.E+04	6.5.E+03	8.7.E+03	2.7.E-01	4.7.E-02
AK14	2.6.E+04	1.3.E+02	1.0.E+00	3.8.E-05	6.8.E-06	R18	3.4.E+04	3.0.E-04	1.0.E+00	3.0.E-05	0.0.E+00
AK15	2.8.E+04	3.0.E-04	1.0.E+00	3.5.E-05	0.0.E+00	S10	1.8.E+04	3.0.E-04	1.0.E+00	5.5.E-05	0.0.E+00
AK16	3.0.E+04	3.0.E-04	1.0.E+00	3.3.E-05	0.0.E+00	S11	2.5.E+04	2.3.E+04	3.8.E+03	1.5.E-01	1.2.E-02
AK17	3.2.E+04	1.1.E+04	3.9.E+02	1.2.E-02	1.1.E-02	S12	3.9.E+04	3.6.E+04	9.2.E+02	2.4.E-02	8.1.E-03
AK18	3.4.E+04	3.3.E+04	1.0.E+03	3.0.E-02	1.1.E-01	S13	2.6.E+04	2.5.E+04	9.7.E+01	3.7.E-03	5.3.E-03
AK19	3.6.E+04	3.5.E+04	2.9.E+03	8.1.E-02	2.2.E-02	S14	2.8.E+04	2.7.E+04	1.2.E+03	4.4.E-02	3.4.E-03
AK20	3.7.E+04	3.5.E+04	3.1.E+03	8.3.E-02	4.2.E-01	S15	2.8.E+04	2.8.E+04	6.5.E+03	2.3.E-01	4.1.E-03
AK21	3.9.E+04	2.7.E+04	1.3.E+05	3.2.E+00	1.6.E+00	S16	3.0.E+04	3.0.E+04	3.3.E+04	1.1.E+00	1.4.E-01
AK22	4.1.E+04	1.2.E+03	1.2.E+05	2.8.E+00	3.0.E-01	S17	3.2.E+04	2.0.E+04	7.7.E+03	2.4.E-01	1.1.E-01
AK23	4.2.E+04	3.8.E+04	1.3.E+06	3.1.E+01	1.2.E+02	S18	3.4.E+04	6.3.E+03	3.2.E+03	9.6.E-02	2.2.E-02
AK24	4.6.E+04	4.0.E+04	5.9.E+06	1.3.E+02	1.5.E+02	S19	3.6.E+04	2.3.E+03	4.4.E+02	1.2.E-02	1.4.E-02
AK25	5.2.E+04	5.0.E+04	7.7.E+06	1.5.E+02	2.0.E+02	T10	1.8.E+04	3.2.E+02	1.0.E+00	5.5.E-05	4.4.E-04
AK26	5.1.E+04	4.9.E+04	1.4.E+06	2.7.E+01	2.2.E+02	T11	2.0.E+04	1.9.E+04	1.0.E+00	4.9.E-05	2.3.E-02
AK27	5.0.E+04	4.9.E+04	1.2.E+06	2.5.E+01	4.9.E+01	T12	2.2.E+04	2.2.E+04	1.0.E+00	4.5.E-05	2.0.E-02
AK28	5.0.E+04	4.9.E+04	1.9.E+06	3.8.E+01	3.4.E+01	T13	2.4.E+04	2.4.E+04	1.9.E+01	7.9.E-04	1.3.E-02
AK29	5.1.E+04	4.8.E+04	8.7.E+05	1.7.E+01	3.3.E+01	T14	2.8.E+04	2.7.E+04	1.0.E+02	3.7.E-03	3.4.E-03
AK30	5.3.E+04	4.2.E+04	1.7.E+06	3.2.E+01	1.8.E+01	T15	3.0.E+04	2.9.E+04	4.4.E+02	1.5.E-02	2.9.E-03
AK31	5.4.E+04	3.3.E+03	1.1.E+05	2.0.E+00	3.2.E+00	T16	3.2.E+04	3.1.E+04	3.2.E+02	1.0.E-02	8.8.E-02
AK32	5.5.E+04	3.0.E-04	1.0.E+00	1.8.E-05	0.0.E+00	T17	3.3.E+04	3.3.E+04	2.9.E+03	8.9.E-02	1.1.E-01
AK4	2.9.E+03	1.6.E+02	1.0.E+00	3.5.E-04	0.0.E+00	T18	3.4.E+04	1.7.E+04	3.5.E+04	1.0.E+00	5.8.E-02
AK5	7.6.E+03	6.6.E+03	2.0.E+00	2.6.E-04	0.0.E+00	T19	3.6.E+04	8.6.E+03	2.7.E+03	7.6.E-02	4.9.E-02
AK6	9.8.E+03	9.1.E+03	2.7.E+00	2.7.E-04	0.0.E+00	T20	3.7.E+04	1.2.E-03	4.2.E+00	1.1.E-04	0.0.E+00
AK7	1.2.E+04	4.7.E+03	1.0.E+00	8.4.E-05	0.0.E+00	U10	1.8.E+04	1.5.E+03	1.0.E+00	5.5.E-05	1.9.E-03

Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))	Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))
AK8	1.4.E+04	8.5.E+03	2.4.E+00	1.7.E-04	8.2.E-06	U11	2.0.E+04	1.9.E+04	1.0.E+00	4.9.E-05	2.2.E-02
AK9	1.6.E+04	1.0.E+04	3.6.E+00	2.2.E-04	5.7.E-03	U12	2.2.E+04	2.2.E+04	5.5.E+02	2.5.E-02	2.1.E-02
AL10	1.8.E+04	1.5.E+04	5.8.E+00	3.2.E-04	7.5.E-03	U13	2.4.E+04	2.4.E+04	1.0.E+00	4.1.E-05	7.4.E-03
AL11	2.0.E+04	4.4.E+03	1.0.E+00	4.9.E-05	1.5.E-03	U14	2.6.E+04	2.6.E+04	2.1.E+01	8.0.E-04	6.4.E-03
AL12	2.2.E+04	1.7.E+04	6.9.E+02	3.1.E-02	5.6.E-03	U15	3.0.E+04	2.9.E+04	1.2.E+03	4.1.E-02	1.9.E-02
AL13	2.4.E+04	1.5.E+04	1.0.E+00	4.1.E-05	3.6.E-03	U16	3.2.E+04	3.1.E+04	4.3.E+02	1.3.E-02	6.7.E-02
AL14	2.6.E+04	5.6.E+03	1.0.E+00	3.8.E-05	1.5.E-03	U17	3.4.E+04	3.3.E+04	6.9.E+02	2.0.E-02	1.2.E-01
AL15	2.8.E+04	3.0.E-04	1.0.E+00	3.5.E-05	0.0.E+00	U18	3.5.E+04	3.5.E+04	3.4.E+04	9.6.E-01	1.4.E-01
AL16	3.0.E+04	3.0.E-04	1.0.E+00	3.3.E-05	0.0.E+00	U19	3.6.E+04	2.4.E+04	7.2.E+03	2.0.E-01	9.6.E-02
AL17	3.2.E+04	5.7.E+00	1.0.E+00	3.1.E-05	1.6.E-05	U20	3.7.E+04	5.6.E+03	1.7.E+03	4.4.E-02	1.1.E-01
AL18	3.4.E+04	2.8.E+04	9.4.E+02	2.8.E-02	8.8.E-03	U21	3.9.E+04	3.0.E-04	1.0.E+00	2.6.E-05	0.0.E+00
AL19	3.6.E+04	3.5.E+04	1.3.E+03	3.7.E-02	2.2.E-02	V10	1.8.E+04	7.0.E+02	3.7.E+01	2.0.E-03	7.1.E-04
AL20	3.7.E+04	3.6.E+04	2.5.E+03	6.6.E-02	2.2.E-01	V11	2.0.E+04	1.7.E+04	1.0.E+00	4.9.E-05	1.1.E-02
AL21	3.9.E+04	3.1.E+04	2.1.E+04	5.3.E-01	2.5.E+00	V12	2.2.E+04	2.1.E+04	7.7.E+01	3.5.E-03	2.3.E-02
AL22	4.1.E+04	1.6.E+03	6.5.E+04	1.6.E+00	4.2.E-01	V13	2.4.E+04	2.3.E+04	1.1.E+03	4.4.E-02	1.1.E-02
AL23	4.2.E+04	1.2.E+04	3.4.E+05	8.1.E+00	4.1.E+01	V14	2.6.E+04	2.6.E+04	5.6.E+00	2.1.E-04	6.4.E-03
AL24	4.6.E+04	1.8.E+04	2.0.E+06	4.3.E+01	6.6.E+01	V15	3.0.E+04	2.8.E+04	5.7.E+01	1.9.E-03	7.0.E-02
AL25	4.7.E+04	4.4.E+04	3.2.E+06	6.7.E+01	1.2.E+02	V16	3.2.E+04	3.1.E+04	3.3.E+02	1.0.E-02	1.4.E-01
AL26	5.1.E+04	5.0.E+04	3.7.E+06	7.2.E+01	1.9.E+02	V17	3.4.E+04	3.3.E+04	2.9.E+03	8.7.E-02	1.4.E-01
AL27	5.1.E+04	5.0.E+04	2.3.E+06	4.5.E+01	5.9.E+01	V18	3.6.E+04	3.4.E+04	2.4.E+04	6.7.E-01	2.1.E-01
AL28	5.0.E+04	4.9.E+04	2.4.E+06	4.8.E+01	2.4.E+01	V19	3.6.E+04	3.5.E+04	5.5.E+03	1.6.E-01	2.5.E-01
AL29	5.2.E+04	4.9.E+04	2.3.E+06	4.6.E+01	2.3.E+01	V20	3.7.E+04	3.4.E+04	3.3.E+04	9.0.E-01	6.5.E-01
AL30	5.3.E+04	4.0.E+04	1.4.E+06	2.7.E+01	6.1.E+01	V21	3.9.E+04	1.2.E+04	5.2.E+04	1.3.E+00	3.3.E-01
AL31	5.4.E+04	1.1.E+03	1.5.E+04	2.8.E-01	1.3.E+00	V22	4.1.E+04	3.0.E-04	1.0.E+00	2.5.E-05	0.0.E+00
AL32	5.5.E+04	3.0.E-04	1.0.E+00	1.8.E-05	0.0.E+00	V23	4.2.E+04	3.0.E-04	1.0.E+00	2.4.E-05	0.0.E+00
AL4	2.9.E+03	8.1.E+02	1.0.E+00	3.5.E-04	0.0.E+00	V24	4.5.E+04	3.0.E-04	1.0.E+00	2.2.E-05	0.0.E+00
AL5	7.6.E+03	6.8.E+03	2.3.E+00	3.0.E-04	0.0.E+00	V25	4.7.E+04	3.0.E-04	1.0.E+00	2.1.E-05	0.0.E+00
AL6	9.8.E+03	8.5.E+03	2.4.E+00	2.5.E-04	0.0.E+00	V26	4.8.E+04	3.0.E-04	1.0.E+00	2.1.E-05	0.0.E+00
AL7	1.2.E+04	9.3.E+03	2.7.E+00	2.3.E-04	0.0.E+00	V9	1.6.E+04	1.1.E+03	6.9.E+01	4.2.E-03	3.5.E-04
AL8	1.4.E+04	8.2.E+03	2.4.E+00	1.7.E-04	0.0.E+00	W10	1.8.E+04	5.1.E+03	4.7.E+00	2.6.E-04	1.4.E-03
AL9	1.6.E+04	1.0.E+04	7.6.E+02	4.7.E-02	5.7.E-03	W11	2.0.E+04	1.8.E+04	2.4.E+02	1.2.E-02	2.8.E-03
AM10	1.8.E+04	1.7.E+04	6.3.E+00	3.5.E-04	7.4.E-03	W12	2.2.E+04	2.0.E+04	1.0.E+00	4.5.E-05	1.9.E-02
AM11	2.0.E+04	1.9.E+04	6.2.E+00	3.0.E-04	7.6.E-03	W13	2.4.E+04	2.1.E+04	5.3.E+02	2.2.E-02	1.4.E-02
AM12	2.2.E+04	1.8.E+04	5.5.E+00	2.5.E-04	7.2.E-03	W14	2.6.E+04	2.6.E+04	1.6.E+02	6.2.E-03	5.5.E-03
AM13	2.4.E+04	1.6.E+04	8.2.E+02	3.4.E-02	4.9.E-03	W15	2.8.E+04	2.8.E+04	5.8.E+02	2.0.E-02	1.1.E-01
AM14	2.6.E+04	8.3.E+03	1.0.E+00	3.8.E-05	2.2.E-03	W16	3.2.E+04	3.1.E+04	5.1.E+03	1.6.E-01	5.5.E-01
AM15	2.8.E+04	3.0.E-04	1.0.E+00	3.5.E-05	0.0.E+00	W17	3.4.E+04	3.3.E+04	3.9.E+03	1.2.E-01	2.2.E+00
AM16	3.0.E+04	3.0.E-04	1.0.E+00	3.3.E-05	0.0.E+00	W18	3.6.E+04	3.4.E+04	3.3.E+04	9.3.E-01	5.1.E-01
AM17	3.2.E+04	3.0.E-04	1.0.E+00	3.1.E-05	0.0.E+00	W19	3.7.E+04	3.6.E+04	8.3.E+04	2.2.E+00	9.0.E-01
AM18	3.4.E+04	2.1.E+04	5.1.E+02	1.5.E-02	2.9.E-03	W20	3.8.E+04	3.7.E+04	2.6.E+04	6.7.E-01	1.9.E+00
AM19	3.7.E+04	3.6.E+04	8.8.E+02	2.3.E-02	5.8.E-02	W21	4.0.E+04	2.1.E+04	3.0.E+06	7.6.E+01	6.7.E-01
AM20	3.9.E+04	3.8.E+04	1.0.E+03	2.6.E-02	3.4.E-01	W22	4.0.E+04	3.0.E+04	1.1.E+06	2.6.E+01	3.4.E+01
AM21	4.1.E+04	3.9.E+04	1.6.E+04	4.0.E-01	1.1.E+00	W23	4.2.E+04	3.0.E+04	2.7.E+06	6.3.E+01	1.3.E+01

Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))	Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))
AM22	4.1.E+04	3.0.E+04	1.5.E+05	3.7.E+00	5.7.E+00	W24	4.5.E+04	3.6.E+04	5.2.E+05	1.2.E+01	1.4.E+01
AM23	4.2.E+04	1.6.E+03	3.1.E+05	7.2.E+00	4.6.E-01	W25	4.6.E+04	3.3.E+04	2.1.E+05	4.5.E+00	4.2.E+01
AM24	4.7.E+04	4.0.E+03	7.1.E+06	1.5.E+02	1.5.E+01	W26	4.8.E+04	2.0.E+04	7.1.E+05	1.5.E+01	3.0.E+01
AM25	4.8.E+04	1.1.E+03	3.1.E+06	6.4.E+01	5.6.E-01	W27	4.9.E+04	3.8.E+02	1.6.E+05	3.2.E+00	4.9.E-01
AM26	5.1.E+04	4.9.E+04	4.8.E+06	9.4.E+01	9.8.E+01	W28	5.1.E+04	3.0.E-04	1.0.E+00	2.0.E-05	0.0.E+00
AM27	5.0.E+04	4.9.E+04	1.8.E+06	3.5.E+01	3.7.E+01	W7	1.2.E+04	2.0.E+02	1.0.E+00	8.4.E-05	0.0.E+00
AM28	5.1.E+04	4.9.E+04	3.2.E+06	6.4.E+01	2.4.E+01	W8	1.4.E+04	2.8.E+03	1.0.E+00	7.1.E-05	1.0.E-03
AM29	5.2.E+04	4.9.E+04	4.8.E+06	9.1.E+01	3.2.E+01	W9	1.6.E+04	1.5.E+04	5.6.E+00	3.5.E-04	4.8.E-03
AM30	5.2.E+04	4.6.E+04	1.3.E+06	2.4.E+01	8.2.E+01	X10	1.8.E+04	4.7.E+03	1.0.E+00	5.5.E-05	1.4.E-03
AM31	5.4.E+04	9.8.E+03	9.0.E+05	1.7.E+01	2.2.E+01	X11	2.0.E+04	1.8.E+04	1.0.E+00	4.9.E-05	1.8.E-03
AM32	5.5.E+04	1.1.E+03	1.2.E+06	2.2.E+01	7.5.E-03	X12	2.2.E+04	1.9.E+04	1.0.E+00	4.5.E-05	1.4.E-02
AM33	5.6.E+04	3.7.E-02	1.2.E+02	2.2.E-03	0.0.E+00	X13	2.4.E+04	1.9.E+04	2.7.E+00	1.1.E-04	1.7.E-02
AM4	2.9.E+03	1.3.E+03	1.0.E+00	3.5.E-04	0.0.E+00	X14	2.6.E+04	2.4.E+04	5.6.E+00	2.1.E-04	7.7.E-03
AM5	7.6.E+03	7.5.E+03	2.8.E+00	3.7.E-04	0.0.E+00	X15	2.8.E+04	2.6.E+04	1.3.E+03	4.6.E-02	2.6.E-02
AM6	1.0.E+04	1.0.E+04	3.2.E+00	3.1.E-04	0.0.E+00	X16	3.2.E+04	3.1.E+04	4.6.E+02	1.5.E-02	6.3.E-01
AM7	1.2.E+04	9.8.E+03	1.8.E+02	1.5.E-02	0.0.E+00	X17	3.4.E+04	3.2.E+04	2.1.E+04	6.3.E-01	7.2.E+00
AM8	1.4.E+04	9.3.E+03	2.7.E+00	1.9.E-04	0.0.E+00	X18	3.6.E+04	3.4.E+04	3.0.E+04	8.5.E-01	8.7.E+00
AM9	1.6.E+04	1.2.E+04	3.6.E+02	2.2.E-02	7.0.E-03	X19	3.7.E+04	3.7.E+04	4.2.E+04	1.1.E+00	7.0.E-01
AN10	1.8.E+04	1.8.E+04	6.0.E+00	3.3.E-04	6.7.E-03	X20	3.9.E+04	3.8.E+04	1.3.E+05	3.3.E+00	1.7.E+00
AN11	2.0.E+04	5.7.E+03	2.1.E+03	1.0.E-01	2.3.E-03	X21	3.9.E+04	2.9.E+04	7.0.E+05	1.8.E+01	2.4.E+00
AN12	2.2.E+04	5.0.E+03	1.6.E+00	7.4.E-05	2.0.E-03	X22	4.2.E+04	1.1.E+03	3.3.E+06	7.9.E+01	1.2.E-02
AN13	2.4.E+04	2.9.E+03	1.0.E+00	4.1.E-05	9.4.E-04	X23	5.0.E+04	4.8.E+04	5.3.E+05	1.1.E+01	2.4.E+01
AN14	2.6.E+04	5.9.E+03	1.0.E+00	3.8.E-05	1.7.E-03	X24	5.0.E+04	4.8.E+04	1.1.E+05	2.2.E+00	2.8.E+01
AN15	2.8.E+04	3.3.E+02	1.0.E+00	3.5.E-05	8.1.E-05	X25	5.0.E+04	4.8.E+04	2.9.E+05	5.9.E+00	6.0.E+01
AN16	3.0.E+04	2.0.E+02	1.0.E+00	3.3.E-05	4.8.E-05	X26	5.1.E+04	4.9.E+04	3.5.E+06	6.9.E+01	6.6.E+01
AN17	3.2.E+04	6.5.E+01	1.0.E+00	3.1.E-05	1.4.E-05	X27	5.1.E+04	4.2.E+04	7.8.E+06	1.5.E+02	4.4.E+01
AN18	3.4.E+04	1.5.E+03	3.4.E+01	1.0.E-03	2.0.E-04	X28	5.1.E+04	1.3.E+04	4.8.E+05	9.5.E+00	1.8.E+00
AN19	3.6.E+04	1.1.E+04	2.9.E+03	8.3.E-02	4.3.E-02	X29	5.2.E+04	1.9.E-01	6.3.E+02	1.2.E-02	0.0.E+00
AN20	3.7.E+04	3.5.E+04	3.8.E+03	1.0.E-01	1.2.E+00	X7	1.2.E+04	4.3.E+03	1.0.E+00	8.4.E-05	0.0.E+00
AN21	3.9.E+04	3.9.E+04	7.5.E+04	1.9.E+00	3.6.E+00	X8	1.4.E+04	4.1.E+03	1.0.E+00	7.1.E-05	1.4.E-03
AN22	4.1.E+04	3.8.E+04	1.8.E+05	4.4.E+00	7.5.E+00	X9	1.8.E+04	1.7.E+04	1.0.E+00	5.5.E-05	5.0.E-03
AN23	4.2.E+04	1.6.E+04	2.4.E+05	5.8.E+00	2.5.E+01	Y10	1.8.E+04	3.8.E+03	1.3.E+03	7.2.E-02	1.1.E-03
AN24	4.6.E+04	2.8.E+04	2.3.E+06	5.0.E+01	1.9.E+02	Y11	2.0.E+04	1.3.E+04	9.7.E+03	4.8.E-01	1.3.E-03
AN25	4.9.E+04	2.1.E+03	6.1.E+06	1.3.E+02	9.2.E-02	Y12	2.2.E+04	1.9.E+04	2.2.E+04	1.0.E+00	1.4.E-02
AN26	5.0.E+04	4.8.E+04	1.5.E+06	2.9.E+01	4.6.E+01	Y13	2.4.E+04	2.0.E+04	1.7.E+04	6.9.E-01	1.9.E-02
AN27	4.9.E+04	4.8.E+04	2.0.E+06	4.2.E+01	1.7.E+01	Y14	2.6.E+04	2.4.E+04	3.6.E+04	1.4.E+00	1.1.E-02
AN28	5.1.E+04	4.8.E+04	4.4.E+06	8.6.E+01	2.2.E+01	Y15	2.8.E+04	2.7.E+04	4.7.E+04	1.7.E+00	1.8.E-02
AN29	5.2.E+04	4.5.E+04	2.1.E+06	4.0.E+01	7.2.E+00	Y16	3.2.E+04	3.1.E+04	1.6.E+05	5.1.E+00	7.4.E-01
AN30	5.3.E+04	2.2.E+04	1.6.E+06	3.0.E+01	7.8.E+00	Y17	3.4.E+04	3.3.E+04	3.1.E+05	9.2.E+00	9.8.E+00
AN31	5.5.E+04	2.7.E+04	4.5.E+06	8.1.E+01	1.2.E+01	Y18	3.6.E+04	3.4.E+04	5.5.E+05	1.5.E+01	1.9.E+01
AN32	5.6.E+04	3.5.E+04	5.4.E+06	9.6.E+01	2.5.E+01	Y19	3.8.E+04	3.7.E+04	1.7.E+06	4.5.E+01	6.6.E+00
AN33	5.7.E+04	7.9.E+03	2.7.E+06	4.9.E+01	2.0.E-02	Y20	4.0.E+04	3.9.E+04	1.8.E+06	4.5.E+01	2.0.E+00
AN4	2.9.E+03	2.5.E+03	1.0.E+00	3.5.E-04	0.0.E+00	Y21	3.9.E+04	3.2.E+04	1.1.E+06	2.9.E+01	2.5.E+00

Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))	Air cell No.	Area (km2)	Land (soil + paved) (km2)	Population (pers)	Population density (pers/km2 (total area))	Agricultural intensity (t/(km2·y))
AN5	8.5.E+03	8.5.E+03	2.8.E+00	3.3.E-04	0.0.E+00	Y22	4.1.E+04	1.3.E+03	1.0.E+05	2.5.E+00	9.7.E-02
AN6	1.1.E+04	1.1.E+04	3.4.E+00	3.1.E-04	0.0.E+00	Y23	5.0.E+04	4.9.E+04	1.0.E+05	2.0.E+00	1.9.E+01
AN7	1.2.E+04	1.1.E+04	3.0.E+00	2.5.E-04	0.0.E+00	Y24	5.0.E+04	4.9.E+04	1.0.E+05	2.0.E+00	8.6.E+00
AN8	1.4.E+04	9.1.E+03	2.6.E+00	1.9.E-04	0.0.E+00	Y25	5.0.E+04	5.0.E+04	1.0.E+05	2.0.E+00	4.1.E+00
AN9	1.6.E+04	1.1.E+04	4.0.E+00	2.5.E-04	6.6.E-03	Y26	5.0.E+04	4.9.E+04	1.0.E+05	2.0.E+00	8.4.E+00
AO10	1.8.E+04	1.6.E+04	5.5.E+00	3.0.E-04	5.7.E-03	Y27	5.1.E+04	4.9.E+04	1.5.E+06	2.9.E+01	4.1.E+01
AO11	2.0.E+04	2.9.E+03	1.0.E+00	4.9.E-05	7.9.E-04	Y28	5.0.E+04	4.7.E+04	2.6.E+06	5.2.E+01	5.0.E+01
AO12	2.2.E+04	3.0.E-04	1.0.E+00	4.5.E-05	0.0.E+00	Y29	5.3.E+04	1.9.E+04	4.1.E+06	7.8.E+01	2.3.E+01
AO13	2.4.E+04	2.1.E+03	1.0.E+00	4.1.E-05	5.2.E-04	Y7	1.2.E+04	6.9.E+03	2.7.E+01	2.3.E-03	0.0.E+00
AO14	2.6.E+04	4.4.E+03	2.8.E+02	1.1.E-02	1.1.E-03	Y8	1.4.E+04	3.3.E+03	1.9.E+02	1.3.E-02	6.4.E-04
AO15	2.8.E+04	4.4.E+03	4.3.E+02	1.5.E-02	8.4.E-04	Y9	1.6.E+04	1.4.E+04	2.7.E+02	1.7.E-02	4.5.E-03
AO16	3.0.E+04	5.4.E+03	1.2.E+03	4.1.E-02	8.8.E-04	Z10	1.8.E+04	1.4.E+04	1.3.E+03	7.2.E-02	4.1.E-03
AO17	3.2.E+04	2.6.E+03	7.6.E+02	2.4.E-02	5.8.E-04	Z11	2.0.E+04	1.5.E+04	9.7.E+03	4.8.E-01	2.4.E-03
AO18	3.4.E+04	1.3.E+04	9.0.E+02	2.7.E-02	9.2.E-03	Z12	2.2.E+04	2.0.E+04	2.2.E+04	1.0.E+00	1.4.E-02
AO19	3.6.E+04	1.7.E+04	5.1.E+03	1.4.E-01	1.2.E-02	Z13	2.4.E+04	2.0.E+04	1.7.E+04	6.9.E-01	1.9.E-02
AO20	3.7.E+04	3.2.E+04	1.9.E+03	5.1.E-02	1.7.E+00	Z14	2.6.E+04	2.5.E+04	3.6.E+04	1.4.E+00	1.5.E-02
AO21	3.9.E+04	3.9.E+04	1.1.E+05	2.9.E+00	4.0.E+00	Z15	3.4.E+04	3.2.E+04	4.7.E+04	1.4.E+00	6.1.E-02
AO22	4.1.E+04	3.9.E+04	1.4.E+05	3.4.E+00	7.6.E+00	Z16	3.2.E+04	3.1.E+04	1.6.E+05	5.1.E+00	2.2.E+00
AO23	4.2.E+04	3.1.E+04	1.1.E+06	2.7.E+01	3.1.E+01	Z17	3.4.E+04	3.2.E+04	3.1.E+05	9.2.E+00	1.1.E+01
AO24	4.8.E+04	6.1.E+03	8.6.E+06	1.8.E+02	1.4.E+01	Z18	3.6.E+04	3.4.E+04	5.5.E+05	1.5.E+01	2.2.E+01
AO25	5.1.E+04	4.9.E+04	2.6.E+06	5.2.E+01	5.0.E+01	Z19	3.8.E+04	3.7.E+04	1.7.E+06	4.5.E+01	2.5.E+01
AO26	4.8.E+04	4.6.E+04	1.9.E+06	3.9.E+01	1.7.E+01	Z20	4.0.E+04	3.9.E+04	1.8.E+06	4.5.E+01	2.1.E+01
AO27	4.9.E+04	4.6.E+04	2.4.E+06	5.0.E+01	2.8.E+01	Z21	3.9.E+04	3.9.E+04	1.1.E+06	2.9.E+01	3.1.E+00
AO28	5.1.E+04	4.5.E+04	3.3.E+06	6.5.E+01	1.3.E+01	Z22	4.1.E+04	5.4.E+03	1.0.E+05	2.5.E+00	4.2.E-01
AO29	5.2.E+04	1.1.E+04	6.0.E+05	1.2.E+01	2.9.E+00	Z23	5.0.E+04	4.9.E+04	1.0.E+05	2.0.E+00	1.9.E+01
AO30	5.3.E+04	3.0.E-04	1.0.E+00	1.9.E-05	0.0.E+00	Z24	5.0.E+04	4.9.E+04	1.0.E+05	2.0.E+00	8.6.E+00
AO31	5.4.E+04	3.0.E-04	1.0.E+00	1.9.E-05	0.0.E+00	Z25	5.0.E+04	5.0.E+04	1.0.E+05	2.0.E+00	4.1.E+00
AO32	5.5.E+04	1.4.E+01	4.6.E+04	8.5.E-01	0.0.E+00	Z26	5.0.E+04	4.9.E+04	1.0.E+05	2.0.E+00	8.4.E+00
AO33	5.6.E+04	9.5.E+00	3.2.E+04	5.6.E-01	0.0.E+00	Z27	5.1.E+04	4.9.E+04	1.5.E+06	2.9.E+01	4.1.E+01
AO4	3.7.E+03	3.6.E+03	1.0.E+00	2.7.E-04	0.0.E+00	Z28	5.0.E+04	4.7.E+04	2.6.E+06	5.2.E+01	5.0.E+01
AO5	8.4.E+03	8.3.E+03	2.8.E+00	3.3.E-04	0.0.E+00	Z29	5.3.E+04	1.9.E+04	4.1.E+06	7.8.E+01	2.3.E+01
AO6	1.2.E+04	1.2.E+04	3.4.E+00	2.9.E-04	0.0.E+00	Z30	5.3.E+04	3.0.E-04	1.0.E+00	1.9.E-05	0.0.E+00
AO7	1.2.E+04	7.5.E+03	2.2.E+00	1.8.E-04	0.0.E+00	Z6	9.8.E+03	3.0.E-04	1.0.E+00	1.0.E-04	0.0.E+00
AO8	1.4.E+04	9.6.E+02	1.0.E+00	7.1.E-05	0.0.E+00	Z7	1.2.E+04	5.6.E+03	2.7.E+01	2.3.E-03	0.0.E+00
AO9	1.6.E+04	1.0.E+04	1.4.E+03	8.6.E-02	6.0.E-03	Z8	1.4.E+04	5.6.E+03	1.9.E+02	1.3.E-02	1.4.E-04
AP10	1.8.E+04	1.8.E+04	5.7.E+00	3.1.E-04	6.8.E-03	Z9	1.6.E+04	1.4.E+04	2.7.E+02	1.7.E-02	4.6.E-03
AP11	2.0.E+04	1.0.E+04	3.4.E+00	1.7.E-04	2.8.E-03						

Table 3-5 presents the oral and inhalation intake fraction for the different pollutants evaluated in this chapter and used to plot Figure 3-3.

Table 3-5: Oral and inhalation intake fraction for the different pollutants evaluated in this chapter.

Pollutant	Oral intake fraction, using (for the weighted average):			Inhalation intake fraction, using (for the weighted average):		
	Population	Agricultural production intensity	Land area	Population	Agricultural production intensity	Land area
PM	0.0E+00	0.0E+00	0.0E+00	6.0E-06	7.6E-07	2.9E-07
Benzo(a)pyrene	3.6E-03	8.5E-03	3.4E-03	5.8E-06	5.6E-07	2.2E-07
Tetrachloroethylene	1.5E-08	3.0E-08	1.8E-08	1.3E-05	8.2E-06	5.4E-06
Carbon tetrachloride	1.0E-06	1.7E-06	8.9E-07	5.3E-04	5.2E-04	2.8E-04
1,3-Butadiene	3.9E-12	9.7E-12	3.5E-12	5.2E-06	1.3E-07	4.9E-08
Methomyl	2.2E-04	5.6E-04	1.9E-04	5.8E-06	5.2E-07	2.0E-07
Acephate	1.8E-06	3.3E-06	1.1E-06	5.3E-06	1.5E-07	5.5E-08
Formaldehyde	5.6E-07	1.5E-06	4.6E-07	5.2E-06	1.2E-07	4.4E-08
PCBs _s	4.0E-04	8.0E-04	4.5E-04	8.2E-06	2.9E-06	1.7E-06
Di(n-octyl) phthalate	6.7E-06	1.7E-05	5.5E-06	5.5E-06	3.2E-07	1.2E-07
Benzene, hexabromo-	3.6E-03	7.4E-03	4.2E-03	8.5E-06	3.4E-06	1.9E-06
Cypermethrin	3.2E-06	6.3E-06	2.5E-06	5.3E-06	1.9E-07	6.9E-08
Mirex	1.5E-04	3.7E-04	1.6E-04	6.2E-06	8.6E-07	3.6E-07
Trifluralin	1.4E-04	3.3E-04	1.6E-04	6.7E-06	1.4E-06	6.3E-07
Dicofol	2.5E-05	6.1E-05	2.1E-05	5.7E-06	4.3E-07	1.6E-07
p-Dichlorobenzene	2.5E-07	4.9E-07	3.0E-07	1.3E-05	8.1E-06	5.4E-06
Aldrin	5.3E-08	1.3E-07	4.4E-08	5.2E-06	1.3E-07	4.8E-08
1,1,2,2-Tetrachloroethane	6.4E-06	1.2E-05	7.5E-06	2.5E-05	2.1E-05	1.5E-05
Captan	5.5E-07	1.4E-06	5.1E-07	5.6E-06	4.1E-07	1.5E-07
Pronamide	7.4E-05	1.6E-04	9.2E-05	8.9E-06	4.1E-06	2.3E-06
Anthracene	1.7E-05	4.2E-05	1.7E-05	6.3E-06	9.7E-07	3.8E-07
gamma-Hexachlorocyclohexane	6.2E-05	1.5E-04	7.1E-05	7.0E-06	1.8E-06	7.9E-07
Dimethyl phthalate	1.4E-05	3.3E-05	1.5E-05	7.1E-06	1.9E-06	8.6E-07
Methanol	5.0E-06	1.2E-05	5.6E-06	7.1E-06	2.1E-06	9.5E-07
1,2-Dichloroethane	9.2E-07	1.7E-06	1.0E-06	2.5E-05	2.1E-05	1.5E-05
Ethyl acetate	4.2E-08	1.0E-07	4.0E-08	6.5E-06	1.2E-06	4.9E-07
N-Nitrosodiethylamine	1.3E-07	3.5E-07	1.1E-07	5.2E-06	1.3E-07	4.7E-08
Thioperoxydicarbonic diamide, tetramethyl-	6.9E-05	1.8E-04	6.9E-05	5.9E-06	5.9E-07	2.4E-07
Propoxur	1.9E-06	4.2E-06	1.4E-06	5.2E-06	9.5E-08	3.5E-08
1H-Isoindole-1,3(2H)-dione, 2-(trichloromethyl)thio -	2.2E-05	4.1E-05	1.5E-05	5.5E-06	2.6E-07	9.9E-08

4. Intake fractions and characterization factors for particulate matter: Review and recommendations for life-cycle assessment

Particulate matter (PM) is a significant cause of adverse human health effects (Pope et al. 2009). However, the IMPACT North America model developed in Chapter 2 is actually more specifically developed for organic molecules rather than PM. The IMPACT North America model can evaluate the fate and exposure of primary PM but not of secondary PM. Therefore I initiated an effort to evaluate how to model the intake fractions of PM using alternative approaches and models.

4.1. Summary

PM is a significant cause of adverse human health effects. To foster health assessment of PM in life-cycle assessment, I review values for PM for two parameters: intake fraction (the fraction of emissions that are inhaled) and characterization factor (the human health damage per mass emitted). My approach presents impacts from PM based on the emissions source height and the “archetypal” emissions environment (indoor versus outdoor; urban, rural, or remote locations). Recommended intake fraction and interim characterization factor values are provided for primary PM₁₀, primary coarse PM (PM_{10-2.5}), primary PM_{2.5}, and secondary inorganic PM (from SO₂, NO_x, and NH₃). Considering an emission-weighted average stack height, intake fraction values (and characterization factors in micro disability-adjusted life years (μDALY, the number of years that are lost within the population because of mortality or morbidity) per kg) for primary PM_{2.5} for urban, rural, and remote areas are 19 (3,400 μDALY/kg), 2.2 (390 μDALY/kg), and 0.084 (14 μDALY/kg) ppm, respectively. For secondary PM, source location and source characteristics have only a minor influence on the magnitude of the intake fraction, except for remote locations where results can be lower than elsewhere by an order of magnitude. For indoor household emissions, typical intake fractions and characterization factors are 4,700 ppm and 610,000 micro μDALY per kg, respectively — values that are about two to three orders of magnitude greater than outdoor urban or rural releases. The intake fractions (and characterization factors) averaged over source location and stack height distribution are 7.0 ppm (n/a⁴ μDALY/kg) for primary PM_{10-2.5} and 10 ppm (1,800 μDALY/kg) for primary PM_{2.5}. For secondary PM, intake fraction (and characterization factor) values are 0.79 ppm (110 μDALY/kg) for SO₂, 0.16 ppm (13 μDALY/kg) for NO_x, and 1.5 ppm (120 μDALY/kg) for NH₃. This chapter aims to provide as complete and consistent an archetype framework as possible, given current understanding of each pollutant. Values presented here facilitate incorporating regional impacts into life-cycle assessment for human health damage from PM.

4.2. Introduction

This chapter aims to review and recommend a consistent set of factors for intake of, and human health damage from, primary and secondary PM.

Several studies show that PM causes serious adverse health effects, including reduced life expectancy, lung cancer, asthma, low birth weight and premature birth (Dockery et al. 1993, Dockery and Pope, 1994, Pope et al. 1995, Pope et al. 2002, Pope et al. 2009, Kuenzli et al.

⁴ Not available. See main text for explanation.

2000, Laden et al. 2000, Laden et al. 2006, Cooke et al. 2007, Bell et al. 2008, Schwartz et al. 2008, Siddiqui et al. 2008, Jerrett et al. 2008). Ambient PM can be primary (i.e., directly emitted) or secondary (i.e., formed in the atmosphere from precursors). Precursors involved in secondary PM formation include sulfur dioxide (SO₂), nitrogen oxides (NO_x), ammonia (NH₃), and volatile and semi-volatile organic compounds.

Several life-cycle impact assessment methods have evaluated the human health damage per mass of particles emitted (Hofstetter 1998, Bare et al. 2003, Jolliet et al. 2003, Van Zelm et al. 2008). Hofstetter (1998) generated one of the first life-cycle impact assessment approaches evaluating damage factors for PM, based on a consistent integration of data from existing models and epidemiological studies. Since then, researchers have continued to develop fate and exposure models (Levy et al. 2002, Marshall et al. 2005, Greco et al. 2007, Rosenbaum et al. 2008) and revise epidemiological data (Pope et al. 2002, Kuenzli et al. 2000, Laden et al. 2006, Schwartz et al. 2008). Previous reviews suggest that human health damage needs to be assessed in a regional context to increase the confidence in, accuracy of, and acceptance of life-cycle impact assessment results (Potting and Hauschild 2006, Sedlbauer et al. 2007, Reap et al. 2008). Potting et al. (2007), reviewing life-cycle impact assessment research on PM, suggested the need for consistency in fate, exposure, and effect evaluation. The present work aims to fill this gap by providing consistent damage factors that consider archetypal differences among regions (“regionalization”).

The objectives of this chapter are to review methods that provide intake fractions and characterization factors of PM, and to recommend source-location intake fraction and interim characterization factor matrices that facilitate straightforward application to life-cycle assessment, in a manner consistent with current approaches for organic pollutants.

I first describe the general framework for calculating damage factors, considering regionalization and suggesting parameters to evaluate. Then I review estimated intake fraction values. I evaluate and suggest parameters to represent the different environmental and exposure situations, and propose a matrix of recommended intake fractions. I close by proposing a set of interim effect factors and characterization factors, and by discussing the outlook for further research. This chapter is based on a meta-review complemented with expert judgment.

The work reported here was done in coordination with and is compatible with the UNEP-SETAC Life Cycle Initiative task force responsible for PM and with the review and recommendations on life-cycle impact assessment made for the European Union by a consortium of experts (European Commission, DG Joint Research Centre, available at <http://lct.jrc.ec.europa.eu>). While this chapter evaluates PM only, a similar approach could be developed for other pollutants or environmental media (not evaluated in this dissertation).

4.3. Methods

General framework

Human health impacts can be expressed using disability-adjusted life years (DALYs – Murray and Lopez 1996). Other health indicators also exist, such as premature death, life-expectancy or quality-adjusted life years (QALY). One of the advantages of the DALY is that it accounts for several forms of burden such as mortality and morbidity. Furthermore, most impact methods used in life-cycle assessment use the unit of DALY to express damage to human health. Using a unit that is compatible with units that are already used in life-cycle impact assessment makes the results herein directly usable in life-cycle impact assessment.

In life-cycle assessment, the characterization factor (CF , $\text{DALY kg}_{\text{emitted}}^{-1}$) of a given atmospheric emission can be evaluated as the product of four parameters.

$$CF = SF \times DR \times XF \times FF = EF \times iF \quad (4-1)$$

The fate factor (FF , kg_{air} per $[\text{kg}_{\text{emitted}} \text{d}^{-1}]$) relates the emission rate ($\text{kg}_{\text{emitted}} \text{d}^{-1}$) to the mass in the exposure medium (kg_{air}); the exposure factor (XF , $[\text{kg}_{\text{inhaled}} \text{d}^{-1}]$ per kg_{air}) determines the change in intake rate per change in mass in the environment; the dose-response (DR , health impact per $\text{kg}_{\text{inhaled}}$) indicates the change in morbidity or mortality attributable to a change in intake; and, the severity factor (SF , DALY per health impact) is the severity per change in morbidity or mortality. The emitted pollutant can be a single chemical or a group of chemicals, and it can be a primary pollutant or a contributor to a secondary pollutant (Rosenbaum et al. 2007).

As shown in Equation 4-1, two terms XF and FF are often combined into intake fraction (iF , $\text{kg}_{\text{inhaled}}$ per $\text{kg}_{\text{emitted}}$; a common shorthand, employed below, is to convert to ppm, i.e., $\text{mg}_{\text{inhaled}}$ per $\text{kg}_{\text{emitted}}$) (Bennett et al. 2002), and the remaining terms SF and DR can be combined in the effect factor (EF , $\text{DALY kg}_{\text{inhaled}}^{-1}$). Intake fraction for primary pollutants indicates the fraction of the emission taken in (inhaled) by the population; intake fraction for secondary PM is the mass of PM attributable to a specific precursor inhaled per mass emission of the precursor. Note that in cases secondary PM is composed of both nitrogen and sulfur such as in $(\text{NH}_4)_2\text{SO}_4$, an issue of double counting may appear when adding the damage from NH_3 emissions and SO_2 emissions. This is limitation of the approach developed in the present dissertation and would require further work to better address this issue of double counting. Figure 4-1 illustrates the cause-effect chain of fate, exposure, dose-response, and severity assessment.

Life-cycle impact assessment studies often assume a linear, no-threshold dose response curve, an approach that for PM is supported by several studies (WHO 2006, Roman et al. 2008, Schwartz et al. 2008). However, in cases where PM concentrations are higher or lower than those observed in epidemiological studies (typically, $\sim 10\text{--}35 \mu\text{g m}^{-3}$ for $\text{PM}_{2.5}$), the linearity assumption may not apply.

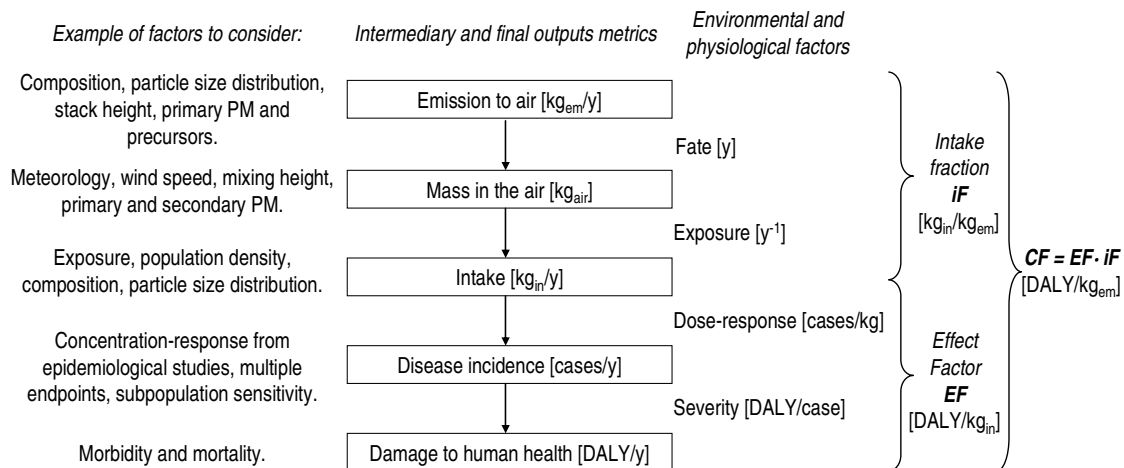


Figure 4-1: Emission-to-damage framework for particulate matter.

Intake fraction

The inhalation intake fraction (iF) of a pollutant p is evaluated using Equation 4-2 (Marshall et al. 2005, Bennett et al. 2002):

$$iF_p = \frac{\int_{time} N(t) \times BR(t) \times C_{p,air}(t) \times dt}{S_p}, \quad (4-2)$$

where N (persons) is the number of persons exposed as a function of time t (s), BR ($\text{m}^3 \text{ person}^{-1} \text{ d}^{-1}$) is the volumetric breathing rate, and $C_{p,air}$ (kg m^{-3}) is the incremental exposure concentration attributable to emission S_p (kg). I employ here a population average breathing rate of $13 \text{ m}^3 \text{ person}^{-1} \text{ d}^{-1}$ (USEPA 1997).

Factors influencing the intake fraction

Regionalization. Recent studies emphasize the importance of regionalization in life-cycle assessment (EC 2005, Rochat et al. 2006) and fate and exposure of PM (Levy et al. 2002, Wang et al. 2006, Zhou et al. 2006, Greco et al. 2007, Tainio et al. 2009). Several studies show that intake fraction is strongly correlated with population density (Greco et al. 2007, Rosenbaum et al. 2008, EC 2005, Tainio et al. 2009, Heath et al. 2006, Preiss et al. 2008, Spadaro and Rabl 2004) and meteorological conditions (Levy et al. 2002, Spadaro and Rabl 2004), especially mixing height and wind speed (Marshall et al. 2005), but also relative concentrations of sulfate-nitrate-ammonium as well as temperature. The averaging method—for example, arithmetic versus harmonic mean—may be important for considering impacts of meteorology on intake fraction (Marshall et al. 2005).

Height of emission. Fate and exposure of PM is influenced by the height at which pollutants are emitted (Van Zelm et al. 2008, Levy et al. 2002, Heath et al. 2006, Heath and Nazaroff 2007, Spadaro and Rabl 2004). Life-cycle inventories (e.g., ecoinvent — Frischknecht 2005) often distinguish among high-stack (e.g., power plants), low-stack (e.g., residential wood combustion), and ground-level sources (e.g., road transportation). Levy et al. (2002) found that primary $\text{PM}_{2.5}$ intake fractions are at least four times greater for mobile (ground-level) emissions as for stationary-source (elevated) emissions.

Archetypes addressing regionalization and the height of emission. Although the exact location of emissions is often unknown in life-cycle assessment, some life-cycle inventories (e.g., ecoinvent — Frischknecht 2005) and software give information regarding the location of emissions (e.g., high versus low population density) and some source types can be extrapolated (e.g., coal power-plants generally involve high-stack emissions). Archetypal environments aim to include vertical and horizontal spatial considerations in cases where full details (e.g., exact emission location or population density) are unavailable (Sedlbauer et al. 2007).

I employ four archetypal environments: indoor sources, and outdoor sources in urban, rural, and remote locations. I further delineate three categories for emission height: ground-level, low-stack and high-stack. Here, stack height generally refers to the physical stack height, and not effective stack height considering plume rise. Emissions at high altitude (e.g., from airplanes) are not considered here owing to a lack of relevant studies addressing the fate and exposure of PM emitted at high altitude. I do not distinguish among ground-level sources (e.g., area sources, on-road mobile, off-road mobile), though I recognize that further refinement on this point may be beneficial. Marshall (2005) concluded that intake fractions for ground-level urban sources are ~1.3 to 5.1 times higher for on-road emissions than for off-road emissions.

Types of PM. The PM-related pollutants considered in this work are primary PM₁₀, primary PM_{10-2.5}, primary PM_{2.5} and secondary PM from SO₂, NO_x, and NH₃. I also summarize results for direct exposure to SO₂, NO_x, and NH₃, so that effects from direct exposure can also be taken into account in characterization factors that, though being dominated by secondary PM exposure, would be applied to emissions of SO₂, NO_x and NH₃, and should therefore also account for the damage from direct exposure. Because of the lack of data, secondary PM from volatile organic compounds is outside the scope of this chapter but is recommended as an area of further research (Muller and Mendelsohn 2007, Ilacqua et al. 2007, Kanakidou et al. 2005). Finally, carbon monoxide (CO) is also evaluated because this chemical is often reported along with primary and secondary PM in the so-called “respiratory inorganics” impact category by life-cycle impact assessment methods.

The intake fraction of secondary PM is calculated by dividing the mass of secondary PM inhaled by the mass of precursors emitted. I assume that SO₂ creates ammonium sulfate ((NH₄)₂SO₄), that NO_x creates ammonium nitrate (NH₄NO₃) and that NH₃ creates both ammonium sulfate ((NH₄)₂SO₄) and ammonium nitrate (NH₄NO₃). Further research is needed to better capture the composition and mass of secondary PM attributable to precursors as well as the potential double counting such as in the case of ammonium sulfate (NH₄)₂SO₄ and the influence of precursors present in the air and influence the secondary PM formation of other precursors.

The two main attributes of PM — the size distribution and the chemical composition — vary among sources, and may influence the dose-response relationship. Franklin et al. (2008) show that certain chemical species significantly modify the association between PM and mortality, suggesting that mass alone may be an imperfect metric when evaluating the health effects of PM. I do not evaluate the influence of chemical composition — outside of distinguishing the type of PM described above — on effect factor because of limited available research on this topic.

Source-location framework. The three emission heights (vertical consideration), four emission locations (horizontal consideration), and six pollutants yield up to 72 possible combinations. Factors reducing the actual size of the source-location-pollutant matrix include that indoor emissions do not require a stack height, and stack height is of limited importance for intake fraction and characterization factor of secondary PM.

4.4. Archetype parameterization

This section characterizes representative parameters for indoor, urban, rural, remote, and unknown environments.

The need for one geographically average value, per archetype, is for cases in life-cycle assessment when the only information known is that the emissions occurred in a specific archetype, but not where the emission occurred (e.g., continent). However, if the user knows the continent where the emissions took place, then the user can use a customized, re-calculated value based on the information presented in section 4.11.

Indoor. Intake fraction values are approximately three orders of magnitude greater for indoor than for outdoor emissions (Lai et al. 2000, Ilacqua et al. 2007, Smith 1988, Klepeis and Nazaroff 2006, Hellweg et al. 2009). A steady-state one-compartment model (Equation 4-3 — Hellweg et al. 2009) is commonly used to estimate indoor intake fraction values, although researchers have also considered episodic emissions (Nazaroff 2008) and multicompartment (Klepeis and Nazaroff 2006) indoor environments.

$$iF_{indoor} = f_{TE} \times N \times BR / (V \times m \times k_{ex}) \quad (4-3)$$

Here N is the number of persons affected and breathing at a rate BR (m^3/h), V is the indoor volume (m^3), m is the mixing factor (unitless), k_{ex} is the air exchange rate (h^{-1}), and f_{TE} (unitless) is the fraction of time people are exposed, i.e., for emissions that occur even when people are not present. Representative parameters and resulting intake fractions are presented in Table 4-1.

Table 4-1: Representative parameters and resulting intake fractions (iF) for indoor emissions.

Type of environment	V/N (m^3/person)	m (unitless) ^a	k_{ex} (d^{-1}) ^a	iF (ppm) for full-time exposure	f_{TE} (d/d)	iF (ppm) for real time exposure
household	160 ^a	1	12	6,800	0.7	4,700
office	100 ^b	1	24	5,400	0.3	1,600
industrial	1,000 ^b	1	240	54	0.6 (assuming two shifts)	33

^a Adapted from Hellweg et al. (2009).
^b Assumption. In need for further research – currently under investigation by Hellweg et al. (2009).

My treatment of PM intake fraction for indoor emissions is based on the recommendations from Hellweg et al. (2009). However, I need here to acknowledge three weaknesses that should be considered when improving the modeling of indoor intake fractions: (i) Equation 4-3 is missing deposition as a removal process; (b) Equation 4-3 is missing the temporal correlation between occupancy and emissions (which influences the factor f_{TE} — if PM releases is correlated with occupancy, then $f_{TE} \rightarrow 1.0$); and (c) Equation 4-3 misses the potentially important removal process in some indoor environments (e.g., offices) from recirculated air with HVAC filters.

Urban. Definitions for “urban area” vary. The United States Census defines an area as urban if the population density is at least 1,000 persons mile⁻² (390 persons km⁻²) and so long as the surrounding area has a density of at least 500 persons mile⁻² (190 persons km⁻²). TRACI (Bare et al. 2003) employs a threshold population density of 100 persons km⁻² to distinguish urban versus non-urban. USES-LCA (Van Zelm et al. 2009, Huijbregts et al. 2005b) employs an urban box with average population density of 2,000 persons km⁻². The population-weighted average urban area in the United States can be represented as a 49 km × 49 km square with a population density of 753 persons km⁻² (see section 2.3 in Chapter 2). On average, population density is generally lower in United States cities than in cities worldwide (Marshall 2007). Globally, urban population densities can reach extremes of 30,000 persons km⁻² (Marshall 2007, Marcotullio and Marshall 2007).

I propose here to parameterize the default urban box to reflect the population-weighted arithmetic average intake fraction for all urban areas worldwide. Having a default urban box parameterize based on all urban areas worldwide permits that to have the resulting intake fraction directly valid for unknown worldwide supply chains and directly usable in impact assessment methods. In section 4.11 I provide an approach to evaluate the intake fractions for North America, for both urban and rural conditions. For intake fraction calculations, linear population density (the cross population per unit of distance, e.g., the population that is present in a “band” with a width of one kilometer; Marshall et al. 2005, Marshall 2007 — see section 4.11) is often a more useful parameter than areal population density. The default urban box has a linear population density of 80,000 persons km⁻¹, a population density of 4,000 persons km⁻², a 20 km × 20 km area and a population of 1.6 million people (see section 4.11). The mixing height is 250 m and dilution rate (the product of the mixing height and wind speed) is of 610 m² s⁻¹, based on an analysis (Marshall et al. 2005) of USEPA SCRAM mixing height data (USEPA 2002) for 75

urban areas. I employed harmonic means rather than arithmetic means because the urban one-compartment intake fraction is inversely proportional to dilution rate and mixing height (Marshall et al. 2005).

Rural. I represent rural areas with an average mixing height of 1,000 m and a wind speed of 3 m s^{-1} (Rosenbaum et al. 2008). When PM emissions occur in rural areas, the population within a few hundred kilometers is exposed (Levy et al. 2002, Greco et al. 2007 — often includes both rural and nearby urban areas), which is represented here by the globally averaged population density of inhabited regions of $90 \text{ persons km}^{-2}$.

Remote. Emissions in remote areas (e.g., ships in oceans, oil drilled in the middle of the desert or in the Arctic) generally have low intake fractions, as they occur far from areas with significant population density. Remote areas are evaluated as having a population density of one person per km^2 , which represents the approximate population density over a few million square kilometers in remote areas.

Emissions- and population-weighted arithmetic average. Most life-cycle inventories do not specify exactly where emissions occur. Several models indicate the country without specifying whether the emission took place in an urban, rural, or remote area within that country. In such cases, a generic intake fraction for an unknown emission location is needed. If an emission-weighted intake fraction was available for a specific context, then the emission-weighted intake fraction should be used. For cases when the emission-weighted intake fraction is unavailable, I suggest using the population-weighted intake fraction (see section 2.4 in Chapter 2 and section 0 in Chapter 3).

Table 4-2 summarizes the main parameters used in the models to generate the recommended intake fractions.

Table 4-2: Parameters used in the models to generate the recommended intake fractions.

Archetype	Urban	Rural	Remote
Worldwide characteristics	3.2 billion people, 2.3 million km^2	6.7 billion people ^a , 75 million km^2	75 million people, 73 million km^2
Parameters used for the generic archetype	80,000 people km^{-1} , 1.6 million people, 400 km^2	900 million people, 10 million km^2	10 million people, 10 million km^2
Meteorological parameters: mixing height, wind speed	250 m, 2.4 m s^{-1} (dilution rate of $610 \text{ m}^2 \text{ s}^{-2}$)	1,000 m, 3 m s^{-1}	1,000 m, 3 m s^{-1}
Other common parameters	breathing rate: $13 \text{ m}^3 \text{ person}^{-1} \text{ d}^{-1}$ (USEPA 1997); global average temperature: 285 K (Seinfeld and Pandis 1998); relative humidity: 70%		
^a All archetypes (including size and population for urban) are based on global data. However, because of data availability, urban meteorological conditions are only based on U.S. data.			
^b The rural archetype also takes into account the population of urban areas that will be exposed from rural emissions.			

4.5. Intake fractions

Comparison of available models and data

Several publications provide intake fractions for one or more of the emission archetypes (Hofstetter 1998, Van Zelm et al. 2008, Levy et al. 2002, Marshall et al. 2005, Greco et al. 2007, Rosenbaum et al. 2008, Heath et al. 2006, Preiss et al. 2008, Spadaro and Rabl 2004, Krewitt et al. 2001, Evans et al. 2002). When possible, values compared (Figure 4-2) were harmonized to correct for parameter differences using values in Table 4-2 (e.g., breathing rates were adjusted to $13 \text{ m}^3 \text{ person}^{-1} \text{ d}^{-1}$ — USEPA 1997). A complete list of literature-derived values, including pre-harmonized values, is presented in section 4.11.

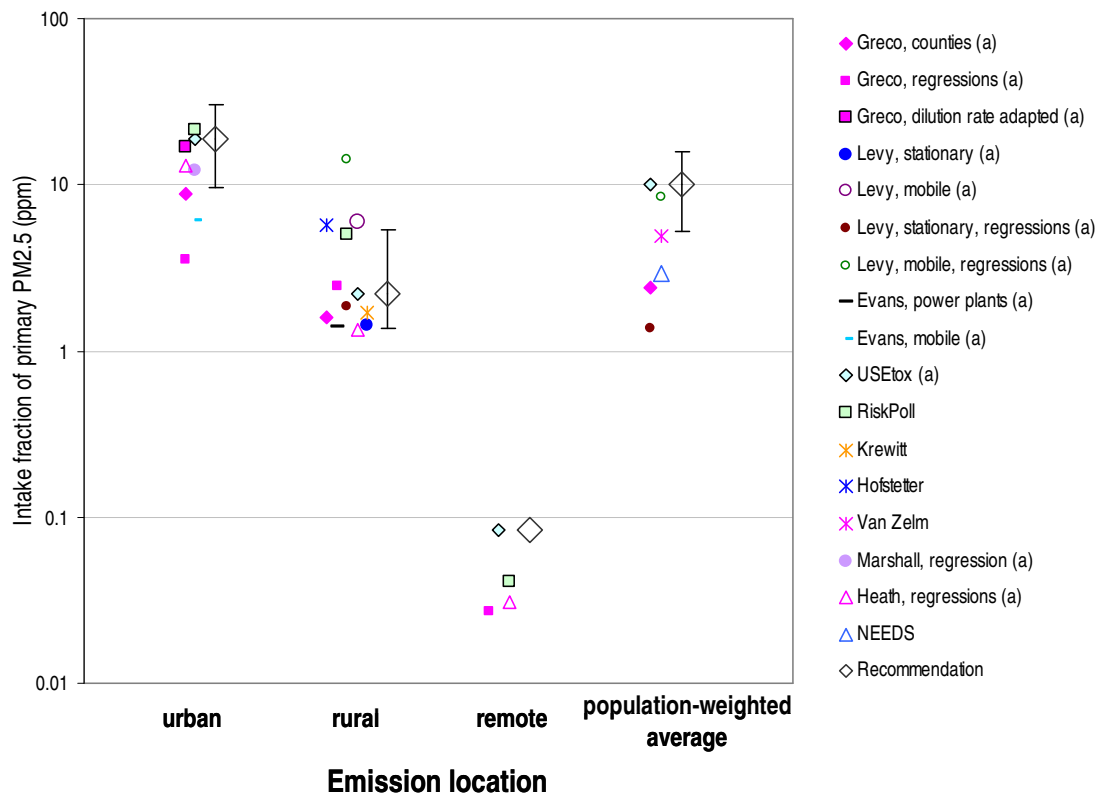


Figure 4-2: Intake fractions for different models and recommended values for primary PM_{2.5}. Values marked with (a) were adapted from the original model to a breathing rate of 13 m³ person⁻¹ d⁻¹ and to parameters for urban, rural, and remote areas. Recommended values are for an emission-weighted average stack height. Error bars show the range for high-stack and ground-level (the low-stack falling in between but not shown) emissions, assuming 41%, 17%, and 42% of total PM_{2.5} emissions are emitted from high-stack, low-stack, and ground-level sources, respectively (see section 4.11). Recommended values range includes high-stack, low-stack and ground-level emissions, and therefore extends beyond the displayed literature values, which are, in general, average values only.

Primary PM_{2.5}

Figure 4-2 indicates, for primary PM intake fraction, one order of magnitude variation between urban and rural areas and an even greater variation between rural and remote areas. Thus, the ability to differentiate between low and high population densities is likely at least as important in intake fraction assessment as the choice of model or method. Variations in intake fraction within an archetype are often linked to model limitations that could not be easily harmonized, such as meteorological dilution rates. USEtox (Rosenbaum et al. 2008) and Greco et al. (2007) give similar results for urban archetype when parameterized consistently, with central tendencies of 19 ppm and 17 ppm, respectively (see section 4.11). Those values are consistent with model- and measurement-based estimates of 14±7 ppm for United States urban ground-based emissions (Marshall et al. 2005). The USEtox rural intake fraction of 2.2 ppm is close to the value reported by Greco et al. (2007) of 2.5 ppm. For remote areas, models that can be adapted to low population density conditions give similar results, in the range 0.03 to 0.08 ppm.

Primary PM₁₀, primary PM_{10-2.5} and secondary PM

Figure 4-3 presents a summary of intake fractions for primary PM₁₀, PM_{10-2.5} and PM_{2.5}, and secondary PM from SO₂, NO_x, and NH₃ from different models. The numerical values are provided in section 4.11.

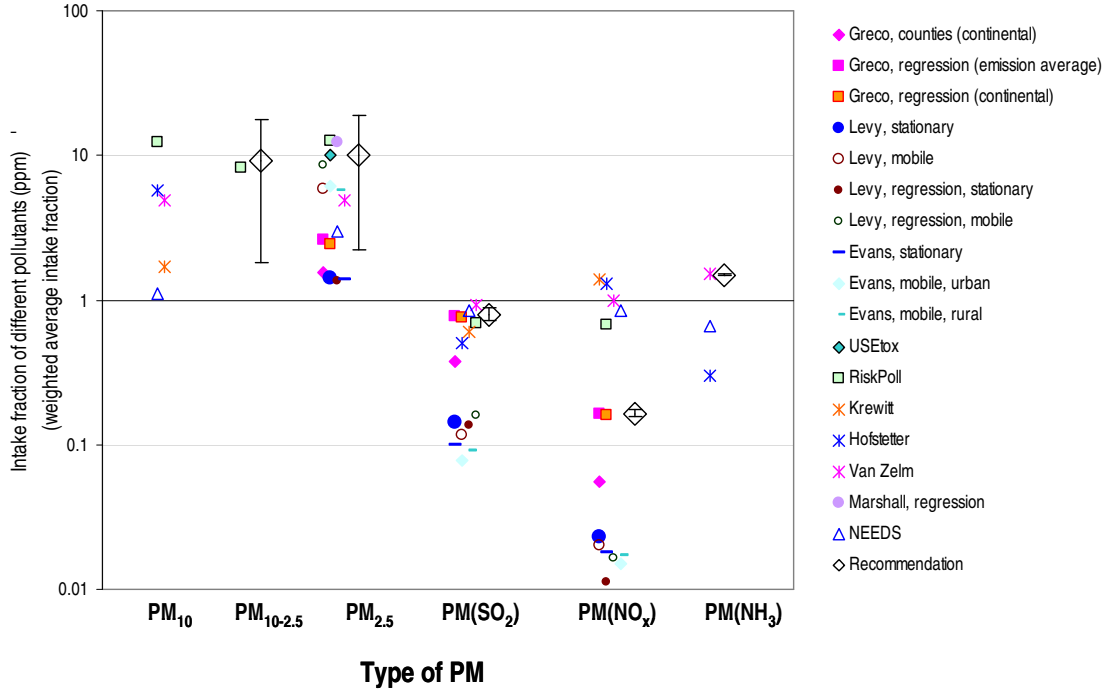


Figure 4-3: Intake fractions for different models and recommended values for different types of PM. Values are adapted to breathing rates of 13 m³ person⁻¹ d⁻¹. Recommended values are for an emission location and stack height weighted average intake fraction. Error bars represent variations depending on the location archetype of emission (from rural to urban). PM₁₀, PM_{10-2.5} and PM_{2.5} are primary PM and PM(SO₂), PM(NO_x) and PM(NH₃) are secondary PM.

Hofstetter (1998), Krewitt (2001), and Van Zelm et al. (2008) suggest no significant difference in the intake fractions of secondary PM from SO₂ and secondary PM from NO_x. In contrast, Levy et al. (2002), Evans et al. (2002), and Greco et al. (2007) suggest that the intake fraction of secondary PM from NO_x is lower than the intake fraction of secondary PM from SO₂. For Levy et al. (2002), this difference is derived primarily from dividing the nitrate concentrations results by a factor of four to reflect their assumption that nitrates only form during winter. Only Hofstetter (1998), Preiss et al. (2008) and Van Zelm et al. (2008) provide intake fractions for secondary PM from NH₃.

The share of PM₁₀ that is greater than 2.5 μm (i.e., PM_{10-2.5}, “coarse”) is generally removed from the environment faster than the PM below 2.5 μm (“fine”). For example, Seinfeld and Pandis (1998) report a U-shaped trend, where removal rates are rapid for large and for small particles, but intermediate sizes (generally, the accumulation mode, ~ 0.1–1 μm) experience slow removal. Because removal rates are typically faster for PM_{10-2.5} than for PM_{2.5}, average intake fractions are expected to be lower for PM_{10-2.5} than for PM_{2.5}.

The intake fraction of PM₁₀ can be expressed using Equation 4-4,

$$iF(PM_{10}) = f_{<2.5,e} \times iF(PM_{2.5}) + f_{10-2.5,e} \times iF(PM_{10-2.5}) \quad (4-4)$$

where $f_{<2.5,e}$ and $f_{10-2.5,e}$ are the mass fractions of emitted PM_{10} that are, respectively, smaller than $2.5 \mu m$ and between 2.5 and $10 \mu m$ at the time of emission. The factor $f_{<2.5,e}$ depends on the emission source. Average values in the United States for $f_{<2.5,e}$ are ~ 0.12 for transportation emissions (including off-road equipment and road dust), ~ 0.73 for tailpipe-only emissions from road transportation, ~ 0.73 for low-stack emissions and ~ 0.60 for high-stack emissions (see section 4.11). These ratios can vary and can be adapted to specific situations (see section 4.11).

Current evidence suggests that health damage per mass inhaled is lower for $PM_{10-2.5}$ than for $PM_{2.5}$ (Dockery et al. 1993, WHO 2006, EC 2005, Brunekreef and Forsberg 2005, USEPA 2009). Based on these differences in intake fraction and toxicity, the public health damage per mass emission is likely greater for $PM_{2.5}$ than for $PM_{10-2.5}$.

Note that further research is needed in this area. Indeed, Perez et al. (2009) show that all PM size fractions have health effects. However, they suggest that PM smaller than $2.5 \mu m$ and PM greater than $2.5 \mu m$ have different health outcomes. When calculating the human health damage from PM_{10} , Equation 4-4, that can be refined to allow consideration of the differences among sizes. By providing intake fractions for $PM_{10-2.5}$ I give the opportunity to have them used with effect factors for $PM_{10-2.5}$ once those become available.

Summary intake fraction values

Table 4-3 summarizes the above values by providing a matrix of internally-consistent estimates. Our goal is to provide the life-cycle assessment community with as complete and consistent a framework as possible, with recommended values for each archetypal environment. Whenever possible, I use a value directly from the relevant model, but equally importantly I choose values that are consistent within the matrix framework (e.g., the remote intake fraction should be smaller than the rural intake fraction, which should be smaller than the urban intake fraction).

Indoor. The intake fractions for indoor emissions are based on the approach in the previous section. Ventilation is likely the dominant elimination mechanism for the considered substances. Note that for $PM_{10-2.5}$, ventilation might actually not be the dominant removal mechanism as PM deposition through settling might be more important. Indeed, Thatcher et al. (2002) shows that for $PM_{10-2.5}$, $k_{settling}$ is between 1 and 10 per hour, whereas $k_{ventilation}$ used in Table 4-1 is between 0.5 and 1 per hour. However, because of the possible resuspension of settled PM, it is not given that settling can actually be considered a removal mechanism. More research would be needed to evaluate overall removal rate of $PM_{10-2.5}$. Using values in Table 4-1, intra-indoor intake fraction values by indoor emissions are 4,700 ppm for household, 1,600 ppm for office, and 33 ppm for industrial environments.

Urban. The urban intake fraction includes intra-urban, as well as global intakes attributable to the urban emissions. The urban intake fraction depends on the urban dilution rate. USEtox (Rosenbaum et al. 2008) uses a dilution rate based on a large set of urban data (see previous section), and has the advantage of ensuring a consistent treatment as for organic chemicals, which can be assessed using the same model parameterization. The intake fraction for an urban emission of primary PM from an unknown stack height is 19 ppm, as calculated by USEtox using the global average urban parameters in Table 4-2. USEtox can be easily adapted to specific urban (and rural) conditions.

For secondary PM from SO_2 and NO_x , the regression model by Greco et al. (2007) is employed here. It provides regressions that consider population densities at incremental distances from the emission and is consistent with USEtox for primary PM. Greco et al. (2007) provide appropriate regressions for estimating the intake fractions of these longer-range (several

hundreds of kilometers) pollutants, which are not significantly affected by the local population density and dilution rate. The intake fractions of secondary PM from urban ground-level emissions of SO₂ and NO_x are 0.88 ppm and 0.18 ppm, respectively. For secondary PM from NH₃, Van Zelm et al. (2008) indicate 1.5 ppm.

Rural. The intake fraction for rural emissions is based on the continental box in USEtox for primary PM_{2.5}, on the regressions of Greco et al. (2007) for secondary PM from SO₂ and NO_x, and on Van Zelm et al. (2008) for secondary PM from NH₃.

Remote. For primary PM, the intake fraction for remote emissions is based on the continental box in USEtox. For secondary PM, the intake fraction for remote emissions is based on the value obtained for rural areas and then using the ratio between intake fractions from rural and remote PM_{2.5} emissions given by USEtox.

PM_{10-2.5}. The intake fractions of PM_{10-2.5} are calculated as the USEtox-derived intake fractions of PM_{2.5} times the RiskPoll-derived (Spadaro and Rabl 2004) ratio of intake fractions of PM_{10-2.5} (section 4.11, Table 4-11) to intake fraction of PM_{2.5} (section 4.11, Table 4-10).

CO. The intra-urban intake fraction of CO, emitted in urban areas, is taken to be the same as the intra-urban intake fraction of PM_{2.5} emitted in urban areas (Marshall et al. 2005). Since the atmospheric residence time of CO (Jolliet and Crettaz 1997) is approximately 42 times greater than the PM_{2.5} residence time of 1.8 days used by USEtox, I calculated the intake fraction of CO emitted in rural and remote areas using an atmospheric life-time of 76 days. Intake fractions calculated with USEtox for rural and remote emissions are 22 ppm and 11 ppm, respectively. Note that because of the long atmospheric life-time of CO, the same limitation as for carbon tetrachloride applies (see section 2.4) regarding the potential overestimated intake fraction because of an underestimated mixing height, which is here chosen to be 1,000 m.

Emission release height. For the urban and rural emissions of primary PM_{2.5}, the USEtox results are used and correspond to an emission from an unknown stack height. Height-specific intake fractions are determined by ratios of modeled intake fractions from the different stack heights using RiskPoll (Spadaro and Rabl 2004). Because of higher urban population densities, the ratios between ground-level and low-stack (25 m), and between low-stack (25 m) and high-stack (100 m) intake fractions for urban emissions are larger than for rural emissions. The intake fraction ratio of ground-level to low-stack emissions is 2.2 for urban and 1.7 for rural conditions, and the intake fraction ratio of low-stack to high-stack emissions is 1.4 for urban and 1.3 for rural conditions.

Levy et al. (2002) found that the secondary particulate intake fraction does not differ significantly by source category. Therefore, the intake fractions of secondary PM from high- and low-stack emissions are assumed to be the same as the intake fraction of ground-level emissions. For remote emissions, differentiating among stack heights is also not necessary, since the air will generally be well-mixed before it reaches the exposed population.

Weighted arithmetic average. The average continental or rural intake fraction values should not be applied to emissions in unknown locations, because emissions are generally correlated to population size and therefore situated, on average, closer to urban areas than a continental intake fraction value would suggest. For this reason, the recommended weighted average intake fraction of an emission in an unknown location is calculated as a function of the urban, rural, and remote values:

$$iF_{p, \text{average location}} = \frac{1}{M_p} \times \sum_l (m_{p,l} \times iF_{p,l}) \quad (4-5)$$

where $iF_{p,average\ location}$ is the emission-weighted average intake fraction of a pollutant p ; $m_{p,l}$ and $iF_{p,l}$ are the mass and intake fraction, respectively, of pollutant p emitted in a location l ; and M_p is the total mass emitted. For outdoor emissions, Equation 4-5 can be simplified as

$$iF_{p,average\ location} \approx f_{p,e,urban} \times iF_{p,urban} + f_{p,e,rural} \times iF_{p,rural} + f_{p,e,remote} \times iF_{p,remote} \quad (4-6)$$

where $f_{p,e,urban}$, $f_{p,e,rural}$, and $f_{p,e,remote}$ represent the respective fractions of PM emissions occurring in urban, rural, and remote regions. As noted above, if mass-weighted values are unavailable, population-weighted values may provide a useful, albeit imperfect, approximation for average intake fraction. I set $f_{p,e,urban}$, $f_{p,e,rural}$, and $f_{p,e,remote}$ to 0.47, 0.52, and 0.01, representing global average population-based values. Note that secondary pollutants have very similar urban and rural intake fractions (10% to 20% variation); therefore, the actual ratio of emissions between these two archetypes does not significantly affect the population-weighted average intake fraction.

The emission-weighted average intake fraction developed for an unknown stack height is evaluated using Equation 4-7:

$$iF_{p,average\ stack} = f_{p,e,high-stack} \times iF_{p,high-stack} + f_{p,e,low-stack} \times iF_{p,low-stack} + f_{p,e,ground-level} \times iF_{p,ground-level} \quad (4-7)$$

where $f_{p,e,high-stack}$, $f_{p,e,low-stack}$, and $f_{p,e,ground-level}$ are the mass fraction of pollutant p emitted by high-stack, low-stack, and ground-level sources in the same geographical limits. Here, I estimated these fractions using data from USEPA (2008a) (see section 4.11 Table 4-8).

Table 4-3 summarizes the recommended intake fractions within a complete source-location framework. Section 4.11 summarizes the different assumptions behind each of the recommended intake fractions. The reason that I overlooked intake fraction of precursors for outdoor exposure is that damage from precursor emissions will be dominated by the damage from secondary PM. The damage from precursors' direct intake will be small in comparison to the damage from secondary PM. However, this is a limitation that would require some further analysis to validate this simplification.

Table 4-3 : Summary of recommended intake fractions (ppm) for PM_{10-2.5}, PM_{2.5}, SO₂, NO_x, NH₃, and CO.

Pollutant emitted	Type of emission source	Intake fractions for the respective location of emission:							Unit
		Indoor ^a (household)	Indoor ^a (office)	Indoor ^a (industrial)	Urban	Rural	Remote	Population-weighted average (only for outdoor emissions)	
PM ₁₀	High-stack ^b	4,700	1,600	33	8.3	0.93	0.055	4.4	10 ⁻⁶ kg PM ₁₀ inhaled/kg PM ₁₀ emitted
	Low-stack				13	1.4	0.055	6.6	
	Ground-level				20	2.1	0.055	1.0	
	Emission-weighted average				18	1.9	0.055	9.3	
PM _{10-2.5}	High-stack ^b	4,700	1,600	33	8.5	0.88	0.034	4.4	10 ⁻⁶ kg PM _{10-2.5} inhaled/kg PM _{10-2.5} emitted
	Low-stack				13	1.3	0.034	6.6	
	Ground-level				18	1.9	0.034	9.6	
	Emission-weighted average				17	1.8	0.034	9.1	
PM _{2.5}	High-stack ^b	4,700	1,600	33	9.6	1.4	0.084	5.2	10 ⁻⁶ kg PM _{2.5} inhaled/kg PM _{2.5} emitted
	Low-stack				14	1.8	0.084	7.4	
	Ground-level				30	3.1	0.084	16	
	Emission-weighted average				19	2.2	0.084	10 ^c	
SO ₂		4,700	1,600	33	0.88	0.72	0.044	0.79 ^c	10 ⁻⁶ kg secondary PM ^d (or SO ₂ for indoor) inhaled/kg SO ₂ emitted
NO _x		4,700	1,600	33	0.18	0.16	0.010	0.16 ^c	10 ⁻⁶ kg secondary PM ^d (or NO _x for indoor) inhaled/kg NO _x emitted
NH ₃		4,700	1,600	33	1.5	1.5	0.090	1.5 ^c	10 ⁻⁶ kg secondary PM ^d (or NH ₃ for indoor) inhaled/kg NH ₃ emitted
CO	High-stack ^b	4,700	1,600	33	30	22	11	26	10 ⁻⁶ kg CO inhaled/kg CO emitted
	Low-stack				34	22	11	27	
	Ground-level				49	22	11	35	
	Emission-weighted average				47	22	11	34 ^c	

^a The intake fraction for indoor emissions includes only indoor exposure. For the total intake fraction for indoor emissions, one should add the intake fraction for outdoor emissions (in general as a low-stack emission) of the archetype where the building is situated.

^b To estimate the intake fractions of very high-stack emissions (>250 m), values from RiskPoll (35) show that the high-stack (100 m) intake fraction can be multiplied by 0.55 and 0.79 for urban and rural emissions, respectively (see section 4.11).

^c The grey cell is a emission-weighted average of the different stack heights of a population-weighted average intake fraction of the different archetypes.

^d SO₂ can form ammonium sulfate and NO_x and NH₃ can form ammonium nitrate.

4.6. Effect factors

The inhalation of PM can lead to many different health outcomes, and existing studies show significant variations in the frequency and estimated damage of each outcome as a function of mass PM inhaled. A complete analysis of effect factor determination is not within the scope of this chapter. This section details a preliminary literature review and outlines a method for determining recommended effect factors. This section leads to preliminary effect factor values for each pollutant.

Computation of the effect factor of PM. I derive effect factors by endpoint in a consistent fashion by reporting both the number of cases of different diseases (i.e., the dose-response factor) as well as the DALYs (i.e., the severity factor). For many endpoints, the concentration–response curve is found to be or assumed to be linear (WHO 2006, Schwartz et al. 2008). Most epidemiology studies have been done using PM₁₀; therefore the computation of the effect factors is done based on PM₁₀. The effect factors for primary PM_{2.5}, secondary PM from SO₂, secondary PM from NO_x, and secondary PM from NH₃ are then derived from the effect

factors computed for PM₁₀. Table 4-4 summarizes the type of endpoint, dose-response, severity, and effect factors, as well as the sources used to calculate the provisional effect factors of PM₁₀.

Table 4-4 : Evaluation of dose-response, severity, and effect factors of PM₁₀.

Type of endpoint	Dose-response factor (case/kg _{inh})	Source/ comment	Severity factor (DALY/ case)	Source/ comment	Effect factor (DALY/ kg _{inh})	Source/ comment
Chronic mortality ^a	5.76	Van Zelm et al. (2008) (RR based on Kuenzli et al. (2000) and F _{inc} on European Commission (2007))	10	Van Zelm et al. (2008) (based on Kuenzli et al. (2001) and Pye and Watkiss (2005)) Note that Bare et al. (2003) uses 10.9 DALY/case based on De Hollander et al. (1999)	57.6	Van Zelm et al. (2008) (For comparison, Torfs et al. (2007) uses 82.2 DALY/kg _{inh})
Acute respiratory morbidity	0.73	Van Zelm et al. (2008) (RR based on Medina et al. (2005) and F _{inc} on Knol and Staatsen (2005))	0.025	Van Zelm et al. (2008) (based on Knol and Staatsen (2005))	0.018	Van Zelm et al. (2008)
Acute cardiovascular morbidity	0.55	Van Zelm et al. (2008) (RR based on Le Tertre et al. (2002) and F _{inc} on Knol and Staatsen (2005))	0.027	Van Zelm et al. (2008) (based on Knol and Staatsen (2005))	0.015	Van Zelm et al. (2008)
Chronic bronchitis (adults)	9.5	Kuenzli et al. (2000)	2	Hofstetter (1998)	19	
Chronic bronchitis (children)	140	Kuenzli et al. (2000)	0.025	Hofstetter (1998)	3.6	
Restricted activity days	6100	Kuenzli et al. (2000)	2.7E-4	Hofstetter (1998)	1.7	
Asthmatics: asthma attacks (children)	56	Kuenzli et al. (2000)	2.7E-4	Hofstetter (1998)	0.015	
Asthmatics: asthma attacks (adults)	140	Kuenzli et al. (2000)	2.7E-4	Hofstetter (1998)	0.037	
				TOTAL:	82	DALY/kg inh PM ₁₀

^a Here, chronic refers to the temporal pattern of exposure and mortality to the outcome.

The total effect factor for PM exposure accounts for premature mortality and other endpoints such as asthma and restricted activity days. Premature mortality (“chronic mortality”) is referring to the mortality associated with chronic diseases. Note that premature mortality also includes short-term increases in mortality (“acute mortality”) from respiratory effects, as well as long-term mortality from carcinogenic effects. Acute data are based on time-series studies on daily mortality that measure the proportional increase in the daily death rate attributable to recent exposure to air pollution. Chronic data are based on cohort studies. Chronic data include those who died from chronic disease caused by long-term exposure, but also those whose death is advanced by recent exposure to air pollution (Kuenzli et al. 2000, WHO 2006, Van Zelm 2009)

By combining all outcomes, I find a final effect factor of approximately 82 DALYs/kg PM_{10,inhaled}.

The effect factor of primary PM_{2.5} is derived from the effect factor for primary PM₁₀ by assuming that 60% of the ambient mass of PM₁₀ in the studies reviewed was smaller than 2.5 μm (Dockery et al. 1993). This approach is based on the assumption that most health effects are attributed to PM smaller than 2.5 μm in size (Hofstetter 1998, WHO 2006, Dockery et al. 1993, EC 2005, Brunekreef and Forsberg 2005, USEPA 2009). This assumption is debated and more

research is needed to better address the effect factor of PM_{10-2.5}. Effect factors of secondary PM are difficult to evaluate as they are often correlated with primary PM. Reiss et al. (2007) suggest that toxicological evidence does not support a causal association between particulate nitrate and sulfate compounds and excess health risks. Hofstetter (1998) suggests that secondary PM from NO_x are better represented by PM₁₀ and secondary PM from SO₂ are better represented by PM_{2.5}. The ExternE project (EC 2005) recommends treating primary PM_{2.5} as 1/0.6 times the toxicity of primary PM₁₀, sulfates as equivalent to the toxicity of PM₁₀ (or 0.6 times PM_{2.5}), nitrates as equivalent to 0.5 times the toxicity of PM₁₀, primary particles from power stations (i.e., high-stack) as equivalent to PM₁₀, and primary particles from vehicles as equivalent to 1.5 times the toxicity of PM_{2.5}. The NEEDS project (Preiss et al. 2008, Torfs et al. 2007), a follow up of the ExternE project, in accordance with the work of the World Health Organization (WHO 2006) and the early findings of Milford and Davidson (1987), suggest that secondary inorganic aerosols are equally toxic to primary PM, but not necessarily equally toxic to PM₁₀. It recommends treating primary sulfates as equivalent to the toxicity of PM_{2.5} and nitrates as equivalent to the toxicity of PM₁₀. Note that this assumption is subject to discussion and further research is certainly needed in this area.

Effect factors of SO₂, NO_x, NH₃ and CO. Hofstetter (1998) provides effect factors for SO₂, NO_x and CO of, respectively, 0.97, 0.46, and 0.019 DALY per kg of SO₂, NO_x and CO inhaled (with breathing rate adjusted to 13 m³ person⁻¹ y⁻¹). The effect factor for direct inhalation of NH₃ is estimated using the method suggested by Pennington et al. (2002) for non-cancer effects, the non-observed adverse effect level of 2.3 mg m⁻³ suggested by USEPA's IRIS database (USEPA 2008b), and an estimated damage factor of 2.2 DALY/case (based on Huijbregts et al. (2005a) for respiratory diseases). For outdoor emissions, the effect factors of SO₂, NO_x and NH₃ should be added to the effect factors evaluated for secondary particles from SO₂, NO_x and NH₃ emissions. Indoor emissions of SO₂, NO_x, and NH₃ can be assumed to have no time to transform into secondary PM before leaving the building and thus only effect factors caused by direct inhalation of SO₂, NO_x, and NH₃ should be accounted for when calculating the damage occurring within the building.

A summary of the effect factors of primary PM₁₀, primary PM_{2.5}, secondary PM from SO₂, NO_x, and NH₃, and direct intake of SO₂, NO_x, NH₃ and CO is given in Table 4-5.

Table 4-5 : Summary of interim effect factors for exposure to various types of PM and other respiratory inorganics.

Pollutant	Abbreviation	EF	Unit	Comment
Primary PM ₁₀	PM ₁₀	82	DALY/kg PM ₁₀ inhaled	based on Van Zelm et al. (2008), Kuenzli et al. (2000), and Hofstetter et al. (1998), see Table 4-4
Primary PM _{2.5} (undefined)	PM _{2.5}	140	DALY/kg PM _{2.5} inhaled	extrapolated from PM ₁₀ by dividing by 0.6 (assumed fraction of ambient PM ₁₀ smaller than 2.5 µm) (EC 2005)
Primary PM _{2.5} (ground-level) ^a	PM _{2.5, ground-level}	200	DALY/kg PM _{2.5, ground-level} inhaled	assumed equivalent to 1.5 × PM _{2.5} (EC 2005)
Secondary PM from SO ₂	PM(SO ₂)	140	DALY/kg PM(SO ₂) inhaled	assumed equivalent to PM _{2.5} (EC 2008)
Secondary PM from NO _x	PM(NO _x)	82	DALY/kg PM(NO _x) inhaled	assumed equivalent to PM ₁₀ (EC 2008)
Secondary PM from NH ₃	PM(NH ₃)	82	DALY/kg PM(NH ₃) inhaled	assumed equivalent to PM ₁₀ (EC 2008)
SO ₂ (direct exposure)	SO ₂	0.97	DALY/kg SO ₂ inhaled	acute mortality, Hofstetter (1998)
NO _x (direct exposure)	NO _x	0.46	DALY/kg NO _x inhaled	acute mortality, Hofstetter (1998)
NH ₃ (direct exposure)	NH ₃	0.84	DALY/kg NH ₃ inhaled	based on Pennington et al. (2002), USEPA (2008b), and Huijbregts et al. (2005a)
CO (direct exposure)	CO	0.019	DALY/kg CO inhaled	acute mortality, Hofstetter (1998)

^a i.e., mainly tailpipe emissions – road dust emissions might be better represented by undefined values – would require further research.

^b Relevant further resources are Harrison et al. (1983), Hering et al. (1997) and Huang et al. (2004).

The exact mode of action of PM in the body is still not well understood. It has been suggested that reactive oxidant stress is a major determinant in the health effects of PM (Donaldson et al. 2001, Dick et al. 2003), but it has not yet been determined whether these impacts are from a physical effect of PM or from the adsorption of other organic or inorganic substances on to the particulates (in which case particle surface areas would be a better proxy than mass to evaluate the adverse health effects).

Since the human health impacts by secondary PM are still being debated by human health experts, the values for effect factors, and characterization factors associated with secondary PM provided here should be considered with care.

4.7. Characterization factors

Characterization factor of PM_{10} and PM_{tot}

The characterization factor of PM_{10} is computed using Equation 4-1 and Equation 4-4:

$$CF(PM_{10}) = iF(PM_{10}) \times EF(PM_{10}) = f_{<2.5,e} \times iF(PM_{2.5}) \times EF(PM_{2.5}) + f_{10-2.5,e} \times iF(PM_{10-2.5}) \times EF(PM_{10-2.5}) \quad (4-8)$$

Assuming $iF(PM_{10-2.5}) < iF(PM_{2.5})$ (coarse particles are removed faster from the atmosphere than fine particles —, Seinfeld and Pandis 1998) and $EF(PM_{10-2.5}) \ll EF(PM_{2.5})$ (Hofstetter 1998, Dockery et al. 1993, EC 2005, Brunekreef and Forsberg 2005, USEPA 2009), then $f_{10-2.5,e} \times iF(PM_{10-2.5}) \times EF(PM_{10-2.5})$ can be considered substantially smaller compared to $f_{<2.5,e} \times iF(PM_{2.5}) \times EF(PM_{2.5})$, and:

$$CF(PM_{10}) \approx f_{<2.5,e} \times iF(PM_{2.5}) \times EF(PM_{2.5}) \quad (4-9)$$

The same approach can be applied for the total amount of particle (PM_{tot}) in case this elementary flow is reported in life-cycle inventories:

$$CF(PM_{tot}) \approx f_{<2.5,emissions\ of\ PM\ tot} \times iF(PM_{2.5}) \times EF(PM_{2.5}) \quad (4-10)$$

Interim characterization factors of PM_{10} , $PM_{10-2.5}$, $PM_{2.5}$, SO_2 , NO_x , NH_3 and CO

Interim characterization factors (Table 4-6) are evaluated by applying Equation 4-1 to the factors in Table 4-3 and Table 4-5.

Table 4-6: Summary of interim characterization factors (microDALY/kg_{emitted}) for PM₁₀, PM_{10-2.5}, PM_{2.5}, SO₂, NO_x, NH₃ and CO.

Pollutant emitted	Type of emission source:	Characterization factor for the respective location of emission:							Unit
		Indoor ^a (household)	Indoor ^a (office)	Indoor ^a (industrial)	Urban	Rural	Remote	Population-weighted average (only for outdoor emissions)	
PM ₁₀ ^b	High-stack	610,000	210,000	4,300	810	120	7.0	440	microDALY/kg PM ₁₀ emitted
	Low-stack				1,400	190	8.5	760	
	Ground-level				730	76	2.0	380	
	Emission-weighted average				780	88	3.2	410	
PM _{10-2.5}	High-stack	n/a ^c	n/a ^c	n/a ^c	n/a ^c	n/a ^c	n/a ^c	n/a ^c	microDALY/kg PM _{10-2.5} emitted
	Low-stack				n/a ^c	n/a ^c	n/a ^c	n/a ^c	
	Ground-level				n/a ^c	n/a ^c	n/a ^c	n/a ^c	
	Emission-weighted average				n/a ^c	n/a ^c	n/a ^c	n/a ^c	
PM _{2.5}	High-stack	660,000	220,000	4,600	1,400	190	12	730	microDALY/kg PM _{2.5} emitted
	Low-stack				1,900	260	12	1,000	
	Ground-level				6,000	630	17	3,100	
	Emission-weighted average				3,400	390	14	1800	
SO ₂		4,600	1,600	32	120	100	6.2	110	microDALY/kg SO ₂ emitted
NO _x		2,200	740	15	14	13	0.79	13	microDALY/kg NO _x emitted
NH ₃		4,000	1,300	28	120	120	7.6	120	microDALY/kg NH ₃ emitted
CO	High-stack	89	30	0.63	0.58	0.42	0.21	0.49	microDALY/kg CO emitted
	Low-stack				0.65	0.42	0.21	0.52	
	Ground-level				0.93	0.42	0.21	0.66	
	Emission-weighted average				0.90	0.42	0.21	0.64	

^a The characterization factor for indoor emissions includes only direct effects from indoor exposure. For the total characterization factor for indoor emissions, one should add the characterization factor for outdoor emissions (in general as a low-stack emission) of the archetype where the building is situated.

^b The characterization factor of PM₁₀ is calculated according to Equation 4-8, assuming the fractions of PM₁₀ smaller than 2.5 µm and between 2.5 µm and 10 µm indicated in section 4.11. The effect factor of PM_{10-2.5} is assumed to be substantially lower compared to those of PM_{2.5} and has therefore been disregarded (see section 4.6). Shall the user know her or his own particle size distribution, she or he should use it with Equation 4-8 to recalculate the characterization factor of PM₁₀ provided in Table 4-7

^c To be taken with care. Seems to be significantly lower than values for PM_{2.5} and PM₁₀. Further research needed.

Most life cycle inventories and life-cycle assessments are still performed without knowledge of the type of source and location of PM emissions. In these cases, the weighted average interim characterization factor would be used (grey cells in Table 4-7). When the type of source and its location are known for foreground processes (i.e., the processes directly evaluated in the life-cycle assessment), the characterization factor for the respective source and location should be used. The interim characterization factors suggested in Table 4-7 use the archetype approach to better assess human health damage from regionalized emissions of PM.

4.8. Discussion

Variability and uncertainty

Accounting for the emission-specific population density reduces the variability (not the uncertainty) of the estimated intake fraction and characterization factor, which in turn reduces the uncertainty of the life-cycle assessment results. Variability becomes an uncertainty if it is not accounted for in the calculation. One of the main constraints in life-cycle assessment regarding

regionalization is that most of the inventories of background processes do not give information (or even do not provide the option to give information) on the country of emission, providing only information on the archetype (e.g., high or low population density) where emissions occur. Certain specific processes may include the country of origin, but this information is then lost when the life-cycle assessment software aggregates inventories before performing the impact assessment. Since aggregated inventories are still distinguished by archetype, the recommended intake fractions and characterization factors presented here can be directly used within current life-cycle assessment constraints (including life-cycle assessment software).

Uncertainty and the importance of spatial differentiation

In discussing impacts of PM, there are many sources of variability and uncertainty along the emission-to-impact chain.

The intake fraction source-location matrix addresses many of the key sources of uncertainty. Differentiation among emissions in urban, rural, and remote areas is a key factor for the fate and exposure of primary and secondary PM, which are also strongly influenced by the height of emissions. When the available information allows, accounting for archetypes will reduce the uncertainty of the final life-cycle assessment results. Intake fraction varies with mixing height and wind speed. I employed here the harmonic mean dilution rate to estimate urban intake fractions (Marshall et al. 2005), recognizing that future work could usefully compare and evaluate methods for summarizing meteorological data. Also, currently, I am relying on meteorological data for United States conditions in the modeling of global intake fractions. Future work is needed to use local data in order to better adapt fate and exposure modeling for other continents.

This chapter focuses on the transport of, and exposure to, PM; rigorous investigation of health effects, dose-response, and mechanisms of impact were outside of our scope. However, additional information of health effects may influence how fate, transport, and exposures are evaluated. For example, PM regulations and epidemiology studies typically focus on PM mass; if PM number, area, or speciation were shown to be important and robustly quantifiable in dose-response relationships, then it would be necessary to reevaluate results presented here. In addition, the use of epidemiological data means that PM-attributed impacts can in fact be from other pollutants whose concentrations could be correlated to PM (Reiss et al. 2007). Care must be taken to avoid double-counting the impacts of PM and the impact of other correlated variables in cases of common endpoints.

I estimate the uncertainty of the source-location characterization factor matrix by examining the variability among the existing models. Setting the same population density and breathing rate in all emissions-to-intake models, and assuming lognormal distributions, I estimate the square geometric standard deviations (GSD^2) of the factors contributing to characterization factors. The GSD^2 means that 95% of the values fall between the median divided by the GSD^2 and the median times the GSD^2 . It provides the upper and lower bounds of the distribution, leaving a 2.5% tail on each side. The GSD^2 of the intake fraction (iF) within a given archetype is approximately 3.3, while the GSD^2 of the interim dose-response factor (DR) is evaluated to be approximately a factor of 1.4 and the GSD^2 of the interim severity factor (SF) evaluated to be another factor of 1.4. If uncertainties in intake fraction, dose-response and severity factors are uncorrelated, the characterization factor has a GSD^2 of 3.6 (Equation 4-8).

$$GSD_{CF}^2 = e^{\sqrt{(\ln GSD_{iF}^2)^2 + (\ln GSD_{DR}^2)^2 + (\ln GSD_{SF}^2)^2}} \quad (4-8)$$

For the pollutants considered here, this modeling uncertainty is smaller than the variation in intake fraction caused by population density patterns, which can be up to two orders of magnitude between emissions in high population density areas (e.g., truck emissions in a city) and low density population areas (e.g., emissions from a diesel train crossing remote areas). Therefore, regionalizing characterization factors by considering variability in population density patterns is an essential step towards the reduction of overall variability and uncertainty in evaluating human health damage when using life-cycle assessment.

Outlook

The matrices of factors that I present above provide a consistent framework for life-cycle assessment practitioners to evaluate the fate, exposure, effect, and damage of primary and secondary PM. The recommended intake fractions and interim characterization factors allow life-cycle assessment models to include information on both the type of source and the location of the PM emission.

However, given the uncertainties outlined above, further research is needed to improve the evaluation of PM within life-cycle assessment. Research should focus on two areas of key topics: better understanding of the fate and exposure of PM and of the effects.

First, further improvements would come with the optimal averaging method of wind speeds and mixing heights, which should be determined to best estimate intake fractions for rural and remote emissions. The influence of the season on the fate and exposure of PM should be evaluated, as the season has a large impact on mixing height, transport, and deposition (Ries et al. 2009). The spatial differentiation of fate and exposure needs to be improved to capture emissions in other types of environments (such as oceans or high altitudes), which would involve the evaluation of intake fractions and characterization factors for different geographical regions in the world not evaluated in this chapter. Furthermore, fate, exposure and effects from secondary PM from volatile organic compounds need to be quantified. To further evaluate the influence of composition and size distribution, the intake fraction and effect factor should be differentiated depending on the PM source such as diesel, coal or road dust.

Since chronic bronchitis in adults accounts for one-third of the PM effect factor, this aspect should be assessed with higher certainty. The influence of PM inhalation on low birth weight (Bell et al. 2008) and expressing it in terms of DALYs also deserves further attention. Furthermore, dose-responses from chronic exposure to precursors, CO, and secondary particulate matter from NH₃ require greater understanding. Finally, characterization factors, both for PM smaller than 2.5 μm (e.g., PM₁ or PM_{0.1}) and for PM between 2.5 and 10 μm, should be studied further.

Throughout this dissertation I have not considered the influence of buildings in my modeling of the fate, exposure or effect factors. Buildings can change the particle size distribution and exposure to PM from outdoor origin (Riley et al. 2002, Liu and Nazaroff 2003) as well as the interpretation of epidemiological data.

Because of the uncertain mode of action for PM, modeling of effect factors may consider surface area and number of particles instead of only mass as a proxy for adverse health effects. Similarly, fate and exposure modeling may consider the evolution of particle size distribution.

Among the different issues raised here, this dissertation will tackle the latter two (i.e., surface area and number of particles as well as particle size distribution) in detail in Chapters 6, 7 and 8.

While these recommended intake fractions and interim characterization factors are still in need of further work, the source-location matrices suggested in the present chapter provide a

framework for life-cycle assessment practitioners to improve their evaluations of adverse health effects caused by primary and secondary particulate matter. In many damage-oriented life-cycle assessment studies, PM is responsible for a large or dominant fraction of the total human health damage. Harmonizing the values used in life-cycle assessment studies and making those values consistent with the characterization of organics (Rosenbaum et al. 2008, Hauschild et al. 2008) will increase accuracy, consistency, and comparability among results for human health damage. This significant improvement in the quality of human health damage quantification will foster the assessment of human health impacts in life-cycle assessment.

In Chapters 5, 9 and 10, I will apply these different characterization factors to different processes to evaluate the influence of regionalization in life-cycle assessment.

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4.11. Supporting information for this chapter

In the following, I first describe the archetype parameterization (section 4.11.1) and characterize the emission sources (section 4.11.2). I then detail the intake fraction calculation (section 4.11.3), providing information on the regression models, the values used for the charts comparing the intake fractions and the formulas used to calculate the recommended values.

4.11.1. Archetype parameterization

Breathing rate

Different models use different breathing rates, varying between 9.5 (Phonboon 1996) and 25 m³ person⁻¹ d⁻¹ (mentioned but not use in Marshall 2005). Because I aim to calculate an intake fraction representative for the average population, I suggest using the population average breathing rate of 13 m³ person⁻¹ d⁻¹ (USEPA 1997), which is applied to all intake fractions calculated in this chapter, unless specifically mentioned.

Indoor environment parameterization

No strong evidence was found to justify amplification for indoor pollutant exposure above room average conditions. Therefore, in Equation 4-3 in the main body of the chapter, the mixing factor m is assumed to be 1. Additional information can be found in Hellweg et al. (2005) and Meijer et al. (2005a, 2005b).

“World city” parameterization

The intra-urban intake fraction (iF) of a pollutant emitted in an urban area i is computed as:

$$iF_i = \frac{a \times BR \times N_i}{u_i \times H_i \times W_i} = \frac{a \times BR}{u_i \times H_i} \times d_i \times L_i, \text{ with } d_i = \frac{N_i}{L_i \times W_i} \quad (4-9)$$

where N (persons) is the number of persons in the urban area; BR (m³ person⁻¹ d⁻¹) is the average breathing rate; a (unitless) is the correction factor to account for the fact that (i) a pollutant can be emitted anywhere in the urban area i and not only along the periphery, and (ii) the air that left the urban area i can return with some of the pollutant (i.e., a back-and-forth movement of air) (the factor a varies between 0.5 and 1, and can be approximated to be 0.75 — Benarie 1998); u (m d⁻¹) and H (m) are the dominant wind speed and mixing height of the urban area; L (m) is the length of the urban area (measured in the direction of the dominant wind); W (m) is the width of the urban area (often, the urban area is assumed to be a square, thus $L = W$); and d (persons m⁻²) is the population density of the urban area. Equation 4-9 assumes that deposition and degradation rates within the urban area are negligible relative to advection out of the area.

Note that the value of the correction factor a can be discussed. For example, the coherence between plume modeling (Heath et al. 2006) and well-mixed box modeling (Marshall et al. 2005) for ground-based emissions in south California suggests that a is close to 1

Equation 4-9 shows that the intake fraction is proportional to the term $d \times L$ (persons m⁻¹), defined as the linear population density (Marshall et al. 2005, Marshall 2007). The term $u \times H$ (m² d⁻¹) is the dilution rate. The dilution rate is 610 m² s⁻¹, calculated as the harmonic average of the mixing height and wind speed of 75 urban areas (based on Marshall et al. (2005) analysis of USEPA SCRAM mixing height data (USEPA 2002)).

The emission-weighted average intra-urban intake fraction $iF_{average}$ of a pollutant over all urban areas is computed as:

$$iF_{average} = \frac{\sum_i m_i \times iF_i}{\sum_i m_i} = \frac{a \times BR}{u \times H} \times \frac{\sum_i m_i \times d_i \times L_i}{\sum_i m_i} = \frac{a \times BR}{u \times H} \times d_{average} \times L_{average} \quad (4-10)$$

where m_i is the mass emitted in urban area i and $d_{average} \times L_{average}$ is the equivalent linear population density representative of the average of all the urban areas:

$$d_{average} \times L_{average} = \frac{1}{\sum_i m_i} \times \sum_i m_i \times d_i \times L_i \quad (4-11)$$

If I assume that emissions are proportional to population, then:

$$d_{average} \times L_{average} = \frac{1}{\sum_i N_i} \times \sum_i N_i \times d_i \times L_i \quad (4-12)$$

The total urban population is $\sum_i N_i = 3.15 \times 10^9$ persons (year 2005 — UN 2008). Using the United Nations Statistics Division data (UNSD 2008) as well as the World Bank data (Angel et al. 2005), I find that $\sum_i N_i \times d_i \times L_i \approx 2.6 \times 10^{14}$ persons² km⁻¹, giving an average linear population density of $d_{average} \times L_{average} \approx 80,000$ persons km⁻¹ = 80 persons m⁻¹. Considering the different sources of uncertainty, the uncertainty in the global population-weighted average linear population density value is estimated to be approximately 25%.

With these values, the simple model presented in Equation 4-9 produces an average intra-urban intake fraction $iF_{average}$ of 15 ppm (Equation 4-13):

$$iF_{average} = \frac{a \times BR}{u \times H} \times d_{average} \times L_{average} = \frac{0.75 \times 13}{610 \times 3600 \times 24} \times 80 = 15 \text{ ppm} \quad (4-13)$$

Apte (2008) estimated the population-weighted median intraurban intake fraction for the world's 50 largest megacities to 80 ppm (to be taken with care as this work is being currently reviewed by the author himself). This may suggest that 15 ppm is an underestimate of the global urban average intake fraction for ground-level releases of non-reactive species. This possible underestimation could be caused by an underestimation of the linear population density in Equation 4-13 as well as an overestimate of dilution when assumed mixed instantly or overestimation of the urban dilution rate.

In this dissertation, the linear population density of the default world city is set as 80,000 persons km⁻¹, that can be represented by a population density of 4,000 persons km⁻² over an area of 20 km × 20 km. These parameters represent a total population of 1,600,000 persons over 400 km².

Parameterizations by continent

Table 4-7 summarizes the parameters by continent and for the world.

Table 4-7 : Summary of the parameters by continent and for the world.

Parameter	World	Generic continent	North America	South America	Europe	Africa	Middle-East (incl. Egypt and Turkey)	Central Asia (incl. Asian Russia)	Asia (continental)	South Asia (Indian sub-continent)	East Asia (China w/o east, Japan, Korea, South East)	Australia/Oceania	Antarctica
Size, actual (km ²)	150 M	-	24 M (incl. Greenland and central America)	18 M	10 M	30 M	~7 M	~20 M	44 M	18 M	~10 M	8.5 M	14 M
Population (persons)	6.7 B	-	520 M	370 M	710 M	920 M	~350 M	~50 M	3.8 B	~1.7 B	~2 B	30 M	1,000
Population density (persons/km ²)	45	-	21	21	70	29	50	2.5	87	340	200	3.5	0.0001
“Continental” size (km ²) ^a	77 M (urban + rural)	10 M	~10 M (US + south Canada)	18 M	10 M	20 M	~7 M	~20 M	-	~5 M	~10 M	~10 M	14 M
“Continental” population (persons) ^a		900 M	~350 M	370 M	710 M	700 M	~350 M	~50 M	-	~1.7 B	~2 B	~40 M	0
“Continental” population density (persons/km ²) ^a (urban + rural)		90	35	21	71	35	50	2.5	-	340	200	4	0
Urban population (persons)	3.2 B	-	-	-	-	-	-	-	-	-	-	-	-
Urban size (km ²)	2.3 M	-	-	-	-	-	-	-	-	-	-	-	-
Urban linear population density (persons/km) (see Equation 4-14)		80,000	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	-
Urban population density (persons/km ²)		4,000	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	-
Urban length (km) ^b		20	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	-
a_i (see Equation 4-14)		0.47	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	-
b_i (see Equation 4-14)		6.5	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	n/a	-

Parameter	World	Generic continent	North America	South America	Europe	Africa	Middle-East (incl. Egypt and Turkey)	Central Asia (incl. Asian Russia)	Asia (continental)	South Asia (Indian sub-continent)	East Asia (China w/o east, Japan, Korea, South East)	Australia/Oceania	Antarctica
4-14)													
Rural population (persons)	3.5 B	-	-	-	-	-	-	-	-	-	-	-	-
Rural size (km ²)	75 M (50%)	-	-	-	-	-	-	-	-	-	-	-	-
Rural population density (persons/km ²)	50		~20	~10	~35	~20	~25	~1		~170	~100	~2	
Remote population (persons)	73 M	10 M	-	-	-	-	-	-	-	-	-	-	-
Remote size (km ²)	73 M (50%)	10 M	-	-	-	-	-	-	-	-	-	-	-
Remote population density (persons/km ²)	1		1	1	1	1	1	1	1	1	1	1	

^a Continental refers to the archetype defined to model rural emissions (see main text).
^b Assuming a square urban area.
Note: 1) M = million, B = billion, 2) Because of rounding, values might not match perfectly.

Based on the World Bank (Angel et al. 2005) data, I find that the population density d_{urban} (persons km⁻²) of an urban area is correlated to its population N_{urban} , with an approximately linear relationship between the logs of the two variables:

$$d_{urban,i} = (N_{urban,i})^{a_i} \times b_i \quad (4-14)$$

where the parameters a_i and b_i can be specific to each region. The parameters a_i and b_i are very sensitive to the population and area of the cities used in the regression. The results provided by Equation 4-14 are therefore a first approximation and should be taken with care.

4.11.2. Characterization of particulate matter emissions

Total emission by source type and emission height

Table 4-8 shows the total United States annual emissions (t y⁻¹) from different sources for different pollutants (USEPA 2008). H , L , and T represent emissions that are assumed to come from high-stack, low-stack and ground-level sources, respectively. Wildfire and miscellaneous emissions are excluded from the inventory. Table 4-8 also shows the fraction of emissions from high-stack, low-stack, and ground-level sources, based on these data and classifications.

Table 4-8 : Total United States emissions of PM₁₀, PM_{10-2.5}, PM_{2.5}, SO₂, NO_x, and CO, by source (USEPA 2008)^a.

Source Sector	PM ₁₀ (t/y)	PM _{10-2.5} (t/y)	PM _{2.5} (t/y)	SO ₂ (t/y)	NO _x (t/y)	CO (t/y)
Electricity generation (H)	620,000	120,000	500,000	10,000,000	4,700,000	650,000
Fertilizer & Livestock (L)	3,100	1,600	1,500		2,100	
Fires (-)	1,500,000	220,000	1,200,000	100,000	160,000	15,000,000
Fossil fuel combustion (L)	360,000	170,000	190,000	2,000,000	2,400,000	1,500,000
Industrial processes (H)	1,200,000	710,000	490,000	1,200,000	1,200,000	2,400,000
Miscellaneous (-)	5,500,000	5,300,000	250,000	750	2,300	34,000
Non-road equipment (T)	330,000	27,000	300,000	520,000	4,500,000	22,000,000
On-road vehicles (T)	200,000	55,000	150,000	260,000	8,100,000	63,000,000
Residential wood combustion (L)	370,000	29,000	340,000	5,100	37,000	2,700,000
Road dust ^b (T)	10,000,000	9,400,000	850,000	0	0	0
Solvent use (L)	8,300	1,300	7,000	1,000	9,000	3,300
Waste disposal (H)	300,000	22,000	270,000	26,000	120,000	2,000,000
Total^c	14,000,000	10,500,000	3,100,000	14,000,000	21,000,000	95,000,000
Total high-stack (H)	2,100,000	850,000	1,300,000	12,000,000	6,000,000	5,100,000
Total low-stack (L)	740,000	200,000	540,000	2,000,000	2,500,000	4,200,000
Total ground-level (G)	11,000,000	9,500,000	1,300,000	770,000	13,000,000	85,000,000
% from high-stack ($f_{e,high-stack}$)	16%	8%	41%	81%	28%	5%
% from low-stack ($f_{e,low-stack}$)	5%	2%	17%	14%	12%	4%
% from ground-level ($f_{e,ground-level}$)	79%	90%	42%	5%	60%	90%
Fraction of PM₁₀ smaller than 2.5 µm ($f_{<2.5,e}$) and PM₁₀ within the range 2.5 µm and 10 µm ($f_{10-2.5,e}$)						
	$f_{<2.5,e}$	$f_{10-2.5,e}$				
for high-stack	60%	40%				
for low-stack	73%	27%				
for ground-level	12%	88%	$f_{<2.5,e} = 0.73$ for tailpipe-only emissions from road transportation			
for indoor	92%	8%	based on residential wood combustion and solvent emissions			
^a NH ₃ annual emissions in the United States are approximately 4 million tonnes, 95% from low-stack (mainly livestock) and 5% from transportation (i.e., ground-level) (USEPA 2008).						
^b If the inventory database used does not contain road dust emissions, then this value should be removed from the calculations.						
^c Excluding wildfire and miscellaneous emissions.						

To estimate typical stack height values, I use a detailed list of stack heights from ten German states found in Pregger and Friedrich (2009). SO₂, NO_x, and CO emission-weighted average stack heights from industrial point sources are, respectively, 144, 121, and 132 m. Assuming that German industrial point sources have relatively higher stacks, a value of 100 m for worldwide stacks, based on Van Zelm et al. (2008), appears plausible.

Fraction of PM₁₀ smaller than 2.5 µm ($f_{<2.5,e}$)

Different values are suggested in the literature for the fraction of PM₁₀ smaller than 2.5 µm ($f_{<2.5,e}$), from as low as 5% (embarkation of coal - Klimont et al. 2002, IIASA 2004, Passant et al. 2002), 8% for road dust (USEPA 2008), 60% (Klimont et al. 2002, EC 2008), 66% (average in Germany — Pregger 2006), 89% (United States coal power plant average - Frischknecht 2005), 95% (tailpipe — Norbeck et al. 1998), 70% to 95% (coal fired power station with flue gas cleaning) and up to nearly 100% (mobile, internal combustion engine—Klimont et al. 2002, IIASA 2004, Passant et al. 2002).

As shown in Table 4-8, average values in the United States for $f_{<2.5,e}$ are ~0.12 for ground-level emissions [including non-road equipment and road dust – note that if the inventory database used does not contain road dust emissions, then this ratio should be corrected by removing the road dust from the calculations (see Table 4-8 and Table 4-9)], ~0.73 for tailpipe-only emissions from road transportation, ~0.73 for low-stack emissions (based on fossil fuel and residential wood combustion), and ~0.60 for high-stack emissions (based on electricity generation - USEPA 2008). These ratios are variable and can be adapted to specific situations, such as for older power plants.

4.11.3. Intake fractions

Intake fraction regressions from USEtox

Unknown stack height. The intake fraction of primary PM_{2.5} modeled with USEtox (version January 2009 — Rosenbaum et al. 2008) can be approximated using the following regressions (Equations 4-15 to 4-18), where L is the length of the urban area (km) and d_{urban} , d_{rural} and d_{remote} are the respective population densities of the urban, rural, and remote areas (persons km⁻²).

Total intake fraction for a remote emission (R^2 of 1.00 over 100 remote conditions when only d_{remote} varies):

$$iF_{remote} = 2.3 \times 10^{-8} \times d_{remote} + 6.0 \times 10^{-8} \quad (4-15)$$

Total intake fraction for a rural emission (R^2 of 1.00 over 40 rural conditions when only d_{rural} varies):

$$iF_{rural} = 2.3 \times 10^{-8} \times d_{rural} + 6.0 \times 10^{-8} \quad (4-16)$$

Intra-urban intake fraction for an urban emission (R^2 of 0.99 over 200 urban conditions when L and d_{urban} vary):

$$iF_{intra-urban} = 1.7 \times 10^{-10} \times L \times d_{urban} \quad (4-17)$$

Total intake fraction for an urban emission, the sum of Equation 4-17 and Equation 4-16 (R^2 of 0.99 over 8,000 conditions when L , d_{urban} and d_{rural} vary):

$$iF_{urban} = iF_{intra-urban} + iF_{rural} \quad (4-18)$$

If rural conditions are unknown, the intake fraction for a rural emission presented in Table 4-3 can be used as a default iF_{rural} in Equation 4-18. Note that the high R^2 in Equations 4-15 to 4-18 is related to the dominant influence of population density relative to the effects of advection, deposition and degradation. A shorter-lived pollutant would not have as good a linear fit.

Differentiating stack heights. Intake fractions of primary PM_{2.5} evaluated with USEtox (Rosenbaum et al. 2008) are for unknown stack height emissions. The intake fraction for an unknown stack height ($iF_{unknown-stack}$) can be calculated as the emission-weighted average of the intake fractions for high-stack ($iF_{high-stack}$), low-stack ($iF_{low-stack}$) and ground-level ($iF_{ground-level}$):

$$iF_{unknown-stack} = f_{e,high-stack} \times iF_{high-stack} + f_{e,low-stack} \times iF_{low-stack} + f_{e,ground-level} \times iF_{ground-level} \quad (4-19)$$

level

where $f_{e,high-stack}$, $f_{e,low-stack}$, and $f_{e,ground-level}$ are the respective fractions of total emissions from high-stack, low-stack and ground-level emissions (Table 4-8).

To consistently differentiate among stack heights, one can calculate the intake fraction ratios of ground-level to low-stack (X) and low-stack to high-stack (Y) emissions:

$$X = iF_{ground-level}/iF_{low-stack} \quad (4-20)$$

$$Y = iF_{low-stack}/iF_{high-stack} \quad (4-21)$$

RiskPoll (Spadaro and Rabl 2004a) provides these intake fractions for primary PM_{2.5}, with ground-level to low-stack ratios (X) of 2.2 for urban and 1.7 for rural conditions, and ratios of low-stack to high-stack (Y) of 1.4 for urban and 1.3 for rural conditions.

Combining Equations 4-19 to 4-21:

$$iF_{high-stack} = iF_{unknown-stack} / (f_{e,high-stack} + Y \times f_{e,low-stack} + X \times Y \times f_{e,ground-level}) \quad (4-22)$$

$$iF_{low-stack} = Y \times iF_{unknown-stack} / (f_{e,high-stack} + Y \times f_{e,low-stack} + X \times Y \times f_{e,ground-level}) \quad (4-23)$$

$$iF_{ground-level} = X \times Y \times iF_{unknown-stack} / (f_{e,high-stack} + Y \times f_{e,low-stack} + X \times Y \times f_{e,ground-level}) \quad (4-24)$$

Intake fraction regressions from Greco et al. (2007)

The intake fractions for secondary PM from SO₂ and NO_x are evaluated using the regressions of Greco et al. (2007):

$$iF_{(PM(SO_2))urban\ and\ rural} = (P_{<50km} \times 1.31 \times 10^{-13} + P_{50-100km} \times 3.11 \times 10^{-14} + P_{100-200km} \times 6.92 \times 10^{-15} + P_{200-500km} \times 4.04 \times 10^{-15} + P_{>500km} \times 8.35 \times 10^{-16}) \times (13/20) \quad (4-25)$$

$$iF_{(PM(NO_x))urban\ and\ rural} = (P_{<50km} \times 1.56 \times 10^{-14} + P_{50-100km} \times 4.89 \times 10^{-15} + P_{100-200km} \times 6.44 \times 10^{-16} - P_{200-500km} \times 1.69 \times 10^{-16} + P_{>500km} \times 2.75 \times 10^{-16}) \times (13/20) \quad (4-26)$$

where $P_{<50km}$, $P_{50-100km}$, $P_{100-200km}$, $P_{200-500km}$, $P_{>500km}$ are the populations within a radius of 50 km from the location of emission, within a “donut” of 50-100 km, within a “donut” of 100-200 km, within a “donut” of 200-500 km, and further than 500 km, respectively. The ratio at the end adjusts for a breathing rate of 13 m³ person⁻¹ d⁻¹ (USEPA 1997) used in the present chapter, whereas Greco et al. (2007) used originally 20 m³ person⁻¹ d⁻¹. Table 4-9 presents the populations used in the regressions of Greco et al. (2007).

Table 4-9 : Populations (in millions) used in the regressions of Greco et al. (2007).

	urban case	rural for 100 km, then continental case	remote
$P_{<50km}$	2.3	0.40	0.0078
$P_{50-100km}$	2.1	2.0	0.024
$P_{100-200km}$	8.5	8.5	0.094
$P_{200-500km}$	59	59	0.66
$P_{>500km}$	830	830	9.2
Total continent:	900	900	10

Intake fraction regressions from Heath et al. (2006)

Heath et al. (2006) developed two regressions to evaluate the intake fractions for urban and rural emissions of primary PM_{2.5}:

$$iF_{urban} = 5.8 \times (P_{100})^{0.5} \quad (4-27)$$

$$iF_{rural} = 114.6 \times (H_E)^{-1.174} \times (P_{100})^{0.838} \quad (4-28)$$

where P_{100} , in millions of persons, is the population within a radius of 100 km of the emitting facility and H_E is the stack height, in meters.

Intake fractions of PM_{2.5} depending on the emission archetype

Table 4-10 presents the comparison of PM_{2.5} intake fractions of different models. The last column indicates which of the values are reported in Figure 4-2.

Table 4-10 : Comparison of PM_{2.5} intake fractions (ppm) of different models.

Model	Urban	Rural	Remote	Emissions-weighted average	Variation in intake fraction (ratio between max and min intake fraction presented in each line)	Comment	Shown in Figure 4-2
Greco et al. (2007) counties	8.9 (max = 15 in N.Y.)	1.6	(min = 0.088 in Maine – cannot be considered remote)	1.6	170	US, BR adapted to 13 m ³ /(pers·d) (original is 20); urban > 2000 person/km ²	X
Greco et al. (2007) regressions	3.5	2.5 with all 90 persons/km ² ; 2.2 with 50 persons/km ² for 100 km × 100 km	0.027		130	BR adapted to 13 m ³ /(pers·d) (original is 20)	X
Levy et al. (2002)		1.4 ^a (stationary) 5.9 ^a (mobile)				BR adapted to 13 m ³ /(pers·d) (original is 20)	X
Levy et al. (2002) regressions (stationary)	(1.9 – does not capture urban areas)	1.9	(0.61 – cannot be considered remote)		>3	BR adapted to 13 m ³ /(pers·d) (original is 20)	X
Levy et al. (2002) regressions (mobile)	(15 – does not capture urban areas)	14	(2.0 – cannot be considered remote)		>8	BR adapted to 13 m ³ /(pers·d) (original is 20)	X
Evans et al. (2002)	6.1 (mobile)	5.7 (mobile); 1.4 (0.16-4.1) (power plant)			38	based on Wolff (2000), US, original BR is 20 m ³ /(pers·d)	X
USEtox (Rosenbaum et al. 2008)	19	2.2 (2.1 with a rural box of 100 km × 100 km with 50 persons/km ² in the continental box)	0.084	10	230	generic continent	X
RiskPoll (Spadaro and Rabl 2004a, 2004b, Rabl and Spadaro 2005, Hirschberg et al. 2003), transportation	36.9	7.38	0.041		900		
RiskPoll (Spadaro and Rabl 2004a, 2004b, Rabl and Spadaro 2005, Hirschberg et al. 2003), low stack (25 m)	12.5	3.89	0.041		300		
RiskPoll (Spadaro and Rabl 2004a,	9.08	3.16	0.041		220		

Model	Urban	Rural	Remote	Emissions-weighted average	Variation in intake fraction (ratio between max and min intake fraction presented in each line)	Comment	Shown in Figure 4-2
2004b, Rabl and Spadaro 2005, Hirschberg et al. 2003), high stack (100 m)							
RiskPoll (Spadaro and Rabl 2004a, 2004b, Rabl and Spadaro 2005, Hirschberg et al. 2003), very high stack (250 m)	5.02	2.50	0.041		120		
RiskPoll (Spadaro and Rabl 2004a, 2004b, Rabl and Spadaro 2005, Hirschberg et al. 2003), emission-weighted average stack height (using transportation, low and high stacks)	21	5.0	0.041		510		X
Krewitt (2001) (for the reference year 2010)		1.7				Europe	X
Hofstetter (1998)		5.7				Europe	X
Van Zelm et al. (2008)				4.9		Europe, as PM ₁₀ , 90 persons/km ²	X
Marshall et al. (2007) (intra-urban only)	37					Los Angeles	
Marshall et al. (2007) regressions (intra-urban only)	12					BR adapted to 13 m ³ /(pers-d) (original is 12.2) and population to 1.6·10 ⁶ persons	X
Heath (2006)	18	0.78			23	US, original values for California cities and rural areas	
Heath (2006) regressions (adapted with average world conditions)	13	1.3	0.031		420	Regression developed originally for US, sensitive to the stack height, BR adapted to 13 m ³ /(pers-d) (original is 20)	X
NEEDS (MET.NO. 2008, Preiss et al. 2008, EcoSenseWeb 2008)				2.95 (0.34-5.83)	17	Range is high stack in lowest country to low stack in highest country within Europe	X
Overall variation in intake fraction (ratio between max and min intake fraction presented in each column)	15	18	3	30			

^a Although these values are weighted averages of intake fraction estimates using relative emissions in United States areas, these weights are not based on urban or rural differences and therefore cannot be considered to be the "average" intake fraction..

For urban areas, the breathing rate-corrected intake fraction for urban emissions provided by the regressions from Greco et al. (2007) is 3.5 ppm, assuming average global urban parameters (4,000 persons km⁻² over 20 km × 20 km) surrounded by a continental region (90 persons km⁻²). This intake fraction is lower than the 19 ppm found with USEtox (Rosenbaum et al. 2008), assuming the same urban and continental conditions. This lower intake fraction is largely because the Greco et al. (2007) model is based on an arithmetic average dilution rate of 3,000 m² s⁻¹, which is 5 times higher than the urban harmonically averaged dilution rate of 610 m² s⁻¹ used in USEtox (see main text). When correcting for this factor, the model of Greco et al. (2007) obtains a more similar intake fraction of 17 ppm.

Intake fractions for other pollutants

Table 4-11 presents a summary of emission-weighted average intake fractions for primary PM₁₀, primary PM_{10-2.5} and primary PM_{2.5}, secondary PM from SO₂, from NO_x, from NH₃, and CO from different models.

Table 4-11 : Summary of intake fractions (ppm) for primary PM₁₀, primary PM_{10-2.5}, primary PM_{2.5}, secondary PM from SO₂, secondary PM from NO_x, secondary PM from NH₃, and CO, adjusted for a breathing rate of 13 m³/(pers-d).

Model	intake fractions for							Comment	
	PM ₁₀	PM _{10-2.5}	PM _{2.5}	second- dary PM from SO ₂	second- dary PM from NO _x	second- dary PM from NH ₃	CO		
Units	kg PM ₁₀ / kg PM ₁₀	kg PM _{10-2.5} /kg PM _{10-2.5}	kg PM _{2.5} / kg PM _{2.5}	kg PM (SO ₂)/ kg SO ₂	kg PM (NO _x)/ kg NO _x	kg PM (NH ₃)/ kg NH ₃	kg CO/ kg CO		
Greco et al. (2007) counties (mobile)			1.6	0.38	0.055			BR adapted	“continental” (median); U.S. conditions
Greco et al. (2007) regressions (mobile)			2.6	0.77	0.16			BR adapted	emission weighted average
			2.5	0.75	0.16			BR adapted	“continental” (median)
Levy et al. (2002) (stationary)			1.4	0.14	0.023			BR adapted to 13 m ³ /(pers-d) (original is 20); U.S. conditions for the first two lines	
Levy et al. (2002) (mobile)			5.9	0.12	0.020				
Levy et al. (2002) regression (stationary)			1.4	0.14	0.023				
Levy et al. (2002), regression (mobile)			5.9	0.12	0.020				
Evans et al. (2002) (stationary)			1.4	0.10	0.018			based on Wolff (2000); U.S. conditions	
Evans et al. (2002) (mobile) (urban)			6.1	0.078	0.015				
Evans et al. (2002) (mobile) (rural)			5.7	0.091	0.017				
USEtox (Rosenbaum et al. 2008)			10				34		
RiskPoll (Spadaro and Rabl 2004a, 2004b, Rabl and Spadaro 2005, Hirschberg et al. 2003)	12 ^a	8.1 ^b	12	0.69	0.68		22		emission and stack weighted average
Krewitt (2001) (for the reference year 2010)	1.7			0.60	1.4				“continental;” EU conditions
Hofstetter (1998)	5.7			0.50	1.3	0.30	38		“continental;” EU conditions
Van Zelm et al. (2008)	4.9			0.93	1.0	1.5			emission-weighted average, 90 persons/km ² ; EU conditions
Marshall et al. (2005) regressions (intra-urban only)			12						BR adapted to 13 m ³ /(pers-d) (original is 12.2) and population to 1.6·10 ⁶ ; U.S. conditions
NEEDS (MET.NO. 2008, Preiss et al. 2008, EcoSenseWeb 2008)	1.10		2.95	0.85	0.84	0.66			emission-weighted average; EU conditions

^a Intake fractions of PM₁₀ (ppm): Urban: 7.86 (for 100 m), 11.3 (for 25 m), 24.6 (for transportation); Rural: 2.15 (for 100 m), 2.89 (for 25 m), 4.93 (for transportation); Remote: 0.027.

^b Intake fractions of PM_{10-2.5} (ppm): Urban: 6.91 (for 100 m), 10.3 (for 25 m), 15 (for transportation); Rural: 1.38 (for 100 m), 2.09 (for 25 m), 3.00 (for transportation); Remote: 0.017.

Recommended intake fractions

Table 4-12 summarizes the models and assumptions used for the recommended intake fractions presented in Table 4-3. In summary, USEtox (Rosenbaum et al. 2008) is used for primary PM and CO, Greco et al. (2007) for secondary PM from SO₂ and NO_x, Van Zelm et al. (2008) for secondary PM from NH₃ and RiskPoll (Spadaro and Rabl 2004a) to differentiate among high-stack, low-stack, and ground-level emissions of primary PM for urban and rural conditions, respectively.

Table 4-12 : Summary of models or assumptions used for the recommended intake fractions of PM_{10-2.5}, PM_{2.5}, SO₂, NO_x, NH₃, and CO.

Pollutant emitted	Type of source for the PM emission :	Intake fractions for the respective location :						Population-weighted average				
		In-door (household)	In-door (office)	In-door (industrial)	Urban	Rural	Remote					
PM _{10-2.5}	High-stack	Adapted from Hellweg et al. (2009)			= $iF(PM_{2.5}) \times \text{ratios of } iF(PM_{10-2.5})/iF(PM_{2.5}) \text{ from RiskPoll}$			Weighted average among urban, rural and remote emissions				
	Low-stack											
	Ground-level											
	Emission-weighted average								Weighted average among high-, low-stack, and ground-level (based on Table 4-8)			
PM _{2.5}	High-stack				Re-derived from unknown intake fraction, using i) urban ratio from RiskPoll, and ii) weighted average among high-stack, low-stack and ground-level	Re-derived from unknown intake fraction, using i) rural ratio from RiskPoll, and ii) weighted average among high-stack, low-stack and ground-level	USEtox (with parameters adapted), no difference among high-stack, low-stack, and ground-level			Weighted average among urban, rural, and remote emissions		
	Low-stack											
	Ground-level											
	Emission-weighted average										USEtox (with urban parameters adapted)	USEtox (with rural parameters adapted)
SO ₂	High-stack				= ground-level (based on Levy et al. (2002))	= ground-level (based on Levy et al. (2002))	based on rural intake fraction, using same ratio as for PM _{2.5} among rural and unknown			Weighted average among urban, rural and remote emissions		
	Low-stack											
	Ground-level						Greco et al. (2007) for urban (with parameters adapted)				Greco et al. (2007) for rural (with parameters adapted)	= high-stack (no difference among high-stack, low-stack and ground-level)
	Emission-weighted average						Weighted average among high-, low-stack, and ground-level (based on Table 4-8)					
NO _x	High-stack	= ground-level (based on Levy et al. (2002))	= ground-level (based on Levy et al. (2002))	based on rural intake fraction, using same ratio as for PM _{2.5}			Weighted average among urban, rural and remote emissions					
	Low-stack											
	Ground-level			Greco et al. (2007) for urban (with parameters adapted)				Greco et al. (2007) for rural (with parameters adapted)	= high-stack (no difference among high-stack, low-stack and ground-level)			
	Emission-weighted average			Weighted average among high-, low-stack, and ground-level (based on Table 4-8)								

NH ₃	High-stack		= rural intake fraction, assuming no difference in intake fraction between rural and urban emissions	Van Zelm et al. (2008), assuming no difference among high-stack, low-stack and ground-level	based on rural intake fraction, using same ratio as for PM _{2.5}	Weighted average among urban, rural and remote emissions
	Low-stack				= high-stack (no difference among high-stack, low-stack and ground-level)	
	Ground-level					
	Emission-weighted average				Weighted average among high-stack, low-stack, and ground-level (based on Table 4-8)	
CO	High-stack		= PM _{2.5} (based on Marshall et al. (2007) for intra-urban + rural intake fraction	USEtox (with rural parameters adapted), no difference among high-stack, low-stack, and ground-level	USEtox (with remote parameters adapted), no difference among high-stack, low-stack, and ground-level	Weighted average among urban, rural and remote emissions
	Low-stack					
	Ground-level					
	Emission-weighted average					
<i>Legend for the cell colors</i>						
<i>Based on (re-parameterized) model</i>		<i>Equalized or derived from other values, based on strong evidence or models</i>		<i>Weighted average</i>		<i>Equalized or derived from other values, based on weak evidence or models</i>

Table 4-13 provides the equations behind each of the intake fractions provided in Table 4-3.

Table 4-13 : Summary of equations used for the recommended intake fractions of PM_{10-2.5}, PM_{2.5}, SO₂, NO_x, NH₃, and CO.

Pollutant emitted	Type of source for the PM emission	Intake fractions for the respective location						Population-weighted average			
		In-door (household)	In-door (office)	In-door (industrial)	Urban	Rural	Remote				
PM _{10-2.5}	High-stack	= Eq. 4-3			$= iF(PM_{2.5\ urban}) \times (6.91/9.08)$	$= iF(PM_{2.5\ rural}) \times (1.38/3.16)$	$= iF(PM_{2.5\ remote}) \times (0.017/0.041)$	= Eq. 4-6			
	Low-stack				$= iF(PM_{2.5\ urban}) \times (10.3/12.5)$	$= iF(PM_{2.5\ rural}) \times (2.09/3.89)$	$= iF(PM_{2.5\ remote}) \times (0.017/0.041)$				
	Ground-level				$= iF(PM_{2.5\ urban}) \times (15/36.9)$	$= iF(PM_{2.5\ rural}) \times (3.0/7.38)$	$= iF(PM_{2.5\ remote}) \times (0.017/0.041)$				
	Emission-weighted average				= Eq. 4-19						
PM _{2.5}	High-stack							= Eq. 4-22	= Eq. 4-22	= Eq. 4-15	= Eq. 4-6
	Low-stack							= Eq. 4-23	= Eq. 4-23		
	Ground-level							= Eq. 4-24	= Eq. 4-24		
	Emission-weighted average							= Eq. 4-18	= Eq. 4-16	= Eq. 4-19	
SO ₂	High-stack							$= iF_{ground-level}$	$= iF_{ground-level}$	$= iF(SO_{2\ rural}) \times (iF(PM_{2.5\ remote}) / iF(PM_{2.5\ rural}))$	= Eq. 4-6
	Low-stack										
	Ground-level							= Eq. 4-25	= Eq. 4-25	$= iF_{high-stack}$	
	Emission-weighted average				= Eq. 4-19						
NO _x	High-stack				$= iF_{ground-level}$	$= iF_{ground-level}$	$= iF(NO_x\ rural) \times (iF(PM_{2.5\ remote}) / iF(PM_{2.5\ rural}))$	= Eq. 4-6			
	Low-stack										
	Ground-level				= Eq. 4-26	= Eq. 4-26	$= iF_{high-stack}$				
	Emission-weighted average	= Eq. 4-19									
NH ₃	High-stack				$= iF_{rural}$	Van Zelm et al. (2008), assuming no difference between high-stack and low-stack = 1.5 ppm ^a	$= iF(NH_3\ rural) \times (iF(PM_{2.5\ remote}) / iF(PM_{2.5\ rural}))$	= Eq. 4-6			
	Low-stack										
	Ground-level						$= iF_{high-stack}$				
	Emission-weighted average	= Eq. 4-19									
CO	High-stack				= Eq. 4-19	= 22 ppm	= 11 ppm	= Eq. 4-6			
	Low-stack										
	Ground-level										
	Emission-weighted average										

Legend for the cell colors

Based on (re-parameterized) model	Equalized or derived from other values, based on strong evidence or models	Weighted average	Equalized or derived from other values, based on weak evidence or models
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^a As a first approximation, this value can be adapted to a specific rural situation by multiplying it by (x/90), with x being the population density (persons/km²) of the rural area under consideration and 90 (persons/km²) being the population density of the generic rural area.

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5. Bringing the characterization factors into practice: Identifying major contributors and influence of regionalization

As stressed in Chapter 3, performing a complete regionalized life-cycle assessment study based on geographically differentiated information remains almost impossible for current life-cycle assessment software because of the significant amount of data that must be handled. Building on developments in Chapter 3 for fate and exposure of organic pollutants and primary PM, as well as on Chapter 4 for secondary PM factors, in this chapter, I apply the characterization factors to real emission scenarios, identify the major contributors, and revisit the questions of regionalization in life-cycle assessment. But in this chapter, I focus on the broader life-cycle assessment perspective of different processes (i.e., considering the amount of emissions from the life-cycle inventory of the different processes) and not only on the impact assessment side of life-cycle assessment as in Chapters 3 and 4.

5.1. Summary

Regionalization of inventory and impact assessment is recognized as an important step towards improving accuracy, precision and confidence in life-cycle assessment results, as well as discriminatory power. Two approaches can be used to perform regionalization in life-cycle assessment: the geographically differentiated approach and the archetype approach. The geographic approach uses information regarding where the process is located in the world (e.g., downtown Chicago), and considers local conditions to estimate the impacts of direct emissions. For the archetype approach, however, the exact location of the emission is not needed, since this approach uses information on the main characteristics of the emission location to evaluate the subsequent impacts. I have examined selected processes and pollutants that contribute significantly to human health impacts, and have found that geographically distributed damage results can vary by two orders of magnitude, depending on the location where the direct emission occurs. For indirect emissions, which occur in background processes, regional damage results vary by a factor of two. Indeed, background processes are often spread throughout a region and thus have a low sensitivity to regionalization. Therefore, processes dominated by impacts from background emissions will have a low sensitivity to regionalization.

In addition, I find that both primary and secondary PM are major contributors to human health damage in life-cycle assessment. Regionalization scheme implemented in life-cycle assessment should therefore address these pollutants in priority.

Since there is currently no tool for gathering large sets of detailed geographic data on inventory and impact assessment in an efficient way, I recommend using the archetype approach as a realistic implementation of regionalization in life-cycle assessment. This will significantly raise confidence in the results while making the regionalization process practical. I also suggest expanding this work to other regions of the world, as well as to other impact categories such as ecotoxicity, land use, and water use. Performing regionalization using the archetype approach can significantly improve the results of life-cycle assessments.

5.2. Introduction

Regionalization in life-cycle assessment

In this chapter I expand the analysis done in Chapters 3 and 4 for impact assessment methods and address the regionalization of life-cycle assessment (see section 3.2 for an introduction on regionalization of life-cycle assessment).

Only a few life-cycle impact assessment methods provide geographically based characterization factors (e.g., Toffoletto et al. 2007, Bare et al. 2003, Potting and Hauschild 2006). However, the resolution used when generating these existing characterization factors is not higher than approximately 100,000 km². In Chapter 3 I discuss the issue of regionalization and conclude that the archetype approach has significant advantages over the geographic approach in performing regionalization for fate and exposure of air pollutants.

Objectives

In this chapter I explore whether regionalization based on the archetype approach significantly improves the evaluative power of life-cycle assessment while minimizing the data requirements needed by the geographically differentiated approach. This chapter has the following objectives: (i) to identify the major contributors to damage to human health in order to identify for which pollutants regionalization schemes should be developed in priority, (ii) to evaluate the influence of regionalization on human health damage using an illustrative set of processes, and (iii) to discuss the advantages and disadvantage of the archetype and geographic approaches for performing regionalization in life-cycle assessment.

5.3. Method

Modeling framework

Figure 2-1 illustrates the framework used to calculate the overall damage to human health of different processes. The unit of the damage used in the present chapter is disability-adjusted life years (DALY — Murray and Lopez 1996). DALY was discussed in detail in section 1.1.1.

The overall damage score associated with a process is therefore a function of both the quantity of the emission and the type of emission – the latter will determine its characterization factor. Therefore, when evaluating the influence of regionalization on different processes, one first needs to evaluate which pollutants are contributing significantly to the overall damage score.

Identifying impact categories dominating human health damage

The contribution from the different impact categories for the total human health damage of all ecoinvent 2.0 (Frischknecht 2005) processes is presented first. Then the contribution from the annual United States air emissions is evaluated. These analyses allow one to identify the dominant impact categories and thus identify the priorities for determining archetypes. The impact assessment tool used is IMPACT 2002+ (Jolliet et al. 2003), for which the category respiratory inorganics is updated using the values from Chapter 4.

Selected process and assessment procedure

A number of processes are selected to evaluate the importance of regionalization. They are selected from those that have a significant share of impacts on human health. The literature suggests that housing, transport, food and other consumption goods are all responsible for a significant portion of the human health damage caused by emissions of pollutants in the environment (Kaenzig and Jolliet 2006, Huppel et al. 2006). Based on this rationale, the following processes are selected as being representative of high production volume processes for each of the four classes introduced above: clinker production for Portland cement (Cement),

electricity from a coal power plant (Electricity), crude oil extraction (Oil), truck driving (Truck), gasoline car driving (Car), trans-oceanic container shipping (Boat), nitrogen-based fertilizer production (Fertilizer), glyphosate application (Pesticide), and polyethylene terephthalate (PET) production.

Characteristics of the selected unit processes

The unit process selected in ecoinvent 2.0 (Frischknecht 2005) and the reference flow (ISO 2006a, 2006b, section 1.1.1) of these processes are given in Table 5-1.

Table 5-1: Characteristics of the selected unit processes.

Process	Unit process selected in ecoinvent 2.01	Reference flow	Comments
Clinker	Clinker, at plant (Swiss conditions)	1 t	This process represents the production of clinker
Electricity	Hard coal, burned in power plant (Southeastern Electric Reliability Council conditions)	1 kWh	This process represents the production of electricity in a coal-fired power plant
Oil	Crude oil, at production offshore (Norway conditions)	1 t	This process represents crude oil extraction
Truck	Transport, lorry >32t, EURO3 (average European conditions)	1 t-km	This process represents goods transportation by a truck
Car	Transport, passenger car, petrol, fleet average 2010 (average European conditions)	1pers-km	This process represents average 2010 fleet gasoline passenger car
Boat	Transport, transoceanic freight ship (average oceanic fleet)	1 t-km	This process represents an average transoceanic freight ship
Fertilizer	Ammonium nitrate, as N, at regional storehouse (average European conditions)	1 t	This process represents fertilizer production; Ammonia (steam reforming, liquid) and Nitric acid (50% in H ₂ O) are performed on-site; Composition is NH ₄ NO ₃
Pesticide	Application of plant protection products, by field sprayer (Swiss conditions) and Glyphosate, at regional storehouse (Swiss conditions)	1 kg-ha	This process represents pesticide production and use; Direct impacts from glyphosate based on Humbert et al. (2007); Composition is Glyphosate (N-(phosphonomethyl) glycine) (the active ingredient of Roundup)
PET	Polyethylene terephthalate, granulate, bottle grade, at plant (average European conditions)	1 t	This process represents PET production

Life-cycle inventory

The inventory database ecoinvent (Frischknecht 2005) is used to calculate the life-cycle inventory. This database, combined with software such as SimaPro (PRé 2006), allows one to perform a regionalization analysis because it allows for the separation of foreground and background emissions as well as for the distinction between high and low population density areas for the zone of emission. The foreground emissions, also referred to as direct emissions, are the emissions occurring directly from the process of interest. Background emissions, also referred to as indirect emissions, are the emissions occurring from all the processes upstream and downstream of the process of interest. The regionalization is first assessed only with respect to direct emissions, using characterization factors specific to different release locations. In this first step, the indirect emissions occurring in unknown areas are assessed with generic characterization factors. In a second step, regionalization is also assessed for indirect emissions, using the archetypes given by default in ecoinvent. However, because the archetypes given by default in ecoinvent are only roughly defined (distinction among air emissions in high, low and “unknown” population densities) and are often set to unknown, the archetypes given by default in ecoinvent only enable one to perform a partial (or rough) regionalization of indirect emissions. Note that version 3 of ecoinvent will improve the regionalization of the database, but details are not yet known (Hischier 2009).

Table 5-2 presents the life-cycle inventory of the selected unit processes.
Table 5-2: Life-cycle inventory of air pollutants emissions (selected substances – full list can be found directly in ecoinvent 2.0 – Frischknecht 2005) from the selected unit processes (emissions are expressed in kg per unit).

Process	Units		CO ₂	PM _{2.5}	NO _x	SO ₂	NH ₃	Dioxin	BaP***
Clinker	per kg	direct	8.4E-01	2.4E-05	1.1E-03	3.6E-04	2.3E-05	9.6E-13	0.0E+00
		indirect	4.3E-02	2.5E-05	2.0E-04	9.1E-05	6.6E-06	1.1E-14	4.0E-10
Electricity	per MJ	direct	9.7E-02	3.4E-06	1.5E-04	5.0E-04	0.0E+00	1.6E-14	7.7E-13
		indirect	4.5E-03	2.1E-06	6.2E-05	1.0E-05	5.1E-06	2.7E-15	4.2E-11
Oil	per kg	direct	4.6E-02	1.8E-05	2.6E-04	3.5E-05	0.0E+00	0.0E+00	1.0E-11
		indirect	4.4E-03	1.6E-06	1.3E-05	2.1E-05	3.4E-07	6.1E-15	4.0E-11
Truck	per t-km*	direct	7.9E-02	1.4E-05	6.8E-04	2.7E-06	4.3E-07	1.1E-15	7.4E-13
		indirect	3.3E-02	1.5E-05	1.5E-04	1.2E-04	1.2E-06	3.7E-14	4.1E-10
Car	per pers-km**	direct	1.2E-01	5.7E-06	1.6E-04	3.8E-06	1.6E-05	0.0E+00	0.0E+00
		indirect	5.5E-02	2.0E-05	1.7E-04	2.9E-04	1.8E-06	6.3E-14	9.1E-10
Boat	per t-km*	direct	9.4E-03	4.0E-06	1.4E-04	1.3E-04	1.0E-06	3.4E-15	2.8E-11
		indirect	2.6E-03	8.5E-07	7.1E-06	1.5E-05	4.8E-08	1.2E-15	3.3E-11
Fertilizer	per kg	direct	8.9E-01	7.5E-04	1.1E-02	6.1E-06	6.2E-03	2.6E-15	2.8E-10
		indirect	2.0E+00	7.2E-04	3.9E-03	5.8E-03	6.3E-05	1.3E-12	1.6E-08
Pesticide	per kg	direct	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
		indirect	1.5E+01	4.8E-03	2.7E-02	6.5E-02	1.8E-04	2.9E-12	1.6E-07
PET	per kg	direct	4.2E-01	9.8E-05	4.2E-04	1.1E-03	8.1E-08	1.8E-14	9.9E-11
		indirect	2.1E+00	4.5E-04	4.4E-03	5.7E-03	4.1E-05	7.8E-13	1.9E-08

* t-km, i.e., one tonne transported over one km; ** pers-km, i.e., one person transported over one km; ***BaP means Benzo(a)pyrene.

Calculating generic versus regional human health damage

Multimedia fate and multipathway exposure models are recognized as suitable modeling approaches to assess chemical fate and exposure in life-cycle impact assessment (Hertwich 2002). To perform the present analysis, I need to use a model that can provide both generic and regional characterization factors. Several models are available to calculate geographic fate and exposure for hundreds of pollutants: BETR North America (MacLeod et al. 2001), BETR World (Toose et al. 2004), IMPACT 2002 Western Europe (Pennington et al. 2005), IMPACT 2002 Continental (Rochat et al. 2006), GLOBOX (Sleeswijk 2006), IMPACT North America (Chapter 2). IMPACT North America (see Figure 2-3 in Chapter 2) is chosen here because it allows one to calculate both intake fractions and characterization factors at a high geographic resolution and includes urban environments. In addition to the air cells presented in Figure 2-3, 292 urban areas are nested in the respective air cells. These urban boxes allow one to more accurately capture the exposure influence of urban emissions. Indeed, Rosenbaum et al. (2008) and Chapters 2, 3 and 4 have shown that considering urban compartments in impact modeling is important for pollutants of low- to medium-persistence when inhalation is the dominant intake pathway. Note that the model IMPACT North America has some limitations in evaluating fate of and exposure for secondary particulate matter owing to the chemistry involved in its formation. Therefore characterization factors for secondary particulate matter are based on the review and recommendations of existing models as presented in Chapter 4. The residues and damage associated with the use of pesticides are based on Humbert et al. (2007). Finally, the generic characterization factor is defined and calculated as the emissions-weighted average characterization factor for the emissions in the different regions of North America. Since the spatial distribution of most emissions is unknown, emissions are considered, as a first approximation, to be correlated to population (see Chapter 3).

Generic versus archetype versus geographic characterization factors

Table 5-3 presents the generic versus archetype versus geographic characterization factors used in this chapter.

Table 5-3: Generic versus archetype versus geographic characterization factors used in this chapter.

	Type of environment	Type of modeling	PM _{2.5}	NO _x	SO ₂	NH ₃	Dioxin	BaP*
Characterization factors (DALY/kg)	Generic	generic	1.8E-3	1.3E-5	1.1E-4	1.2E-4	1E+3	7E-4
	Urban	archetype	3.4E-3	1.4E-5	1.2E-4	1.2E-4	1E+3	8E-4
		geographic	3.0E-3	1.4E-5	1.2E-4	1.2E-4	3E+3	8E-4
	Rural	archetype	3.9E-4	1.3E-5	1.0E-4	1.2E-4	1E+3	8E-4
		geographic	3.4E-4	1.3E-5	1.0E-4	1.2E-4	2E+3	4E-4
	Remote	archetype	1.4E-5	7.9E-7	6.2E-6	7.6E-6	1E-1	8E-8
geographic		2.0E-6	7.9E-7	6.2E-6	7.6E-6	9E+1	6E-10	
Sources	Generic	generic	Chapter 4	Chapter 4	Chapter 4	Chapter 4	Chapter 2 ^b	Chapter 2 ^b
	Urban	archetype	Chapter 4	Chapter 4	Chapter 4	Chapter 4	Chapter 2 ^b	Chapter 2 ^b
		geographic	Chapter 2 ^b	Chapter 4	Chapter 4	Chapter 4	Chapter 2 ^b	Chapter 2 ^b
	Rural	archetype	Chapter 4	Chapter 4	Chapter 4	Chapter 4	Chapter 2 ^b	Chapter 2 ^b
		geographic	Chapter 2 ^b	Chapter 4	Chapter 4	Chapter 4	Chapter 2 ^b	Chapter 2 ^b
	Remote	archetype	Chapter 4	Chapter 4	Chapter 4	Chapter 4	Chapter 2 ^b	Chapter 2 ^b
geographic		Chapter 2 ^b	Chapter 4	Chapter 4	Chapter 4	Chapter 2 ^b	Chapter 2 ^b	

^a BaP means Benzo(a)pyrene; ^b Values are generated with the IMPACT North America model that contains effect factors included in the model available at <http://www.impactmodeling.org>; the effect factors used for PM_{2.5} in the IMPACT North America model is the same as in Chapter 4 (i.e., 140 DALY/kg_{inhaled}), the effect factors for dioxin and benzo(a)pyrene are respectively 100,000 and 2 DALY/kg_{ingested}.

5.4. Results

Human health damage for all ecoinvent processes

The contribution from the different impact categories for the total human health damage of 3,841 processes defined by ecoinvent 2.01 (Frischknecht 2005) is presented in Table 5-5. Such information allows one to identify the dominant impact categories within different processes and therefore allows one to set priorities when determining archetypes. The impact assessment is based on IMPACT 2002+ (Jolliet et al. 2003), given by default in ecoinvent 2.01 (the Excel file containing the human health damage for all 3,841 processes can be downloaded at <http://www.ecoinvent.ch>), for which the category “respiratory inorganics” is updated using the values from Chapter 4.

The impact categories human toxicity, ionizing radiations, ozone layer depletion, and photochemical oxidation are directly taken from the Excel file that can be downloaded at <http://www.ecoinvent.ch>, using the value of 0.0071 DALY/point to transform the units from points (defined in IMPACT 2002+ — Jolliet et al 2003) per functional unit of each process to DALY per functional unit of each process.

For the category “respiratory inorganics,” the substances from ecoinvent 2.01 presented in Table 5-4 have their characterization factors updated.

The Excel table in which the computation was performed can be downloaded from <http://www.impactmodeling.org>. However, this Excel table is simply for information as all calculation can be performed again using the impact assessment results Excel file that can be downloaded at <http://www.ecoinvent.ch> and the values of 0.0071 DALY/point as well as values found in Table 5-4.

Table 5-4: Substances for which the characterization factor (CF, in DALY/kg) is updated. These substances correspond to the category “respiratory inorganics.”

Substance	particulates, <2.5 um	sulphur dioxide	nitrogen oxides	ammonia	carbon monoxide
Original CF	7.0E-04	5.5E-05	8.9E-05	7.3E-07	8.5E-05
CF used to generate the values presented below for the category “respiratory inorganics”	1.8E-03	1.1E-04	1.3E-05	1.2E-04	6.4E-07

Table 5-5 presents the contribution of background emissions as well as of foreground emissions to the total human health damage.

Table 5-5: Contribution of the different impact categories for the 3,841 processes defined by ecoinvent 2.01 (median of the processes represented for each impact category).

Process class	Number of processes represented	Contribution to total human health damage (%)				
		Human toxicity	Ionizing radiation	Ozone layer depletion	Photochemical oxidation	Respiratory inorganics
agriculture	211	15	0.16	0.0048	0.061	84
chemicals	506	6.0	0.41	0.012	0.070	93
energy	1,501	6.2	0.17	0.0042	0.040	93
materials	767	7.4	0.24	0.0039	0.053	92
processing	281	8.8	0.40	0.0039	0.028	91
transport	206	5.4	0.20	0.013	0.12	94
waste management	369	50	0.11	0.0027	0.040	49

Table 5-5 shows that the category “respiratory inorganics” dominates the total human health damage. This category contains the impacts from CO, primary PM smaller than 2.5 μm (PM_{2.5}), and secondary PM attributable to SO₂, NO_x, and NH₃. These pollutants should therefore get extra attention when evaluating the influence of regionalization on processes. Note that secondary PM attributable to volatile and semi-volatile organic compounds emissions is not addressed (see Chapter 4).

Human health damage from annual United States airborne emissions

The human health damage from annual United States airborne emissions are based on the impact categories respiratory inorganics, human toxicity, ionizing radiation, ozone layer depletion and photochemical oxidation.

Impacts from respiratory inorganics from United States air emissions of primary PM, SO₂, NO_x, NH₃ and CO are based on Chapter 4 for both the inventory results (Table 4-8) and the characterization factors (Table 4-7). Table 5-6 lists the inventory and the characterization factors chosen for the category respiratory inorganics.

Table 5-6: List of characterization factors chosen for the category respiratory inorganics.

Pollutant	Archetype of emission (Table 4-8)	United States annual emissions (t/y) (see Chapter 4)	Characterization factor (DALY/kg) (see Chapter 4)	Source/comment
PM _{10-2.5}	Total high-stack	850,000	n/a	
	Total low-stack	200,000	n/a	
	Total transport	9,500,000	n/a	
	Total fires and miscellaneous	5,500,000	n/a	
PM _{2.5}	Total high-stack	1,300,000	7.3E-04	
	Total low-stack	540,000	1.0E-03	
	Total transport (on and non-road)	450,000	3.1E-03	
	Total transport (road dust)	850,000	2.2E-03	Effect factor assumed to be better represented by undefined than tailpipe emissions; therefore the characterization factor chosen here is the characterization factor for ground-level emission that has been corrected by (140/200); further research is needed to better evaluate road dust effects
	Total fires and miscellaneous	1,500,000	1.2E-05	Low-stack remote
SO ₂	Total high-stack	12,000,000	1.1E-04	
	Total low-stack	2,000,000	1.1E-04	
	Total transport	770,000	1.1E-04	
	Total fires and miscellaneous	100,000	6.2E-06	Low-stack remote
NO _x	Total high-stack	6,000,000	1.3E-05	
	Total low-stack	2,500,000	1.3E-05	
	Total transport	13,000,000	1.3E-05	
	Total fires and miscellaneous	160,000	7.9E-07	Low-stack remote
CO	Total high-stack	5,100,000	4.9E-07	
	Total low-stack	4,200,000	5.2E-07	
	Total transport	85,000,000	6.6E-07	
	Total fires and miscellaneous	15,000,000	2.1E-07	Low-stack remote
NH ₃	Transport	200,000	1.2E-04	
	Low-stack rural	3,800,000	1.2E-04	

Table 5-7 presents the main contributors to human health damage from United States air emissions, using the United States Environmental Protection Agency's Toxics Release Inventory (2008) annual air emissions and IMPACT 2002+ (Jolliet et al. 2003, Humbert et al. 2009) for human toxicity, ionizing radiation, ozone layer depletion and photochemical oxidation, as well as a summary of the results of Table 5-6 for respiratory inorganics.

Table 5-7 shows that annual United States airborne emissions cause approximately 7 million DALYs dominated by primary PM_{2.5} (66%), secondary particulate matter from SO₂ (22%), secondary particulate matter from NH₃ (7%), secondary particulate matter from NO_x (4%), and CO (1%). See section **Error! Reference source not found.** for details on calculating DALY from primary and secondary PM exposure. These five pollutants contribute 99% of the human health damage from annual United States outdoor airborne emissions with primary PM_{2.5} as the dominant contributor. The other 266 pollutants evaluated contribute 1% of the total human health damage from annual United States outdoor airborne emissions, lead by dioxin (see section 5.8).

Note that secondary PM from organics has not been included in the modeling. Current results should therefore be taken with care and identification of the contribution from secondary PM from organics should be done in order to improve the robustness of these results.

At the level of United States (~300,000,000 persons), an annual damage of 7 million DALYs represents approximately 0.024 DALYs per person per year or 9 days per person per year of life lost because of annual United States outdoor air emissions, representing, when assuming an average life expectancy of 77 years (Pope et al. 2009), 1.8 DALY per person per

lifetime. In other words, this result implies a 2% life shortening from air pollution, of which two thirds come from primary PM.

Of these 1.8 DALY per person per lifetime, 66% is coming from primary PM_{2.5} and 22% from secondary PM from SO₂. These two pollutants are assumed to represent fine PM (see Chapter 4). This indicates that approximately 88% of those 1.8 DALY, or 1.6 DALY per person per lifetime, can be attributed to fine PM exposure.

As a matter of comparison, Pope et al. (2009) evaluated a loss of 0.7 to 1.6 years of life expectancy that can be attributed to longterm exposure to PM_{2.5} per incremental concentration of 10 µg per m³. Using the United States average PM_{2.5} concentration in the air in 1999-2000, which is 14 µg per m³ (Pope et al. 2009), the values of Pope et al. (2009) suggest a reduction of life expectancy of approximately $(14/10) \times (0.7 \text{ to } 1.6) = 0.98 \text{ to } 2.2$ years. It is interesting to note that the value I found (1.6 DALY per person per lifetime) falls within the range evaluated by Pope et al. (2009) (which is 0.98 to 2.2 DALY per person per lifetime).

Table 5-7: Main contributors to human health damage from United States emissions.

Pollutant	Annual United States emissions (kg/y) (USEPA 2008)	Characterization factor (DALY/kg emitted) (see Chapter 4)	Impact category considered	Total damage score to human health (DALY/y)	Contribution to total human health damage	Cumulative total human health damage	
Primary PM _{2.5}	4,500,000,000	see Table 5-6	respiratory inorganics	4,700,000	66%	66%	
Secondary particulate matter from SO ₂	15,000,000,000	see Table 5-6		1,600,000	22%	88%	
Secondary particulate matter from NH ₃	4,000,000,000	see Table 5-6		480,000	6.7%	94%	
Secondary particulate matter from NO _x	21,000,000,000	see Table 5-6		270,000	3.8%	98%	
Carbon monoxide (CO)	110,000,000,000	see Table 5-6		64,000	0.89%	99.16%	
Dioxin and dioxin-like compounds	1.4	2.9E+04	human toxicity (IMPACT 2002+ — Jolliet et al. 2003)	41,000	0.57%	99.74%	
Decabromodiphenyl oxide	20,000	6.8E-01		14,000	0.19%	99.93%	
Polycyclic aromatic compounds	200,000	9.9E-03		2,000	0.028%	99.95%	
Arsenic compounds	49,000	3.9E-02		1,900	0.026%	99.98%	
Zinc compounds	2,800,000	2.6E-04		720	0.010%	99.99%	
Selenium compounds	260,000	5.1E-04		130	0.002%	99.99%	
Chromium compounds (except chromite ore mined in the transvaal region)	210,000	3.9E-04		82	0.001%	99.99%	
Zinc (fume or dust)	240,000	2.6E-04		63	0.001%	99.99%	
Mercury compounds	55,000	1.1E-03		59	0.001%	100.00%	
Benzene	2,500,000	2.1E-05		52	0.001%	100.00%	
Formaldehyde	4,200,000	1.2E-05		49	0.001%	100.00%	
All other pollutants (266 identified)	220,000,000			220	0.003%		
All pollutants	150,000,000,000				7,200,000		100%

Human health damage evaluated with generic, geographically-based and archetype-based regional damage factors

For each of the selected processes and locations, human health damage is calculated based on cancer, non-cancer and respiratory effects caused by emitted pollutants (Figure 5-1). Damage from foreground (direct, in gray) and background (indirect, in black) emissions are shown separately.

The three emission locations chosen to illustrate and analyze the influence of regionalization are Chicago for urban emissions, Montana for rural emissions and Alaska for remote emissions. These three locations are chosen because they are good examples of urban areas, rural areas and remote areas. Other cities or regions could have been chosen for the present analysis.

The damage scores from both the generic case and the indirect emissions for the geographically-based case (which occur in an unknown location) are computed using the population emission-weighted characterization factors for North America (Chapter 3). With the archetype characterization factor, both direct and indirect emissions are evaluated with archetype-based characterization factors.

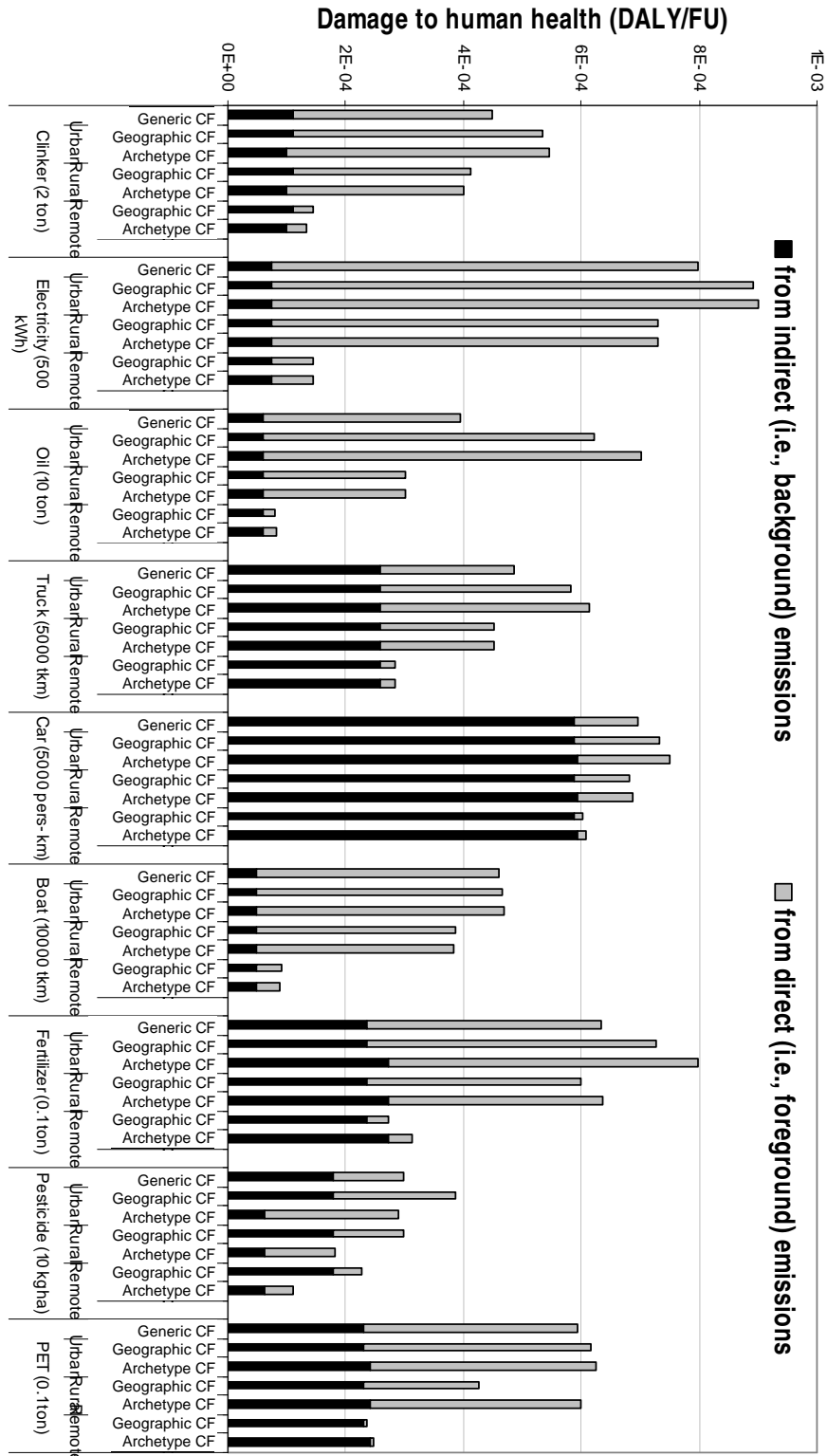


Figure 5-1: Human health damage for selected processes using generic versus geographic-based versus archetype-based regional characterization factors (CF), and distinguishing between damages related to foreground (direct) and background (indirect) emissions. Chicago, Montana and Alaska are taken as an example for urban, rural and remote area respectively.

Table 5-8 lists the pollutants contributing, in total, to more than 99% of the human health damage score caused by direct and indirect emissions for the selected processes, evaluated using generic characterization factors.

Table 5-8: Pollutants^a contributing, in total, to more than 99% of the human health damage score^b caused by direct and indirect emissions for each process (ecoinvent 2.01 – Frischknecht 2005).

		Process considered								
		Clinker	Electricity	Oil	Truck	Car	Boat	Fertilizer	Pesticide	PET
Pollutants contributing to more than 99% of the human health damage score for	direct emissions	PM(SO ₂), PM(NO _x), PM _{2.5} , PM(NH ₃)	PM(SO ₂), PM(NO _x), PM _{2.5}	PM(NO _x), PM _{2.5} , PM(SO ₂)	PM(NO _x), PM _{2.5} , Zinc in soil, PM(SO ₂)	PM(NO _x), PM(NH ₃), PM _{2.5} , PM(SO ₂), Zinc in soil, Benzene	PM(SO ₂), PM(NO _x), PM _{2.5} , PM(NH ₃)	Zinc in soil, PM(NH ₃), PM(NO _x), PM _{2.5}	PM(NO _x), PM _{2.5} , PM(SO ₂)	PAH, PM(SO ₂), PM _{2.5}
	indirect emissions	PM(SO ₂), PM _{2.5} , PM(NO _x), PM(NH ₃)	Arsenic in water, PM(NO _x), PM(NH ₃), PM(SO ₂), PM _{2.5} , PAH	PM(SO ₂), PM _{2.5} , PM(NO _x), PAH, PM(NH ₃), Arsenic in water	PM(SO ₂), PM _{2.5} , PM(NO _x), PAH, Arsenic in water	PM(SO ₂), PAH, PM _{2.5} , PM(NO _x)	PM(SO ₂), PM _{2.5} , PM(NO _x)	PM(SO ₂), PM _{2.5} , PM(NO _x), PAH, Arsenic in air, PM(NH ₃)	PM(SO ₂), PM _{2.5} , PM(NO _x)	PM(SO ₂), PAH, PM _{2.5} , PM(NO _x), Arsenic in water

^a "PM(XX)" means "secondary particulate matter formed from the pollutant XX." PAH refers to polycyclic aromatic hydrocarbons.
^b This damage includes toxicity (i.e., cancer and non-cancer) and respiratory inorganic effects.

Note that the significant contribution of background processes to overall impact score of driving a car can be surprising. This requires further analysis to understand the quality of the life-cycle inventory used for oil extraction and refining. Furthermore, note that this is a car using gasoline: a car using diesel would probably have higher contribution to direct impacts because of its higher amount of PM and NOx tailpipe emissions relative to gasoline cars.

5.5. Discussion

Dominant contributors to human health damage from outdoor emissions

Human health damage from airborne emissions are, in most cases, dominated by primary and secondary PM as well as by CO and dioxin. This can be seen clearly from the results presented in Table 5-5, Table 5-7 and Table 5-8. Therefore, any regionalization system used in life-cycle assessment should address primary and secondary particulate matter as well as carbon monoxide and dioxin. Furthermore, I foresee that for most processes, the sensitivity of the total human damage score will be strongly correlated with how these pollutants are affected by regionalization.

Generic versus regional characterization factors

Figure 5-1 shows that, depending on where a process is located, overall human health damage (i.e., including indirect emissions) can be influenced by less than a factor of two (e.g., for cars, truck, PET production or fertilizers production) to a factor of five (e.g., for oil extraction, electricity production or emissions from boats). However, human health damage from direct emissions can vary by up to a factor of 100 (e.g., for PET production, oil extraction or cars). The use of generic factors can underestimate human health damage for processes situated in urban areas and can overestimate human health damage for processes situated in rural or remote areas. Therefore, when life-cycle processes are mostly in areas with conditions that are significantly different from generic ones (e.g., oil platforms or boats are mostly in areas with low or no population density) or for case studies involving emission scenarios occurring in conditions

significantly different from the generic ones (e.g., a case study comparing truck and train shipping across the Rockies) the use of generic data can underestimate or overestimate the human health damage score by a factor of two or five, respectively. This variation of an order of magnitude observed here for the total impact score underscores the need for regionalization following the recommendation of Margni et al. (2008) stating that differentiation should be considered if sensitivity studies reveal high variations in characterization factors for a given category, that is at least a factor of two to ten, depending on the uncertainty in the category. However, when indirect emissions have a significant contribution to the overall impact score, the need for regionalization in life-cycle impact assessment is reduced.

Table 5-7 lists the pollutants contributing, in total, to more than 99% of the human health damage score caused by direct and indirect emissions for the selected processes, evaluated using generic characterization factors. The damage from those that are inhalation-dominated (e.g., PM_{2.5}) is mainly influenced by population density, whereas the damage of others that are ingestion-dominated (e.g., PAHs) is mainly influenced by the agricultural production intensity (see Chapter 3). Processes having human health damage dominated by primary PM, zinc, benzene, PAHs and arsenic are the most sensitive to the emission location. For these pollutants, regionalization can significantly improve the accuracy in the life-cycle assessment results evaluating human health damage. For processes having human health damage dominated by secondary PM attributable to NO_x, SO₂ and NH₃, the overall human health damage is not very sensitive to whether the emissions occur in urban areas, rural areas, or a generic location. As explained in Chapter 4, secondary PM is not significantly influenced by local population density surrounding the emission location; it is only influenced by regional or continental population densities because of the time required by the transformation processes to form secondary PM from precursors (Levy et al. 2002). It appears that only remote areas (e.g., boat on the ocean, oil platform in the arctic, truck crossing the desert) have impacts where regionalization significantly changes the overall human health damage dominated by secondary PM.

Geographically-based versus archetype-based regional characterization factors

Figure 5-1 shows that the archetype approach gives results consistent with the geographic approach. Because background processes are often scattered around the country or the world, regionalization influences the damage from indirect emissions (i.e., from the background processes) less than the damage from direct emissions (i.e., from foreground processes). Therefore, though the damage from direct emissions can vary up to a factor of 100, the total damage from both direct and indirect emissions varies in a smaller range, within a factor of ten, depending on the share of indirect emissions in the total damage. For several processes, variations of a factor of two are observed for the total damage. Therefore, because the background damage does not vary a lot, and because the background emissions contributes to a significant share of the total human health damage (Table 5-5), even if the foreground damage varies a lot, the total damage will not vary as much as the direct damage.

Outlook

In this chapter I show that regionalization is important for life-cycle impact assessment, and that the archetype approach is a practical way to perform regionalization as it is as accurate as the geographic approach, but more practical to implement. Therefore, as long as no detailed geographic information systems allows the generation of geographically differentiated inventories and the matching of those inventories with impact assessments, I recommend the use of the archetype approach to make regionalization practical for life-cycle assessment. Because most processes have their human health damage score dominated by impacts from primary and

secondary particulate matter, carbon monoxide and dioxin, any regionalization scheme should address these pollutants with care.

Future research

This chapter explores the question of regionalization for non-global human health damage caused by pollutants emitted in North America. This type of work should be expanded to other regions of the world as well as to other types of impact categories such as damage related to tropospheric ozone formation, ecotoxicity, acidification, eutrophication, land use or water use. Future research should be conducted to better assess the optimum archetypes for the different impact categories, media of emissions, world regions, and types of pollutants.

In conclusion, making regionalization a common practice in life-cycle assessment of human health impacts is important, needed and demanded. Indeed, the lack of regionalization in life-cycle assessment has been long discussed and has been considered a limitation of human health damage life-cycle assessment-based results in decision making (Potting and Hauschild 2006, Sedlbauer et al. 2007, Reap et al. 2008, Margni et al. 2008). However, up until now, life-cycle assessment studies that have integrated regionalization have been rare, mainly because of the data intensiveness and large number of computations needed to perform geographically-differentiated regionalization. The archetypes-based regionalization approach would significantly increase confidence in decisions based on life-cycle assessment by improving accuracy, precision and confidence in life-cycle assessment results as well as its evaluative power, and therefore encourage the use of life-cycle assessment in decision making.

5.6. Acknowledgments

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5.8. Supporting information for this chapter

Table 5-9 presents the list of pollutants evaluated to generate Table 5-7.

Table 5-9: List of pollutants evaluated to generate Table 5-7.

Pollutant/Chemical	Emissions (total air release (On-site Fugitive Air + On-site Point Source Air)) (in kg/y)	Source	Characterization factor (CF) for human health (DALY/kg)	Source	Damage score (DALY/y) (i.e., emissions × CF)	Contribution to total damage	Cumulative damage
PM2.5	4.6.E+09	see Chapter 4	see Table 5-6	see Table 5-6	4.7.E+06	66.000%	66.00%
SO2	1.5.E+10				1.6.E+06	22.000%	88.00%
NH3	4.0.E+09				4.8.E+05	6.700%	94.00%
NOx	2.1.E+10				2.7.E+05	3.800%	98.00%
CO	1.1.E+11				6.4.E+04	0.890%	99.00%
DIOXIN AND DIOXIN-LIKE COMPOUNDS	1.4.E+00	TRI (USEPA 2008)	2.9.E+04	Jolliet et al. 2003	4.1.E+04	0.570%	99.70%
DECABROMODIPHENYL OXIDE	2.0.E+04	TRI (USEPA 2008)	6.8.E-01	Jolliet et al. 2003	1.4.E+04	0.190%	99.90%
POLYCYCLIC AROMATIC COMPOUNDS	2.0.E+05	TRI (USEPA 2008)	9.9.E-03	Jolliet et al. 2003	2.0.E+03	0.028%	99.95%
ARSENIC COMPOUNDS	4.9.E+04	TRI (USEPA 2008)	3.9.E-02	Jolliet et al. 2003	1.9.E+03	0.026%	99.98%
ZINC COMPOUNDS	2.8.E+06	TRI (USEPA 2008)	2.6.E-04	Jolliet et al. 2003	7.2.E+02	0.010%	99.99%
SELENIUM COMPOUNDS	2.6.E+05	TRI (USEPA 2008)	5.1.E-04	Jolliet et al. 2003	1.3.E+02	0.002%	99.99%
CHROMIUM COMPOUNDS (EXCEPT CHROMITE ORE MINED IN THE TRANSVAAL REGION)	2.1.E+05	TRI (USEPA 2008)	3.9.E-04	Jolliet et al. 2003	8.2.E+01	0.001%	99.99%
ZINC (FUME OR DUST)	2.4.E+05	TRI (USEPA 2008)	2.6.E-04	Jolliet et al. 2003	6.3.E+01	0.001%	99.99%
MERCURY COMPOUNDS	5.5.E+04	TRI (USEPA 2008)	1.1.E-03	Jolliet et al. 2003	5.9.E+01	0.001%	100.00%
BENZENE	2.5.E+06	TRI (USEPA 2008)	2.1.E-05	Jolliet et al. 2003	5.2.E+01	0.001%	100.00%
FORMALDEHYDE	4.2.E+06	TRI (USEPA 2008)	1.2.E-05	Jolliet et al. 2003	4.9.E+01	0.001%	100.00%
HEXACHLOROBENZENE	1.5.E+02	TRI (USEPA 2008)	2.0.E-01	Jolliet et al. 2003	3.1.E+01	0.0%	100.00%
CHROMIUM	7.1.E+04	TRI (USEPA 2008)	3.9.E-04	Jolliet et al. 2003	2.8.E+01	0.0%	100.00%
BARIUM COMPOUNDS	9.3.E+05	TRI (USEPA 2008)	2.1.E-05	Jolliet et al. 2003	2.0.E+01	0.0%	100.00%
ARSENIC	4.3.E+02	TRI (USEPA 2008)	3.9.E-02	Jolliet et al. 2003	1.7.E+01	0.0%	100.00%
BROMOMETHANE	1.6.E+05	TRI (USEPA 2008)	6.2.E-05	Jolliet et al. 2003	1.0.E+01	0.0%	100.00%
CARBON TETRACHLORIDE	7.5.E+04	TRI (USEPA 2008)	1.1.E-04	Jolliet et al. 2003	8.2.E+00	0.0%	100.00%
AMMONIA	5.3.E+07	TRI (USEPA 2008)	1.4.E-07	Jolliet et al. 2003	7.6.E+00	0.0%	100.00%
ANTIMONY COMPOUNDS	1.7.E+04	TRI (USEPA 2008)	4.5.E-04	Jolliet et al. 2003	7.5.E+00	0.0%	100.00%
NICKEL COMPOUNDS	2.9.E+05	TRI (USEPA 2008)	2.5.E-05	Jolliet et al. 2003	7.2.E+00	0.0%	100.00%
CADMIUM COMPOUNDS	4.3.E+03	TRI (USEPA 2008)	1.6.E-03	Jolliet et al. 2003	7.0.E+00	0.0%	100.00%
SELENIUM	1.3.E+04	TRI (USEPA 2008)	5.1.E-04	Jolliet et al.	6.5.E+00	0.0%	100.00%

Pollutant/Chemical	Emissions (total air release (On-site Fugitive Air + On-site Point Source Air)) (in kg/y)	Source	Characterization factor (CF) for human health (DALY/kg)	Source	Damage score (DALY/y) (i.e., emissions × CF)	Contribution to total damage	Cumulative damage
		2008)		2003			
STYRENE	1.7.E+07	TRI (USEPA 2008)	3.7.E-07	Jolliet et al. 2003	6.4.E+00	0.0%	100.00%
ACROLEIN	7.4.E+04	TRI (USEPA 2008)	7.6.E-05	Jolliet et al. 2003	5.6.E+00	0.0%	100.00%
DIMETHOATE	4.7.E+03	TRI (USEPA 2008)	1.1.E-03	Jolliet et al. 2003	5.4.E+00	0.0%	100.00%
MERCURY	4.8.E+03	TRI (USEPA 2008)	1.1.E-03	Jolliet et al. 2003	5.1.E+00	0.0%	100.00%
ATRAZINE	6.1.E+03	TRI (USEPA 2008)	4.5.E-04	Jolliet et al. 2003	2.8.E+00	0.0%	100.00%
NICKEL	1.1.E+05	TRI (USEPA 2008)	2.5.E-05	Jolliet et al. 2003	2.8.E+00	0.0%	100.00%
LEAD COMPOUNDS	3.6.E+05	TRI (USEPA 2008)	7.3.E-06	Jolliet et al. 2003	2.7.E+00	0.0%	100.00%
BARIUM	1.2.E+05	TRI (USEPA 2008)	2.1.E-05	Jolliet et al. 2003	2.5.E+00	0.0%	100.00%
ACRYLAMIDE	5.4.E+03	TRI (USEPA 2008)	4.4.E-04	Jolliet et al. 2003	2.3.E+00	0.0%	100.00%
TOLUENE DIISOCYANATE (MIXED ISOMERS)	1.2.E+04	TRI (USEPA 2008)	1.9.E-04	Jolliet et al. 2003	2.3.E+00	0.0%	100.00%
PYRIDINE	1.5.E+04	TRI (USEPA 2008)	1.5.E-04	Jolliet et al. 2003	2.2.E+00	0.0%	100.00%
M-DINITROBENZENE	3.3.E+03	TRI (USEPA 2008)	6.1.E-04	Jolliet et al. 2003	2.0.E+00	0.0%	100.00%
1,2,4- TRIMETHYLBENZENE	3.1.E+06	TRI (USEPA 2008)	6.4.E-07	Jolliet et al. 2003	2.0.E+00	0.0%	100.00%
COPPER COMPOUNDS	3.2.E+05	TRI (USEPA 2008)	5.7.E-06	Jolliet et al. 2003	1.8.E+00	0.0%	100.00%
CHLOROFORM	2.7.E+05	TRI (USEPA 2008)	6.3.E-06	Jolliet et al. 2003	1.7.E+00	0.0%	100.00%
DICHLOROMETHANE	2.4.E+06	TRI (USEPA 2008)	6.7.E-07	Jolliet et al. 2003	1.6.E+00	0.0%	100.00%
ANTIMONY	3.3.E+03	TRI (USEPA 2008)	4.5.E-04	Jolliet et al. 2003	1.5.E+00	0.0%	100.00%
2,4-DIAMINOTOLUENE	5.5.E+02	TRI (USEPA 2008)	2.6.E-03	Jolliet et al. 2003	1.5.E+00	0.0%	100.00%
COPPER	2.5.E+05	TRI (USEPA 2008)	5.7.E-06	Jolliet et al. 2003	1.4.E+00	0.0%	100.00%
DI(2-ETHYLHEXYL) PHTHALATE	6.9.E+04	TRI (USEPA 2008)	1.8.E-05	Jolliet et al. 2003	1.2.E+00	0.0%	100.00%
EPICHLOROHYDRIN	6.0.E+04	TRI (USEPA 2008)	2.0.E-05	Jolliet et al. 2003	1.2.E+00	0.0%	100.00%
ACETALDEHYDE	4.8.E+06	TRI (USEPA 2008)	2.4.E-07	Jolliet et al. 2003	1.1.E+00	0.0%	100.00%
1,2-DICHLOROETHANE	1.5.E+05	TRI (USEPA 2008)	7.0.E-06	Jolliet et al. 2003	1.1.E+00	0.0%	100.00%
ACRYLONITRILE	2.0.E+05	TRI (USEPA 2008)	5.0.E-06	Jolliet et al. 2003	1.0.E+00	0.0%	100.00%
ALUMINUM (FUME OR DUST)	7.3.E+05	TRI (USEPA 2008)	1.4.E-06	Jolliet et al. 2003	1.0.E+00	0.0%	100.00%
ACEPHATE	4.5.E+02	TRI (USEPA 2008)	2.1.E-03	Jolliet et al. 2003	9.3.E-01	0.0%	100.00%
ETHYLENE OXIDE	1.3.E+05	TRI (USEPA 2008)	7.0.E-06	Jolliet et al. 2003	9.1.E-01	0.0%	100.00%
CADMIUM	5.5.E+02	TRI (USEPA 2008)	1.6.E-03	Jolliet et al.	8.9.E-01	0.0%	100.00%

Pollutant/Chemical	Emissions (total air release (On-site Fugitive Air + On-site Point Source Air)) (in kg/y)	Source	Characterization factor (CF) for human health (DALY/kg)	Source	Damage score (DALY/y) (i.e., emissions × CF)	Contribution to total damage	Cumulative damage
		2008)		2003			
LEAD	1.0.E+05	TRI (USEPA 2008)	7.3.E-06	Jolliet et al. 2003	7.5.E-01	0.0%	100.00%
METHANOL	5.9.E+07	TRI (USEPA 2008)	1.2.E-08	Jolliet et al. 2003	7.0.E-01	0.0%	100.00%
BENZO(G,H,I)PERYLENE	9.4.E+03	TRI (USEPA 2008)	6.2.E-05	Jolliet et al. 2003	5.8.E-01	0.0%	100.00%
PROPYLENE OXIDE	1.5.E+05	TRI (USEPA 2008)	3.3.E-06	Jolliet et al. 2003	5.0.E-01	0.0%	100.00%
NITROGLYCERIN	5.4.E+04	TRI (USEPA 2008)	9.1.E-06	Jolliet et al. 2003	4.9.E-01	0.0%	100.00%
1,2-DICHLOROPROPANE	5.0.E+04	TRI (USEPA 2008)	9.6.E-06	Jolliet et al. 2003	4.8.E-01	0.0%	100.00%
VINYL CHLORIDE	1.6.E+05	TRI (USEPA 2008)	3.0.E-06	Jolliet et al. 2003	4.7.E-01	0.0%	100.00%
CARBON DISULFIDE	4.1.E+06	TRI (USEPA 2008)	1.1.E-07	Jolliet et al. 2003	4.7.E-01	0.0%	100.00%
BERYLLIUM COMPOUNDS	2.3.E+03	TRI (USEPA 2008)	1.7.E-04	Jolliet et al. 2003	3.9.E-01	0.0%	100.00%
ACRYLIC ACID	1.3.E+05	TRI (USEPA 2008)	2.9.E-06	Jolliet et al. 2003	3.7.E-01	0.0%	100.00%
THIRAM	3.0.E+04	TRI (USEPA 2008)	1.1.E-05	Jolliet et al. 2003	3.4.E-01	0.0%	100.00%
TETRACHLOROETHYLENE	8.0.E+05	TRI (USEPA 2008)	3.7.E-07	Jolliet et al. 2003	3.0.E-01	0.0%	100.00%
TRICHLOROETHYLENE	2.0.E+06	TRI (USEPA 2008)	1.5.E-07	Jolliet et al. 2003	2.9.E-01	0.0%	100.00%
CHLORDANE	2.8.E+01	TRI (USEPA 2008)	1.0.E-02	Jolliet et al. 2003	2.9.E-01	0.0%	100.00%
ANILINE	6.2.E+04	TRI (USEPA 2008)	4.4.E-06	Jolliet et al. 2003	2.7.E-01	0.0%	100.00%
HEXACHLOROCYCLOPENTADIENE	4.2.E+02	TRI (USEPA 2008)	6.4.E-04	Jolliet et al. 2003	2.7.E-01	0.0%	100.00%
XYLENE (MIXED ISOMERS)	1.0.E+07	TRI (USEPA 2008)	2.4.E-08	Jolliet et al. 2003	2.5.E-01	0.0%	100.00%
DINITROBUTYL PHENOL	3.0.E+02	TRI (USEPA 2008)	6.7.E-04	Jolliet et al. 2003	2.0.E-01	0.0%	100.00%
2,6-DINITROTOLUENE	3.2.E+00	TRI (USEPA 2008)	5.5.E-02	Jolliet et al. 2003	1.7.E-01	0.0%	100.00%
TOLUENE	1.8.E+07	TRI (USEPA 2008)	9.5.E-09	Jolliet et al. 2003	1.7.E-01	0.0%	100.00%
N-METHYLOLACRYLAMIDE	2.9.E+03	TRI (USEPA 2008)	4.5.E-05	Jolliet et al. 2003	1.3.E-01	0.0%	100.00%
N-HEXANE	1.6.E+07	TRI (USEPA 2008)	8.3.E-09	Jolliet et al. 2003	1.3.E-01	0.0%	100.00%
TOXAPHENE	1.1.E+01	TRI (USEPA 2008)	1.0.E-02	Jolliet et al. 2003	1.2.E-01	0.0%	100.00%
HEXACHLOROPHENE	3.7.E+01	TRI (USEPA 2008)	3.1.E-03	Jolliet et al. 2003	1.1.E-01	0.0%	100.00%
CARBARYL	2.5.E+02	TRI (USEPA 2008)	4.3.E-04	Jolliet et al. 2003	1.1.E-01	0.0%	100.00%
NAPHTHALENE	7.2.E+05	TRI (USEPA 2008)	1.5.E-07	Jolliet et al. 2003	1.1.E-01	0.0%	100.00%
HYDROQUINONE	7.7.E+03	TRI (USEPA 2008)	1.2.E-05	Jolliet et al. 2003	9.5.E-02	0.0%	100.00%
BENZIDINE	2.6.E+00	TRI (USEPA 2008)	3.1.E-02	Jolliet et al. 2003	8.1.E-02	0.0%	100.00%

Pollutant/Chemical	Emissions (total air release (On-site Fugitive Air + On-site Point Source Air)) (in kg/y)	Source	Characterization factor (CF) for human health (DALY/kg)	Source	Damage score (DALY/y) (i.e., emissions × CF)	Contribution to total damage	Cumulative damage
CHLOROETHANE	3.2.E+05	TRI (USEPA 2008)	2.4.E-07	Jolliet et al. 2003	7.6.E-02	0.0%	100.00%
HEXACHLORO-1,3-BUTADIENE	1.9.E+02	TRI (USEPA 2008)	3.8.E-04	Jolliet et al. 2003	7.2.E-02	0.0%	100.00%
ACETONITRILE	2.1.E+05	TRI (USEPA 2008)	3.4.E-07	Jolliet et al. 2003	7.0.E-02	0.0%	100.00%
CHLORODIFLUOROMETHANE	3.0.E+06	TRI (USEPA 2008)	2.3.E-08	Jolliet et al. 2003	6.8.E-02	0.0%	100.00%
1,3-BUTADIENE	8.1.E+05	TRI (USEPA 2008)	8.2.E-08	Jolliet et al. 2003	6.7.E-02	0.0%	100.00%
HYDROGEN CYANIDE	4.7.E+05	TRI (USEPA 2008)	1.2.E-07	Jolliet et al. 2003	5.9.E-02	0.0%	100.00%
2,4-D	8.5.E+02	TRI (USEPA 2008)	6.7.E-05	Jolliet et al. 2003	5.7.E-02	0.0%	100.00%
DIBUTYL PHTHALATE	2.4.E+04	TRI (USEPA 2008)	2.3.E-06	Jolliet et al. 2003	5.4.E-02	0.0%	100.00%
2,4-DICHLOROPHENOL	2.0.E+03	TRI (USEPA 2008)	2.6.E-05	Jolliet et al. 2003	5.4.E-02	0.0%	100.00%
ETHYLENE THIOUREA	4.2.E+01	TRI (USEPA 2008)	1.2.E-03	Jolliet et al. 2003	4.8.E-02	0.0%	100.00%
DINITROTOLUENE (MIXED ISOMERS)	1.4.E+02	TRI (USEPA 2008)	3.1.E-04	Jolliet et al. 2003	4.2.E-02	0.0%	100.00%
VINYL ACETATE	9.2.E+05	TRI (USEPA 2008)	4.3.E-08	Jolliet et al. 2003	4.0.E-02	0.0%	100.00%
VINYLDENE CHLORIDE	2.5.E+04	TRI (USEPA 2008)	1.5.E-06	Jolliet et al. 2003	3.9.E-02	0.0%	100.00%
ETHYLENE GLYCOL	1.1.E+06	TRI (USEPA 2008)	3.5.E-08	Jolliet et al. 2003	3.8.E-02	0.0%	100.00%
URETHANE	2.2.E+03	TRI (USEPA 2008)	1.7.E-05	Jolliet et al. 2003	3.6.E-02	0.0%	100.00%
PHENOL	1.8.E+06	TRI (USEPA 2008)	1.7.E-08	Jolliet et al. 2003	3.1.E-02	0.0%	100.00%
1,1,2-TRICHLOROETHANE	9.8.E+03	TRI (USEPA 2008)	3.0.E-06	Jolliet et al. 2003	3.0.E-02	0.0%	100.00%
2-ACETYLAMINOFLUORENE	1.1.E+02	TRI (USEPA 2008)	2.5.E-04	Jolliet et al. 2003	2.8.E-02	0.0%	100.00%
DIPHENYLAMINE	1.5.E+04	TRI (USEPA 2008)	1.7.E-06	Jolliet et al. 2003	2.5.E-02	0.0%	100.00%
METHYL PARATHION	7.4.E+02	TRI (USEPA 2008)	3.2.E-05	Jolliet et al. 2003	2.4.E-02	0.0%	100.00%
1,3-PHENYLENEDIAMINE	1.3.E+03	TRI (USEPA 2008)	1.7.E-05	Jolliet et al. 2003	2.2.E-02	0.0%	100.00%
CHLOROMETHYL METHYL ETHER	1.6.E+03	TRI (USEPA 2008)	1.3.E-05	Jolliet et al. 2003	2.1.E-02	0.0%	100.00%
SIMAZINE	3.9.E+01	TRI (USEPA 2008)	5.4.E-04	Jolliet et al. 2003	2.1.E-02	0.0%	100.00%
4,4'-METHYLENEBIS(2-CHLOROANILINE)	1.8.E+03	TRI (USEPA 2008)	1.2.E-05	Jolliet et al. 2003	2.1.E-02	0.0%	100.00%
N-BUTYL ALCOHOL	5.9.E+06	TRI (USEPA 2008)	3.4.E-09	Jolliet et al. 2003	2.0.E-02	0.0%	100.00%
LINDANE	8.3.E+01	TRI (USEPA 2008)	2.4.E-04	Jolliet et al. 2003	2.0.E-02	0.0%	100.00%
MALATHION	3.0.E+03	TRI (USEPA 2008)	6.5.E-06	Jolliet et al. 2003	2.0.E-02	0.0%	100.00%
PENTACHLOROPHENOL	3.9.E+01	TRI (USEPA 2008)	4.5.E-04	Jolliet et al. 2003	1.7.E-02	0.0%	100.00%

Pollutant/Chemical	Emissions (total air release (On-site Fugitive Air + On-site Point Source Air)) (in kg/y)	Source	Characterization factor (CF) for human health (DALY/kg)	Source	Damage score (DALY/y) (i.e., emissions × CF)	Contribution to total damage	Cumulative damage
4,4'-ISOPROPYLIDENEDIPHENOL	5.6.E+04	TRI (USEPA 2008)	3.1.E-07	Jolliet et al. 2003	1.7.E-02	0.0%	100.00%
2,4-DINITROTOLUENE	1.1.E+03	TRI (USEPA 2008)	1.6.E-05	Jolliet et al. 2003	1.7.E-02	0.0%	100.00%
BERYLLIUM	8.9.E+01	TRI (USEPA 2008)	1.7.E-04	Jolliet et al. 2003	1.5.E-02	0.0%	100.00%
ACETAMIDE	7.3.E+02	TRI (USEPA 2008)	2.0.E-05	Jolliet et al. 2003	1.5.E-02	0.0%	100.00%
N,N-DIMETHYLFORMAMIDE	1.4.E+05	TRI (USEPA 2008)	9.7.E-08	Jolliet et al. 2003	1.3.E-02	0.0%	100.00%
1,2-DIBROMOETHANE	1.8.E+03	TRI (USEPA 2008)	7.2.E-06	Jolliet et al. 2003	1.3.E-02	0.0%	100.00%
PHENANTHRENE	2.6.E+04	TRI (USEPA 2008)	4.8.E-07	Jolliet et al. 2003	1.2.E-02	0.0%	100.00%
CATECHOL	1.1.E+03	TRI (USEPA 2008)	1.1.E-05	Jolliet et al. 2003	1.2.E-02	0.0%	100.00%
4,4'-DIAMINODIPHENYL ETHER	1.6.E+02	TRI (USEPA 2008)	7.1.E-05	Jolliet et al. 2003	1.1.E-02	0.0%	100.00%
OXYDIAZON	1.2.E+02	TRI (USEPA 2008)	9.2.E-05	Jolliet et al. 2003	1.1.E-02	0.0%	100.00%
DIURON	1.5.E+02	TRI (USEPA 2008)	6.6.E-05	Jolliet et al. 2003	9.9.E-03	0.0%	100.00%
1,1,2,2-TETRACHLOROETHANE	6.4.E+02	TRI (USEPA 2008)	1.5.E-05	Jolliet et al. 2003	9.8.E-03	0.0%	100.00%
1-CHLORO-1,1-DIFLUOROETHANE	2.7.E+06	TRI (USEPA 2008)	3.5.E-09	Jolliet et al. 2003	9.7.E-03	0.0%	100.00%
MALEIC ANHYDRIDE	1.7.E+05	TRI (USEPA 2008)	4.3.E-08	Jolliet et al. 2003	7.3.E-03	0.0%	100.00%
TRIS(2,3-DIBROMOPROPYL) PHOSPHATE	1.1.E+02	TRI (USEPA 2008)	4.9.E-05	Jolliet et al. 2003	5.5.E-03	0.0%	100.00%
TRIFLURALIN	3.0.E+02	TRI (USEPA 2008)	1.5.E-05	Jolliet et al. 2003	4.6.E-03	0.0%	100.00%
HYDRAZINE	6.2.E+02	TRI (USEPA 2008)	7.4.E-06	Jolliet et al. 2003	4.5.E-03	0.0%	100.00%
1,1,1,2-TETRACHLOROETHANE	8.2.E+02	TRI (USEPA 2008)	4.9.E-06	Jolliet et al. 2003	4.0.E-03	0.0%	100.00%
PERMETHRIN	1.6.E+02	TRI (USEPA 2008)	2.2.E-05	Jolliet et al. 2003	3.3.E-03	0.0%	100.00%
PROPANE SULFONE	1.1.E+02	TRI (USEPA 2008)	2.9.E-05	Jolliet et al. 2003	3.3.E-03	0.0%	100.00%
1,2,3-TRICHLOROPROPANE	5.1.E+02	TRI (USEPA 2008)	6.3.E-06	Jolliet et al. 2003	3.2.E-03	0.0%	100.00%
PROPANIL	2.3.E+02	TRI (USEPA 2008)	1.4.E-05	Jolliet et al. 2003	3.2.E-03	0.0%	100.00%
AMETRYN	1.4.E+02	TRI (USEPA 2008)	2.3.E-05	Jolliet et al. 2003	3.1.E-03	0.0%	100.00%
PENTACHLOROETHANE	1.2.E+04	TRI (USEPA 2008)	2.4.E-07	Jolliet et al. 2003	2.9.E-03	0.0%	100.00%
P-CHLOROANILINE	1.2.E+02	TRI (USEPA 2008)	2.3.E-05	Jolliet et al. 2003	2.7.E-03	0.0%	100.00%
DICAMBA	5.1.E+01	TRI (USEPA 2008)	5.2.E-05	Jolliet et al. 2003	2.6.E-03	0.0%	100.00%
1,4-DIOXANE	5.7.E+04	TRI (USEPA 2008)	4.3.E-08	Jolliet et al. 2003	2.4.E-03	0.0%	100.00%
MECOPROP	7.3.E+01	TRI (USEPA 2008)	3.2.E-05	Jolliet et al.	2.3.E-03	0.0%	100.00%

Pollutant/Chemical	Emissions (total air release (On-site Fugitive Air + On-site Point Source Air)) (in kg/y)	Source	Characterization factor (CF) for human health (DALY/kg)	Source	Damage score (DALY/y) (i.e., emissions × CF)	Contribution to total damage	Cumulative damage
		2008)		2003			
CHLOROTHALONIL	2.2.E+03	TRI (USEPA 2008)	1.0.E-06	Jolliet et al. 2003	2.3.E-03	0.0%	100.00%
PENTACHLOROENZENE	9.8.E+01	TRI (USEPA 2008)	2.1.E-05	Jolliet et al. 2003	2.1.E-03	0.0%	100.00%
QUINTOZENE	1.0.E+02	TRI (USEPA 2008)	2.0.E-05	Jolliet et al. 2003	2.0.E-03	0.0%	100.00%
2-NITROPROPANE	1.2.E+04	TRI (USEPA 2008)	1.7.E-07	Jolliet et al. 2003	2.0.E-03	0.0%	100.00%
MERPHOS	1.4.E+00	TRI (USEPA 2008)	1.4.E-03	Jolliet et al. 2003	2.0.E-03	0.0%	100.00%
TETRACHLORVINPHOS	1.6.E+02	TRI (USEPA 2008)	1.2.E-05	Jolliet et al. 2003	2.0.E-03	0.0%	100.00%
HEPTACHLOR	3.9.E+00	TRI (USEPA 2008)	4.8.E-04	Jolliet et al. 2003	1.9.E-03	0.0%	100.00%
PROPOXUR	6.7.E+00	TRI (USEPA 2008)	2.6.E-04	Jolliet et al. 2003	1.7.E-03	0.0%	100.00%
2-MERCAPTOBENZOTHAZOLE	1.0.E+03	TRI (USEPA 2008)	1.7.E-06	Jolliet et al. 2003	1.7.E-03	0.0%	100.00%
THIOUREA	2.3.E+02	TRI (USEPA 2008)	7.2.E-06	Jolliet et al. 2003	1.7.E-03	0.0%	100.00%
2,4-DIMETHYLPHENOL	7.1.E+03	TRI (USEPA 2008)	2.3.E-07	Jolliet et al. 2003	1.7.E-03	0.0%	100.00%
2,4-DINITROPHENOL	7.3.E+00	TRI (USEPA 2008)	2.2.E-04	Jolliet et al. 2003	1.6.E-03	0.0%	100.00%
ALDRIN	1.7.E+01	TRI (USEPA 2008)	9.3.E-05	Jolliet et al. 2003	1.5.E-03	0.0%	100.00%
ALACHLOR	1.7.E+02	TRI (USEPA 2008)	9.1.E-06	Jolliet et al. 2003	1.5.E-03	0.0%	100.00%
CARBOFURAN	7.7.E+01	TRI (USEPA 2008)	1.9.E-05	Jolliet et al. 2003	1.5.E-03	0.0%	100.00%
TRIBENURON METHYL	2.6.E+02	TRI (USEPA 2008)	5.6.E-06	Jolliet et al. 2003	1.4.E-03	0.0%	100.00%
HEXACHLOROETHANE	2.7.E+02	TRI (USEPA 2008)	5.2.E-06	Jolliet et al. 2003	1.4.E-03	0.0%	100.00%
NITROBENZENE	1.1.E+04	TRI (USEPA 2008)	1.3.E-07	Jolliet et al. 2003	1.4.E-03	0.0%	100.00%
ALLYL CHLORIDE	1.2.E+04	TRI (USEPA 2008)	1.1.E-07	Jolliet et al. 2003	1.3.E-03	0.0%	100.00%
3,3'-DIMETHOXYBENZIDINE DIHYDROCHLORIDE	1.2.E+02	TRI (USEPA 2008)	1.1.E-05	Jolliet et al. 2003	1.3.E-03	0.0%	100.00%
4-DIMETHYLAMINOAZOBENZENE	1.1.E+02	TRI (USEPA 2008)	1.1.E-05	Jolliet et al. 2003	1.2.E-03	0.0%	100.00%
PARAQUAT DICHLORIDE	3.0.E+01	TRI (USEPA 2008)	3.8.E-05	Jolliet et al. 2003	1.2.E-03	0.0%	100.00%
CAPTAN	2.7.E+02	TRI (USEPA 2008)	4.0.E-06	Jolliet et al. 2003	1.1.E-03	0.0%	100.00%
ETHYLBENZENE	1.7.E+06	TRI (USEPA 2008)	6.3.E-10	Jolliet et al. 2003	1.1.E-03	0.0%	100.00%
BIPHENYL	1.6.E+05	TRI (USEPA 2008)	6.1.E-09	Jolliet et al. 2003	1.0.E-03	0.0%	100.00%

Pollutant/Chemical	Emissions (total air release (On-site Fugitive Air + On-site Point Source Air)) (in kg/y)	Source	Characterization factor (CF) for human health (DALY/kg)	Source	Damage score (DALY/y) (i.e., emissions × CF)	Contribution to total damage	Cumulative damage
AMITROLE	4.5.E+00	TRI (USEPA 2008)	2.2.E-04	Jolliet et al. 2003	9.8.E-04	0.0%	100.00%
CHLOROBENZILATE	9.8.E+00	TRI (USEPA 2008)	9.6.E-05	Jolliet et al. 2003	9.4.E-04	0.0%	100.00%
TRIETHYLAMINE	1.9.E+05	TRI (USEPA 2008)	4.8.E-09	Jolliet et al. 2003	9.4.E-04	0.0%	100.00%
METHYL TERT-BUTYL ETHER	2.4.E+05	TRI (USEPA 2008)	3.7.E-09	Jolliet et al. 2003	8.8.E-04	0.0%	100.00%
DICHLORVOS	1.8.E+02	TRI (USEPA 2008)	4.9.E-06	Jolliet et al. 2003	8.7.E-04	0.0%	100.00%
BENZOIC TRICHLORIDE	2.9.E+02	TRI (USEPA 2008)	2.9.E-06	Jolliet et al. 2003	8.4.E-04	0.0%	100.00%
1,4-DICHLOROBENZENE	3.6.E+04	TRI (USEPA 2008)	2.1.E-08	Jolliet et al. 2003	7.6.E-04	0.0%	100.00%
PENDIMETHALIN	7.6.E+02	TRI (USEPA 2008)	9.8.E-07	Jolliet et al. 2003	7.4.E-04	0.0%	100.00%
LINURON	6.4.E+00	TRI (USEPA 2008)	1.1.E-04	Jolliet et al. 2003	7.0.E-04	0.0%	100.00%
1,2,4-TRICHLOROBENZENE	9.9.E+03	TRI (USEPA 2008)	6.9.E-08	Jolliet et al. 2003	6.9.E-04	0.0%	100.00%
1,2-BUTYLENE OXIDE	8.3.E+02	TRI (USEPA 2008)	8.1.E-07	Jolliet et al. 2003	6.7.E-04	0.0%	100.00%
PRONAMIDE	4.9.E+00	TRI (USEPA 2008)	1.3.E-04	Jolliet et al. 2003	6.4.E-04	0.0%	100.00%
BIFENTHRIN	2.0.E+00	TRI (USEPA 2008)	3.2.E-04	Jolliet et al. 2003	6.3.E-04	0.0%	100.00%
ALDICARB	6.0.E+01	TRI (USEPA 2008)	1.0.E-05	Jolliet et al. 2003	6.1.E-04	0.0%	100.00%
ALLYL ALCOHOL	1.3.E+04	TRI (USEPA 2008)	4.0.E-08	Jolliet et al. 2003	5.3.E-04	0.0%	100.00%
ACETOPHENONE	4.9.E+04	TRI (USEPA 2008)	1.1.E-08	Jolliet et al. 2003	5.3.E-04	0.0%	100.00%
2,4,6-TRICHLOROPHENOL	1.1.E+02	TRI (USEPA 2008)	5.0.E-06	Jolliet et al. 2003	5.3.E-04	0.0%	100.00%
CHLOROBENZENE	2.1.E+05	TRI (USEPA 2008)	2.4.E-09	Jolliet et al. 2003	5.0.E-04	0.0%	100.00%
2-CHLORO-1,1,1-TRIFLUOROETHANE	2.2.E+04	TRI (USEPA 2008)	2.2.E-08	Jolliet et al. 2003	4.9.E-04	0.0%	100.00%
PROMETRYN	1.2.E+02	TRI (USEPA 2008)	4.1.E-06	Jolliet et al. 2003	4.7.E-04	0.0%	100.00%
METHOXYCHLOR	1.2.E+01	TRI (USEPA 2008)	3.8.E-05	Jolliet et al. 2003	4.6.E-04	0.0%	100.00%
CUMENE	4.8.E+05	TRI (USEPA 2008)	7.9.E-10	Jolliet et al. 2003	3.8.E-04	0.0%	100.00%
2-ETHOXYETHANOL	2.7.E+04	TRI (USEPA 2008)	1.4.E-08	Jolliet et al. 2003	3.8.E-04	0.0%	100.00%
M-CRESOL	1.3.E+04	TRI (USEPA 2008)	2.9.E-08	Jolliet et al. 2003	3.7.E-04	0.0%	100.00%
CYFLUTHRIN	1.6.E+01	TRI (USEPA 2008)	2.2.E-05	Jolliet et al. 2003	3.5.E-04	0.0%	100.00%
DIMETHYLAMINE	5.2.E+04	TRI (USEPA 2008)	5.4.E-09	Jolliet et al. 2003	2.8.E-04	0.0%	100.00%
DIMETHIPIN	6.3.E+01	TRI (USEPA 2008)	4.3.E-06	Jolliet et al. 2003	2.7.E-04	0.0%	100.00%
ANTHRACENE	5.6.E+03	TRI (USEPA 2008)	4.6.E-08	Jolliet et al. 2003	2.6.E-04	0.0%	100.00%
CHLORSULFURON	1.9.E+01	TRI (USEPA 2008)	1.4.E-05	Jolliet et al.	2.6.E-04	0.0%	100.00%

Pollutant/Chemical	Emissions (total air release (On-site Fugitive Air + On-site Point Source Air)) (in kg/y)	Source	Characterization factor (CF) for human health (DALY/kg)	Source	Damage score (DALY/y) (i.e., emissions × CF)	Contribution to total damage	Cumulative damage
		2008)		2003			
METHOXONE	5.9.E+00	TRI (USEPA 2008)	4.2.E-05	Jolliet et al. 2003	2.5.E-04	0.0%	100.00%
BENZYL CHLORIDE	5.8.E+03	TRI (USEPA 2008)	4.2.E-08	Jolliet et al. 2003	2.4.E-04	0.0%	100.00%
MYCLOBUTANIL	8.2.E+00	TRI (USEPA 2008)	2.8.E-05	Jolliet et al. 2003	2.3.E-04	0.0%	100.00%
BIS(2-CHLOROETHYL) ETHER	4.1.E+01	TRI (USEPA 2008)	5.4.E-06	Jolliet et al. 2003	2.2.E-04	0.0%	100.00%
BROMOXYNIL	5.9.E+00	TRI (USEPA 2008)	3.7.E-05	Jolliet et al. 2003	2.2.E-04	0.0%	100.00%
2-METHOXYETHANOL	1.0.E+04	TRI (USEPA 2008)	2.0.E-08	Jolliet et al. 2003	2.1.E-04	0.0%	100.00%
LACTOFEN	2.3.E+00	TRI (USEPA 2008)	8.9.E-05	Jolliet et al. 2003	2.0.E-04	0.0%	100.00%
PHENYTOIN	1.8.E+01	TRI (USEPA 2008)	1.0.E-05	Jolliet et al. 2003	1.8.E-04	0.0%	100.00%
N,N-DIMETHYLANILINE	2.3.E+02	TRI (USEPA 2008)	7.1.E-07	Jolliet et al. 2003	1.7.E-04	0.0%	100.00%
S,S,S-TRIBUTYLTRITHIOPHOSPHATE	1.4.E+00	TRI (USEPA 2008)	1.2.E-04	Jolliet et al. 2003	1.6.E-04	0.0%	100.00%
4-AMINOBIPHENYL	4.5.E-01	TRI (USEPA 2008)	2.7.E-04	Jolliet et al. 2003	1.2.E-04	0.0%	100.00%
TRICHLOROFLUOROMETHANE	1.3.E+05	TRI (USEPA 2008)	9.0.E-10	Jolliet et al. 2003	1.2.E-04	0.0%	100.00%
METRIBUZIN	2.0.E+01	TRI (USEPA 2008)	4.5.E-06	Jolliet et al. 2003	9.1.E-05	0.0%	100.00%
PROPARGYL ALCOHOL	2.2.E+02	TRI (USEPA 2008)	3.9.E-07	Jolliet et al. 2003	8.7.E-05	0.0%	100.00%
DIHYDROSAFROLE	2.3.E+02	TRI (USEPA 2008)	3.7.E-07	Jolliet et al. 2003	8.3.E-05	0.0%	100.00%
O-CRESOL	5.0.E+03	TRI (USEPA 2008)	1.6.E-08	Jolliet et al. 2003	8.2.E-05	0.0%	100.00%
DICHLORODIFLUOROMETHANE	1.2.E+05	TRI (USEPA 2008)	6.3.E-10	Jolliet et al. 2003	7.7.E-05	0.0%	100.00%
FOLPET	1.5.E+01	TRI (USEPA 2008)	4.5.E-06	Jolliet et al. 2003	6.9.E-05	0.0%	100.00%
BROMOFORM	8.7.E+01	TRI (USEPA 2008)	6.8.E-07	Jolliet et al. 2003	5.9.E-05	0.0%	100.00%
ETHYLENEIMINE	1.9.E+00	TRI (USEPA 2008)	3.0.E-05	Jolliet et al. 2003	5.6.E-05	0.0%	100.00%
METHYL METHACRYLATE	1.1.E+06	TRI (USEPA 2008)	4.1.E-11	Jolliet et al. 2003	4.4.E-05	0.0%	100.00%
FREON 113	2.6.E+05	TRI (USEPA 2008)	1.7.E-10	Jolliet et al. 2003	4.3.E-05	0.0%	100.00%
TRIALATE	1.7.E+01	TRI (USEPA 2008)	2.4.E-06	Jolliet et al. 2003	4.2.E-05	0.0%	100.00%
TEBUTHIURON	4.5.E+00	TRI (USEPA 2008)	9.0.E-06	Jolliet et al. 2003	4.1.E-05	0.0%	100.00%
1,2-DICHLOROBENZENE	1.7.E+04	TRI (USEPA 2008)	2.3.E-09	Jolliet et al. 2003	4.0.E-05	0.0%	100.00%
DIGLYCIDYL RESORCINOL ETHER	4.5.E-01	TRI (USEPA 2008)	8.7.E-05	Jolliet et al. 2003	4.0.E-05	0.0%	100.00%
N-NITROSODI-N-PROPYLAMINE	1.1.E+02	TRI (USEPA 2008)	3.2.E-07	Jolliet et al. 2003	3.6.E-05	0.0%	100.00%
ETHYL ACRYLATE	3.2.E+04	TRI (USEPA 2008)	1.1.E-09	Jolliet et al.	3.5.E-05	0.0%	100.00%

Pollutant/Chemical	Emissions (total air release (On-site Fugitive Air + On-site Point Source Air)) (in kg/y)	Source	Characterization factor (CF) for human health (DALY/kg)	Source	Damage score (DALY/y) (i.e., emissions × CF)	Contribution to total damage	Cumulative damage
		2008)		2003			
CHLORENDIC ACID	3.6.E+00	TRI (USEPA 2008)	8.0.E-06	Jolliet et al. 2003	2.9.E-05	0.0%	100.00%
SAFROLE	2.3.E+02	TRI (USEPA 2008)	9.5.E-08	Jolliet et al. 2003	2.2.E-05	0.0%	100.00%
1,1-DIMETHYL HYDRAZINE	2.4.E+00	TRI (USEPA 2008)	7.6.E-06	Jolliet et al. 2003	1.8.E-05	0.0%	100.00%
PROPICONAZOLE	1.4.E+00	TRI (USEPA 2008)	7.8.E-06	Jolliet et al. 2003	1.1.E-05	0.0%	100.00%
CARBOXIN	6.4.E+00	TRI (USEPA 2008)	1.4.E-06	Jolliet et al. 2003	9.0.E-06	0.0%	100.00%
FLUOMETURON	4.5.E-01	TRI (USEPA 2008)	1.9.E-05	Jolliet et al. 2003	8.4.E-06	0.0%	100.00%
STYRENE OXIDE	3.8.E+01	TRI (USEPA 2008)	1.8.E-07	Jolliet et al. 2003	6.8.E-06	0.0%	100.00%
DIMETHYLCARBAMYL CHLORIDE	2.3.E+00	TRI (USEPA 2008)	2.6.E-06	Jolliet et al. 2003	5.9.E-06	0.0%	100.00%
PROPARGITE	2.2.E+01	TRI (USEPA 2008)	2.7.E-07	Jolliet et al. 2003	5.8.E-06	0.0%	100.00%
HYDRAMETHYLNON	1.0.E+01	TRI (USEPA 2008)	4.3.E-07	Jolliet et al. 2003	4.4.E-06	0.0%	100.00%
DICHLOROBROMOMETHANE	1.3.E+02	TRI (USEPA 2008)	3.2.E-08	Jolliet et al. 2003	4.4.E-06	0.0%	100.00%
BROMOXYNIL OCTANOATE	1.2.E+01	TRI (USEPA 2008)	3.1.E-07	Jolliet et al. 2003	3.7.E-06	0.0%	100.00%
ETHYL DIPROPYLTHIOCARBAMATE	2.3.E+01	TRI (USEPA 2008)	1.6.E-07	Jolliet et al. 2003	3.7.E-06	0.0%	100.00%
BIS(2-CHLORO-1-METHYLETHYL) ETHER	3.3.E+02	TRI (USEPA 2008)	1.1.E-08	Jolliet et al. 2003	3.5.E-06	0.0%	100.00%
1,3-DICHLOROPROPYLENE	2.5.E+03	TRI (USEPA 2008)	1.4.E-09	Jolliet et al. 2003	3.5.E-06	0.0%	100.00%
BENFLURALIN	8.0.E+01	TRI (USEPA 2008)	3.7.E-08	Jolliet et al. 2003	3.0.E-06	0.0%	100.00%
P-CRESIDINE	4.5.E+00	TRI (USEPA 2008)	4.1.E-07	Jolliet et al. 2003	1.9.E-06	0.0%	100.00%
1,2-DIBROMO-3-CHLOROPROPANE	1.4.E-02	TRI (USEPA 2008)	8.8.E-05	Jolliet et al. 2003	1.2.E-06	0.0%	100.00%
NALED	4.5.E+00	TRI (USEPA 2008)	1.7.E-07	Jolliet et al. 2003	7.5.E-07	0.0%	100.00%
3-CHLORO-2-METHYL-1-PROPENE	3.0.E+03	TRI (USEPA 2008)	1.0.E-10	Jolliet et al. 2003	3.1.E-07	0.0%	100.00%
PROPACHLOR	4.5.E-01	TRI (USEPA 2008)	4.9.E-07	Jolliet et al. 2003	2.2.E-07	0.0%	100.00%
CHLORIMURON ETHYL	8.6.E+00	TRI (USEPA 2008)	2.2.E-08	Jolliet et al. 2003	1.9.E-07	0.0%	100.00%
NITRILOTRIACETIC ACID	9.5.E+00	TRI (USEPA 2008)	1.6.E-08	Jolliet et al. 2003	1.5.E-07	0.0%	100.00%
METHACRYLONITRILE	4.5.E+00	TRI (USEPA 2008)	1.9.E-08	Jolliet et al. 2003	8.7.E-08	0.0%	100.00%
O-TOLUIDINE HYDROCHLORIDE	1.4.E-02	TRI (USEPA 2008)	5.1.E-06	Jolliet et al. 2003	6.9.E-08	0.0%	100.00%
2,4,5-TRICHLOROPHENOL	2.7.E-02	TRI (USEPA 2008)	1.3.E-06	Jolliet et al. 2003	3.6.E-08	0.0%	100.00%
TRANS-1,4-DICHLORO-2-BUTENE	2.3.E-01	TRI (USEPA 2008)	1.5.E-07	Jolliet et al. 2003	3.3.E-08	0.0%	100.00%
N-	4.5.E+00	TRI (USEPA	1.6.E-09	Jolliet et al.	7.4.E-09	0.0%	100.00%

Pollutant/Chemical	Emissions (total air release (On-site Fugitive Air + On-site Point Source Air)) (in kg/y)	Source	Characterization factor (CF) for human health (DALY/kg)	Source	Damage score (DALY/y) (i.e., emissions × CF)	Contribution to total damage	Cumulative damage
NITROSODIPHENYLAMINE		2008)		2003			
1,2-DIPHENYLHYDRAZINE	0.0.E+00	TRI (USEPA 2008)	6.9.E-06	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
2,4-DB	0.0.E+00	TRI (USEPA 2008)	1.6.E-06	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
2-PHENYLPHENOL	0.0.E+00	TRI (USEPA 2008)	1.2.E-06	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
5-NITRO-O-TOLUIDINE	0.0.E+00	TRI (USEPA 2008)	2.0.E-05	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
ACIFLUORFEN, SODIUM SALT	0.0.E+00	TRI (USEPA 2008)	1.0.E-04	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
AMITRAZ	0.0.E+00	TRI (USEPA 2008)	1.3.E-05	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
BIS(CHLOROMETHYL) ETHER	0.0.E+00	TRI (USEPA 2008)	3.8.E-02	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
CUPFERRON	0.0.E+00	TRI (USEPA 2008)	1.6.E-05	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
CYHALOTHRIN	0.0.E+00	TRI (USEPA 2008)	2.9.E-04	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
DIALLATE	0.0.E+00	TRI (USEPA 2008)	2.8.E-04	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
DIFLUBENZURON	0.0.E+00	TRI (USEPA 2008)	3.7.E-06	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
FENPROPATHRIN	0.0.E+00	TRI (USEPA 2008)	1.7.E-06	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
FLUOROURACIL	0.0.E+00	TRI (USEPA 2008)	5.4.E-04	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
MANEB	0.0.E+00	TRI (USEPA 2008)	4.1.E-07	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
METHYL HYDRAZINE	0.0.E+00	TRI (USEPA 2008)	1.4.E-08	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
MOLINATE	0.0.E+00	TRI (USEPA 2008)	7.9.E-06	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
N-NITROSO-N-ETHYLUREA	0.0.E+00	TRI (USEPA 2008)	2.7.E-04	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
N-NITROSO-N-METHYLUREA	0.0.E+00	TRI (USEPA 2008)	8.4.E-03	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
N-NITROSODI-N-BUTYLAMINE	0.0.E+00	TRI (USEPA 2008)	4.1.E-05	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
N-NITROSODIETHYLAMINE	0.0.E+00	TRI (USEPA 2008)	6.7.E-02	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
N-NITROSODIMETHYLAMINE	0.0.E+00	TRI (USEPA 2008)	1.7.E-05	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
N-NITROSOPIPERIDINE	0.0.E+00	TRI (USEPA 2008)	1.6.E-05	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
NORFLURAZON	0.0.E+00	TRI (USEPA 2008)	4.3.E-06	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
ORYZALIN	0.0.E+00	TRI (USEPA 2008)	1.5.E-06	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
OXYFLUORFEN	0.0.E+00	TRI (USEPA 2008)	2.5.E-05	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
QUINONE	0.0.E+00	TRI (USEPA 2008)	7.0.E-08	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
QUIZALOFOP-ETHYL	0.0.E+00	TRI (USEPA 2008)	4.2.E-06	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%

Pollutant/Chemical	Emissions (total air release (On-site Fugitive Air + On-site Point Source Air)) (in kg/y)	Source	Characterization factor (CF) for human health (DALY/kg)	Source	Damage score (DALY/y) (i.e., emissions × CF)	Contribution to total damage	Cumulative damage
RESMETHRIN	0.0.E+00	TRI (USEPA 2008)	6.2.E-07	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
SETHOXYDIM	0.0.E+00	TRI (USEPA 2008)	4.1.E-09	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
SODIUM O-PHENYLPHENOXIDE	0.0.E+00	TRI (USEPA 2008)	4.0.E-07	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
THIOACETAMIDE	0.0.E+00	TRI (USEPA 2008)	3.3.E-07	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
THIOBENCARB	0.0.E+00	TRI (USEPA 2008)	1.1.E-05	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
VINCLOZOLIN	0.0.E+00	TRI (USEPA 2008)	3.9.E-06	Jolliet et al. 2003	0.0.E+00	0.0%	100.00%
Total	1.5E+11				7.2E+6		
Other TRI chemicals not evaluated: 221	3.7E+08						
Total	1.5E+11						

6. Uptake fraction: Definition and application to particulate matter

In Chapter 5 I conclude that PM is often found to dominate the human health damage from outdoor airborne pollutants in life-cycle assessment. Indeed, PM is recognized as an important source of adverse health effects caused by air pollution. However, current life-cycle impact assessment methods characterize PM incompletely.

The IMPACT North America model developed in Chapter 2 is well suited to evaluate the fate and exposure for organic pollutants. However, though it is also used to evaluate the fate and exposure for inorganic pollutants (heavy metals, PM, etc.), better approaches exist. Since PM is often recognized as an important source of adverse health effects, special emphasis is placed on PM to better evaluate its fate, exposure and effect. Chapter 4 reviewed the different studies evaluating intake fractions and effect factors for PM.

However, the work performed in Chapters 2 and 4 has its modeling solely based on mass of PM inhaled. I explore here (i.e., Chapters 6, 7 and 8) the possibility of modeling characterization factors of PM using additional parameters such as particle size distribution and particle surface. In addition to the size, the modeling of composition of PM is also important to study. However, I decided to focus the work on size and keep composition outside of the scope of chapters 6, 7 and 8. Including composition in the analysis is important and would require significant future work.

This work is exploratory. It is not based on a detailed review of work that has been previously performed in this field. The work is mainly based on the knowledge acquired during the class “CE218B - Air Pollutant Dynamics” taught by Professor William Nazaroff. Therefore, Chapters 6, 7 and 8 should be taken as exploratory and results interpreted with care. Should this approach be recognized as valid, I would advise to revisit the modeling, addressing systematically the limitations and simplifications mentioned throughout Chapters 6, 7 and 8 and grounding it in a deeper literature. However, the scope of this work would be a Doctoral dissertation in itself and is outside of the present dissertation that aims to explore this approach within a broader issue of regionalization in life-cycle assessment.

This chapter introduces the concept of the uptake fraction metric and demonstrates its potential to improve the human health damage assessment from pollutants such as PM by considering how the particle size distribution influences respiratory tract deposition.

6.1. Summary

I present the concept of the uptake fraction, the fraction of emitted pollutant that comes into direct contact with organs of the body for dermal, oral or inhalation exposure. This metric is of particular interest for PM, which is a significant source of adverse health effects worldwide. PM health damage depends on how far particles penetrate into the lungs, which in turn depends on particle size distribution. I show that for the same level of intake, the uptake fraction can vary by up to a factor of three, depending on the particle size distribution. Furthermore, the zone of particle deposition within the respiratory tract also differs with particle size distribution. The uptake fraction can improve the assessment of PM damage by considering how the particle size distribution influences deposition in the respiratory tract system.

6.2. Introduction

I present here the concept of the uptake fraction metric and illustrate its use in health studies for PM. By explicitly addressing how pollutants’ characteristics influences deposition in

the organs, the uptake fraction extends the widely used intake fraction metric (Bennett et al. 2002, Smith et al. 2002).

The case of PM

This concept is especially relevant to improve the assessment of PM exposures that are shown to cause some of the most serious adverse health effects (Dockery et al. 1993, Dockey and Pope 1994, Pope et al. 1995, 2002, 2009, Kuenzli et al. 2000, Laden et al. 2000, 2006, Nel 2005, Schwarz et al. 2008).

Particle size distribution and composition (Franklin et al. 2008) play a crucial role in the relative human health damage of PM. Nel (2005) recommends that in addition to the PM mass, particle number should also be monitored in health studies for ultrafine particles. Froines (2006) and Oberdörster et al. (2005) stress that particle surface area or diameter may be a better measure of toxicity than total particle mass or number. Size can be an important factor because different particulate sizes deposit in different zones of the lung. When they deposit, individual particles may elicit a health response, either because they are foreign objects or because of the chemicals sorbed on their surface. Indeed, some of the observed adverse health effects are partly associated with chemicals sorbed on the surface of particles, and the incidence of such effects has been found to increase with total surface area of PM deposition in the lungs (Nel 2005).

Ultrafine particles (smaller than 0.1 μm) are potentially the most dangerous (Nel 2005). Human health risk has been observed to increase with decreasing particle size (Peters et al. 1997). Mayer et al. (2001) suggest that the particles deposited into the nasopharyngeal zone are eliminated by natural defense systems, whereas particles penetrating the tracheobronchial and the alveolar zones are less readily removed and can cause adverse health effects.

A number of recent studies have improved our ability to quantify PM intake fractions for specific outdoor sources (Wolff 2000, Krewitt et al. 2001, Nigge 2001a, 2001b, Evans 2002, Levy et al. 2002, 2003, Li and Hao 2003, Marshall et al. 2003, 2005, Marshall and Behrentz 2005, Heath et al. 2006, Greco et al. 2007, Van Zelm et al. 2008, and see Chapter 4) and for indoor emissions (Wilson 2003, Nazaroff 2008, Hellweg et al. 2009, Siddiqui et al. 2008). Yet few have accounted for the influence of the specific particle size distribution on the resulting internal PM exposures and damage (see Chapter 4). At most, these studies differentiate among PM sizes that are smaller than 2.5 μm ($\text{PM}_{2.5}$), between 2.5 μm and 10 μm ($\text{PM}_{10-2.5}$), and greater than 10 μm . Lai et al. (2000) evaluated intake fractions (that they called cumulative population inhalation transfer factor (PITF)) for PM emitted indoor as a function of particle diameter. Lazaridis et al. (2001) developed an approach for modeling the source to lung-deposition chain of events. However, no studies calculated characterization factors for different classes of PM smaller than 2.5 μm within a life-cycle assessment context.

This chapter addresses how to define an uptake fraction metric in relation to the intake fraction metric, how to model the uptake fraction, and examines how the uptake fraction changes with the particle size distribution.

6.3. The uptake fraction as an extension of the intake fraction

Intake fraction

The intake fraction (iF) was defined by Bennett et al. (2002) as:

$$iF = \frac{\sum_{\text{people,time}} \text{intake of pollutant in an individual (kg)}}{\text{mass released into the environment (kg)}} \quad (6-1)$$

and can also be expressed as the ratio of an intake rate to an emission source strength in units of inhaled parts per million (ppm):

$$iF = \frac{\sum(N \times BR \times C)}{S} \quad (6-2)$$

where C (in mg/m^3) is the incremental increase in particle concentration caused by a source S (in kg/d) to which N persons are exposed, each with an average breathing rate BR (in $\text{m}^3/\text{d}\cdot\text{person}$). Note that in a regionalized approach, the terms BR and C can be dependent on location and be specific to each subgroup of persons N , thus allowing the intake fraction to be a metric adaptable to specific locations.

Intake fraction versus uptake fraction

For substances with uniform size distributions, the damage (the burden of a given mass of pollutant emitted) is directly proportional to the emission rate and the intake fraction. But the use of intake fraction is problematic in at least four cases: (i) for air pollutants with varying size distributions that can be deposited in different regions of the respiratory tract depending on physical or chemical properties (e.g., smaller particles may deposit deeper in the lungs); (ii) for ingested pollutants that are absorbed to a different extent in the gastrointestinal tract based on their size and composition; (iii) when pollutant damage studies are based on dose delivered to an organ rather than exposure-response; and (iv) for dermal exposure. Even in cases where the intake fraction is size dependent, it fails to be useful unless size dependent damages are also known. In the case of PM this depends on where the dose of inhaled pollutant is delivered within the respiratory tract. To address the four cases above, I explore the uptake fraction as an alternative metric for exposure or dose. When categorizing pollutants as a function of their potential burden on human health, the uptake fraction reduces the risk of misclassification (i.e., classifying population groups in a class of exposure that they should not be classified into).

Uptake fraction

The uptake fraction is the fraction of emitted pollutant that, after being inhaled or ingested, comes into contact with a part of the body that results in an adverse health impact. For dermal exposure, the uptake fraction simply quantifies the fraction of emitted pollutants that is transferred into the surface of the skin. This dissertation focuses on inhalation uptake fraction. A detailed computation of ingestion or dermal uptake fraction is outside the scope of this dissertation and would require further exposure analysis.

For the specific case of PM, the uptake fraction characterizes the amount of pollutant that is actually deposited in specific regions of the respiratory tract, which is necessary to induce almost any negative health impact. Figure 6-1 shows how the intake fraction and the uptake fraction are evaluated and how they differ from each other. The respiratory tract can be divided into the nasopharyngeal, tracheobronchial, and alveolar zones (Figure 6-1). Inhaled particles either deposit in one of these three zones or are exhaled.

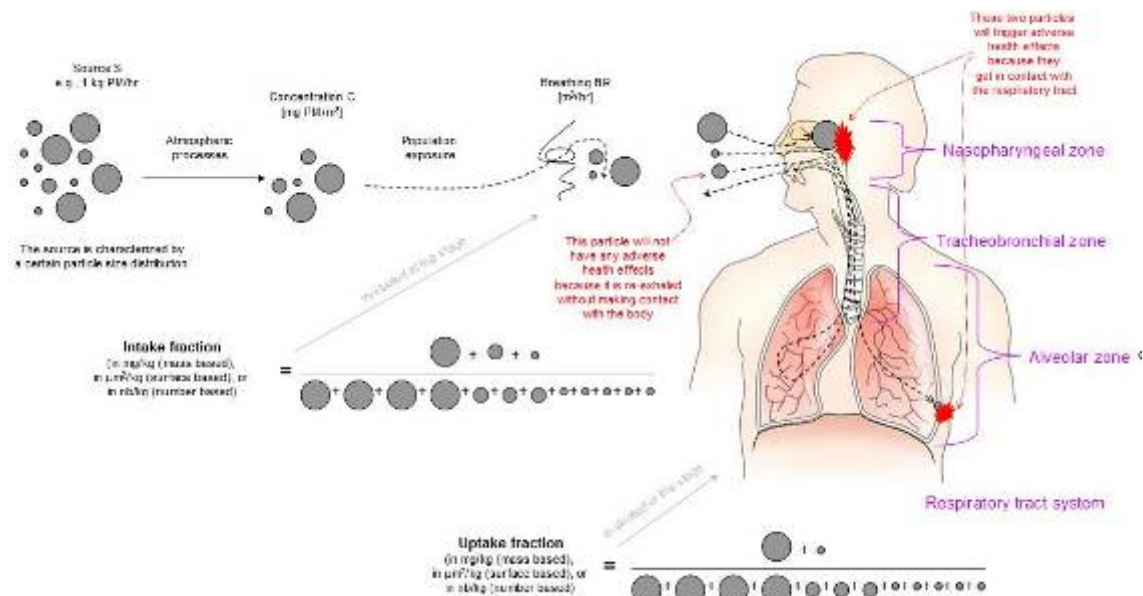


Figure 6-1: Summary of the intake fraction versus uptake fraction concepts for PM (respiratory system image taken from <http://www.edupics.com>).

I suggest using the term “uptake” to express the deposition in the respiratory tract to avoid confusion with the common term “deposition fraction,” which represents the fraction of particle that is deposited on a surface through dry and wet atmospheric deposition processes. Within the respiratory tract, I use “fraction deposited in the respiratory tract” and “fraction taken up in the respiratory tract” interchangeably. Furthermore, considering that when expressed by surface or by number per mass emitted, the term fraction is actually not appropriate because a fraction should not be dimensional. However, in a desire not to change terms throughout the analysis as well as for similarities with the well-known term “intake fraction” I decided to keep the term “uptake fraction” for dimensional results. This is a semantic limitation and could require further analysis, such as using the term “uptake factor”.

Effect factor

In health assessment protocols, the damage can be represented by the product of the emission rate, the effect factor and the intake fraction. The same concept applies to the uptake fraction: the damage can be represented by the product of the emission rate, the effect factor and the uptake fraction. The effect factor (Crettaz et al. 2002, Pennington et al. 2002) is defined as the marginal damage to human health caused by a marginal rate of additional disease per additional unit mass intake. The effect factor is derived from exposure-response or dose-response curves and can depend on where and how much the pollutant deposits in the body. Note that when using the uptake fraction metric, the effect factor concept has to be adapted (this issue will be addressed in Chapter 8).

6.4. Modeling the uptake fraction

Uptake fraction for a single chemical (or a single particle size)

For a single chemical (i.e., with a unique size), the uptake fraction (uF) can be expressed as follows:

$$uF = \frac{\sum_{\text{people,time}} \text{uptake of pollutant by an individual (mass or surface or number)}}{\text{mass released into the environment (mass or surface or number)}} = f_{\text{dep,tot}} \times iF \quad (6-3),$$

where, for the case of inhalation, $f_{\text{dep,tot}} = f_{\text{dep,N}} + f_{\text{dep,T}} + f_{\text{dep,A}}$ is the sum of the deposition fractions in the nasopharyngeal, tracheobronchial, and alveolar parts of the respiratory tract, respectively. Note that the maximum value of $f_{\text{dep,tot}}$ is 1, when the entire amount inhaled is taken up and none is exhaled.

Uptake fraction for a group of chemicals assuming a particle size distribution (or different particle size classes, such as in PM)

For particles of non-uniform diameter D_p , Equation 6-3 can be expressed as follows:

$$uF_{\text{tot}} = \sum_z \left(\int_{D_p} [f_{\text{dep},z}(D_p) \times iF(D_p)] dD_p \right) \quad (6-4)$$

where z can be one of the three zones in the respiratory tract. When explicitly using the mass emitted and inhaled rather than the intake fraction, the uptake fraction can be expressed as follows:

$$uF_{\text{tot}} = \frac{1}{S_{PM}} \times \sum_z \left(\int_{D_p} [f_{\text{dep},z}(D_p) \times M_{\text{inh}}(D_p)] dD_p \right) \quad (6-5)$$

where $iF(D_p)$ (in $\text{kg}_{\text{inhaled}}/\text{kg}_{\text{emitted}}$) has been expressed as the ratio of the intake of particle of size D_p ($M_{\text{inh}}(D_p)$ in kg_{in}) to total mass of particles emitted (S_{PM} in $\text{kg}_{\text{emit}}/\text{d}$).

Figure 6-2 shows the deposition fraction in each respiratory tract zone as a function of particle diameter (D_p) (Yeh et al. 1996) (see section 6.9 for the numerical values).

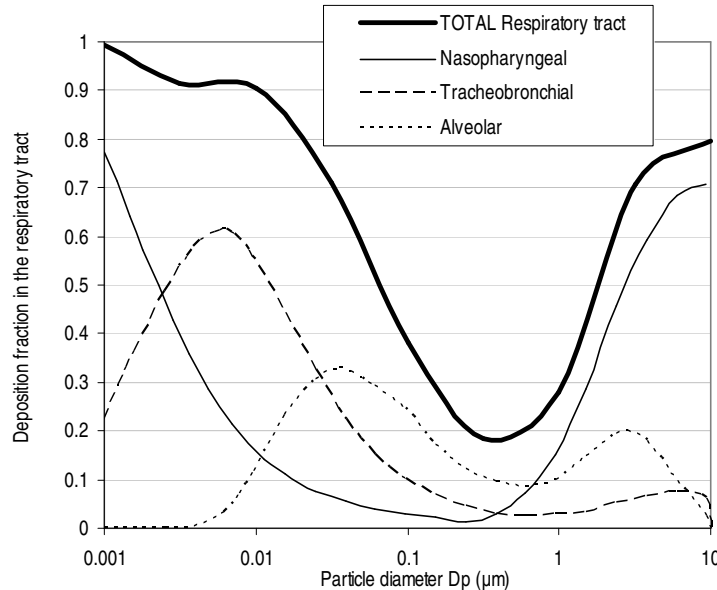


Figure 6-2: Deposition fraction in the respiratory tract (based on Yeh et al. (1996)).

The respiratory tract can be divided into more zones if needed, but the nasopharyngeal, tracheobronchial and alveolar zones used here are the most commonly used.

The units of intake fraction and uptake fraction

The intake fraction is traditionally expressed as the ratio of mass intake to mass emitted into the environment. Using the same approach, the uptake fraction can be expressed as the ratio of mass uptake by the body – or deposited in the respiratory tract when inhaled - to mass emitted into the environment. Furthermore, for particles, both metrics can be expressed using particle surface or number of particles instead of mass (in case these metrics better fit dose-response curves). The alternatives for expressing the intake fractions and uptake fractions are listed in Table 6-1. The common approach for intake fraction and the default approach I suggest for uptake fraction are highlighted in grey, accounting only for particle mass. However if particle surface area is a better measure of toxicity than the mass (or particle number) (Froines 2006, Oberdörster et al. 2005), then using surface area instead of mass appears more useful, at least as an intermediate calculation step.

Table 6-1: Different units to express intake fractions and uptake fractions depending on relevant unit for damage.

				Unit used for the emission		
				per mass emitted	per surface emitted	per number emitted
Metric	intake fraction (or intake factor for dimensional values)	unit used to express the amount taken in (inhaled for inhalation) or taken up (deposited in the respiratory tract for inhalation)	per mass	$\frac{\text{mass intake}}{\text{mass emitted}}$	$\frac{\text{mass intake}}{\text{surface emitted}}$	$\frac{\text{mass intake}}{\text{number emitted}}$
			per surface	$\frac{\text{surface intake}}{\text{mass emitted}}$	$\frac{\text{surface intake}}{\text{surface emitted}}$	$\frac{\text{surface intake}}{\text{number emitted}}$
			per number	$\frac{\text{number intake}}{\text{mass emitted}}$	$\frac{\text{number intake}}{\text{surface emitted}}$	$\frac{\text{number intake}}{\text{number emitted}}$
	uptake fraction (or uptake factor for dimensional values)		per mass	$\frac{\text{mass deposited}}{\text{mass emitted}}$	$\frac{\text{mass deposited}}{\text{surface emitted}}$	$\frac{\text{mass deposited}}{\text{number emitted}}$
			per surface	$\frac{\text{surface deposited}}{\text{mass emitted}}$	$\frac{\text{surface deposited}}{\text{surface emitted}}$	$\frac{\text{surface deposited}}{\text{number emitted}}$
			per number	$\frac{\text{number deposited}}{\text{mass emitted}}$	$\frac{\text{number deposited}}{\text{surface emitted}}$	$\frac{\text{number deposited}}{\text{number emitted}}$

6.5. Example of application for PM

An example application of the uptake fraction concept to PM is presented here.

Assume a PM₁₀ emission *S* of 1 kg/s causes an increase in ambient concentration of 1 µg primary PM₁₀ per m³. If 100 million people are exposed with a breathing rate of 13 m³/(pers·d) (USEPA 1997), then the resulting intake fraction is 1.5×10⁻⁵ kg_{inh}/kg_{emi} or 15 ppm (i.e., 15 mg of PM₁₀ are inhaled by a population with each kilogram of PM₁₀ emitted in the air). The computation of the intake fraction is the following: intake fraction = (100,000,000 pers × 13 m³/(pers·d) × 1 µg/m³) / (24 hr/d × 3,600 s/h × 1,000,000,000 µg/s) = 1.5×10⁻⁵ kg_{inh}/kg_{emi}.

To show how the uptake fraction metric can change compared to the intake fraction metric, I examine three different particle size distributions (Figure 6-3). The emission can result in exposure to particle size distributions dominated by (1) coarse PM (1-10 µm); (2) fine PM (0.1-1 µm); and (3) ultrafine PM (0.01-0.1 µm).

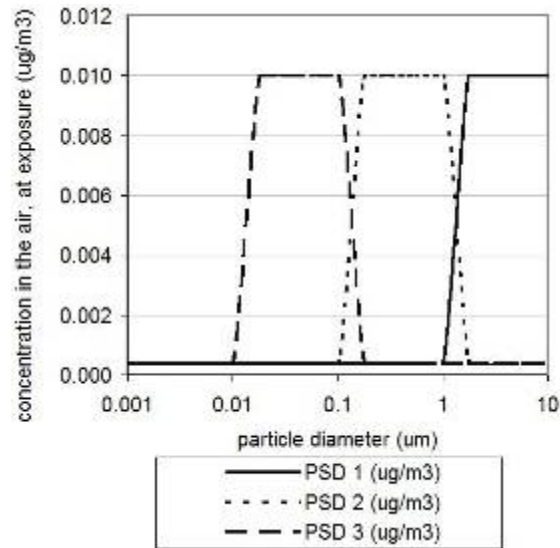


Figure 6-3: Three particle size distributions (PSDs) of particles at time of exposure. “ug” means “micrograms.”

The uptake fraction for each particle size distribution can be found by multiplying the particle size distribution curves suggested in Figure 6-3 by the respiratory tract deposition fraction curves suggested in Figure 6-2. At each particle diameter, the concentration in the air (Figure 6-3) is multiplied by the intake fraction ($1.5 \cdot 10^{-5}$). Then this concentration that enters the respiratory tract is multiplied by the respiratory tract deposition fraction curve for each of the zones (nasopharyngeal, tracheobronchial and alveolar, Figure 6-2). Summing the result of each zone gives the mass (e.g., in μg) of particle that deposit in the respective zone, per m^3 depicted in Table 6-2.

Table 6-2: Uptake fractions (uF) for the different particle size distributions (PSD) and different zones of the respiratory tract.

	uF nasopharyngeal	uF tracheobronchial	uF alveolar	uF tot
PSD 1	7.9E-06 (71%)	1.3E-06 (11%)	2.0E-06 (18%)	1.1E-05 (100%)
PSD 2	1.2E-06 (30%)	9.4E-07 (25%)	1.7E-06 (45%)	3.8E-06 (100%)
PSD 3	1.3E-06 (15%)	3.7E-06 (41%)	4.0E-06 (45%)	9.0E-06 (100%)

The uptake fractions found are depicted in Figure 6-4.

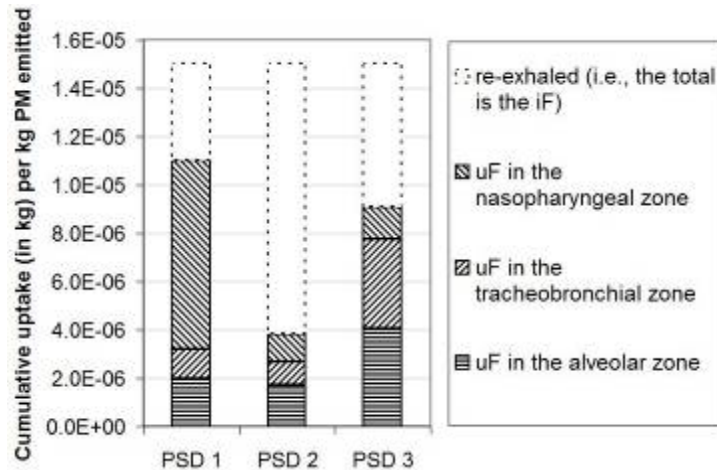


Figure 6-4: Cumulative uptake of PM in the different zones of the respiratory tract for exposure to three particle size distributions (shown in Figure 6-3).

The intake fraction is the same for each of the three cases ($1.5 \times 10^{-5} \text{ kg}_{\text{inh}}/\text{kg}_{\text{emi}}$), therefore using the intake fraction as the only metric for exposure or dose would result in the same calculated damage to human health for each of the three cases. However, the uptake fraction shows the differences in respiratory exposure among the three cases, for both the total amount of particles deposited in the respiratory tract and the distribution of deposition within the respiratory tract. That difference shows that total damage to human health is likely to differ among the three cases. The total uptake fraction is highest for a particle size distribution dominated by coarse particles (around $3 \mu\text{m}$), followed by a particle size distribution dominated by ultrafine particles (around $0.03 \mu\text{m}$); the smallest uptake fraction is dominated by medium-range particles (around $0.3 \mu\text{m}$) at exposure. Regarding the location of deposition within the respiratory tract, PM dominated by coarse particles (around $3 \mu\text{m}$) deposits mainly in the nasopharyngeal zone, while PM dominated by medium-range particles (around $0.3 \mu\text{m}$) deposits in the three zones, with some tendency towards the alveolar zone. PM dominated by ultrafine particles (around $0.03 \mu\text{m}$) deposits mainly in the tracheobronchial and alveolar zones.

For the three cases of PM exposure described above, all yield the same intake fraction, but the uptake fraction and deposition location can vary by up to a factor of three. Thus the increased information of the uptake fraction can lead to improved estimates of the damage to human health.

6.6. Outlook

For PM, different removal mechanisms occur in different zones within the respiratory tract. Studies suggest that the magnitude of harm is directly related to where the particle deposits in the lung, in which case the uptake fraction calculations above show that the damage to human health will differ with particle size distribution and therefore will not necessarily be proportional to the intake because of the heterogeneity of the particle size distribution. With accurate measurements of the PM particle size distribution in the region of exposure where concentration-response curves are derived, it is possible to determine particle size distribution-specific effect factors, which can be multiplied by particle size distribution-specific uptake fractions to better estimate human health damage.

Future investigation should identify how mechanisms involved in the uptake by the body differ depending on the organs, and especially depending on the zone of deposition in the respiratory tract. Establishing mechanism-based rather than concentration-response curve effect factors would help to better capture the adverse health effects caused by pollutants, especially when linked with the zonal deposition uptake fraction model presented here.

In conclusion, the uptake fraction metric can be used in addition to the intake fraction to improve the assessment of pollutants by considering how uptake in organs is influenced by pollutant characteristics. Using the uptake fraction is especially important for the case of PM where particle size distribution influences deposition in the respiratory tract system by up to a factor of three.

Proposal of the uptake fraction metric follows from the original concept of “the dose makes the poison,” often attributed to the medieval physician Paracelsus. This concept was recently updated by Smith (2002) as “the place makes the poison” to indicate that the location of a chemical emission governs its toxicity. The uptake fraction metric extends the concept to “the depth makes the poison,” to reflect the importance of lung penetration.

6.7. Acknowledgments

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6.9. Supporting information for this chapter

Table 6-3 presents the respiratory tract deposition functions (Yeh et al. 1996).

Table 6-3: Respiratory tract deposition functions (Yeh et al. 1996).

Nasopharyngeal (Fd,N,dp)		Tracheobronchial (Fa,T,dp)		Alveolar (Fd,A,dp)		TOTAL Respiratory tract (Fd,tot)	
Diameter (µm)	Deposition fraction	Diameter (µm)	Deposition fraction	Diameter (µm)	Deposition fraction	Diameter (µm)	Deposition fraction
0.0010	0.771	0.0010	0.223	0.0010	0.000	0.0010	0.99
0.0012	0.714	0.0016	0.364	0.0025	0.000	0.0032	0.91
0.0015	0.643	0.0025	0.486	0.0039	0.003	0.0100	0.91
0.0018	0.571	0.0039	0.579	0.0062	0.033	0.0316	0.71
0.0023	0.500	0.0057	0.611	0.0078	0.071	0.1000	0.38
0.0028	0.429	0.0068	0.607	0.0098	0.120	0.3162	0.18
0.0036	0.357	0.0090	0.571	0.0155	0.234	1.0000	0.28
0.0049	0.286	0.0155	0.443	0.0246	0.307	3.1623	0.71
0.0070	0.214	0.0246	0.326	0.0354	0.329	10.0000	0.80
0.0115	0.143	0.0388	0.223	0.0425	0.321		
0.0155	0.111	0.0613	0.150	0.0613	0.293		
0.0246	0.077	0.0968	0.103	0.0968	0.247		
0.0388	0.056	0.1530	0.070	0.1530	0.171		
0.0613	0.040	0.2417	0.046	0.2417	0.123		
0.0968	0.030	0.3819	0.029	0.3819	0.094		
0.1530	0.021	0.6033	0.026	0.6033	0.086		
0.2417	0.014	0.9531	0.029	0.9531	0.100		
0.3819	0.029	1.5058	0.033	1.5058	0.147		
0.6033	0.071	2.3788	0.051	2.3788	0.196		
0.9531	0.150	3.7582	0.067	2.8563	0.200		
1.5058	0.286	5.9372	0.076	3.7582	0.180		
2.3788	0.457	9.3798	0.063	5.9372	0.097		
3.7582	0.593	10.0000	0.000	9.3798	0.026		
5.9372	0.684			10.0000	0.000		
9.3798	0.707						
10.0000							

7. A method for estimating size-dependent uptake fractions of primary particulate matter in the respiratory tract

Building on the concept developed in Chapter 6, a model, based on the same general concepts as the IMPACT North America model developed in Chapter 2, is developed to calculate uptake fractions (uF) for particulate matter (PM) for different fate and exposure conditions.

The identification and analysis of the most influencing parameters when evaluating the fate and exposure of PM as function of the particle size distribution is outside of the scope of this dissertation. Indeed the focus of this dissertation can be formulated “what exists in the current scientific knowledge that can be used to improve geographic differentiation of characterization factors for PM.” The phenomena considered for modeling the fate, exposure and effect of PM emissions are the following:

- Source-specific particle size distribution (influencing the fate, exposure and effect)
- Differential removal mechanisms from the atmosphere under dry and wet conditions (influencing the fate)
- Influence of the variation in population density patterns (influencing the exposure)
- Differential deposition in the respiratory tract (influencing the exposure and the effect)

Further work can be done to update the model presented in this chapter in order to incorporate additional parameters that are currently neglected in the present model.

7.1. Summary

PM is a significant cause of adverse human health effects. Studies suggest that an evaluation of PM-caused damage to human health should integrate surface area and number of particles in addition to overall mass exposure. However, current impact assessment methods consider little if any influence of particle size distribution in fate, exposure or effect analyses. This chapter uses PM particle size distribution to evaluate the uptake fraction, which expresses the ratio of uptake in the respiratory tract (i.e., the deposition in the respiratory tract) to emission rate (in terms of mass and surface area of particles), and shows how this metric can be used as an extension of the intake fraction to improve the assessment of damage caused by PM exposure. The modeling used in this chapter to characterize uptake fraction considers the influence of source-specific particle size distributions, differential removal mechanisms under dry and wet conditions, population density patterns, and differential deposition in the respiratory tract. In addition to the size, the modeling of composition of PM is also important to study but is outside of the scope of this chapter. As an example, the uptake fractions for tailpipe PM_{2.5} and PM_{10-2.5} emitted by diesel engines' private car are determined for different weather conditions and population densities. Results show that, within PM₁₀, exposure expressed as uptake fraction and evaluated in terms of mass or surface area is dominated by particles between 0.01 and 3 μm. When the uptake fraction is evaluated by mass, compared to particles smaller than 2.5 μm, exposure to particles greater than 2.5 μm is not negligible, but if evaluated by surface area of particles, exposure to particles greater than 2.5 μm can be considered negligible. For ambient PM concentrations proposed by Seinfeld and Pandis (1998), as well as for diesel emissions from Norbeck et al. (1998), the ratio of uptake fraction to intake fraction for PM_{2.5} is 0.3. The uptake

fraction can improve fate and exposure analysis and can therefore be used to reevaluate damage factors when estimating damage to human health caused by PM exposure.

7.2. Introduction

Though several studies show that among well-studied pollutant emissions PM is associated with some of the most serious adverse health effects (Dockery et al. 1993, Schwartz et al. 2008, Pope et al. 2009), at present, the life-cycle impact assessment methods that include PM impacts fail to capture the latest findings on PM exposure-disease relationships described in Chapters 4 and 6.

Uptake fraction as an extension of intake fraction

The uptake fraction metric — the product of the intake fraction and the deposition fraction (the fraction of emissions that is deposited in the respiratory tract⁵) — developed in Chapter 6 can be used to improve the evaluation of fate and exposure of PM.

Objective

The objective of this chapter is to evaluate the size-dependent uptake fractions for primary PM based on mass and surface area of particles, taking into account (i) the influence of the source's specific particle size distribution functions, (ii) the influence of the differential removal from the atmosphere under dry and wet weather conditions, (iii) the influence of population density patterns relative to the source, and (iv) the influence of the differential deposition functions in the respiratory tract.

The uptake fraction is calculated here considering PM as spheres. Because of the broadness of the subject, the influence of the different geometry and the influence of the composition of PM in the evaluation of the uptake fraction are both outside the scope of this chapter and require further investigation.

Finally, particle number is not evaluated in the present dissertation. Indeed, the modeling of particle number is more complex than particle surface area because of phenomenon such as coagulation that can significantly influence ultrafine particle number. In addition, as showed by Keywood et al. (1999), particle number may not be correlated with particle mass and therefore, results obtained on a mass basis but using particle number in the modeling will not be valid in a life-cycle assessment context.

7.3. Method

Framework

Extending upon the concept of the intake fraction, the uptake fraction (uF) is defined in Chapter 6 according to Equation 7-1.

$$uF = \frac{\sum_{\text{people, time}} \text{deposition of pollutant in an individual (mass or surface or number)}}{\text{amount released into the environment (mass or surface or number)}} \quad (7-1)$$

Figure 7-1 depicts the framework used to evaluate the damage to human health from PM, modeling successively (as described below) (i) atmospheric fate relating emissions to air masses or concentrations, (ii) population exposure to calculate intake, and (iii) respiratory tract deposition to estimate PM uptake. Special care is given to phenomena that affect or are affected by particle size distribution, such as distinguishing the zone of uptake within the respiratory tract.

⁵ In this chapter, “deposited in the respiratory tract” and “taken up by the respiratory tract” are considered the same.

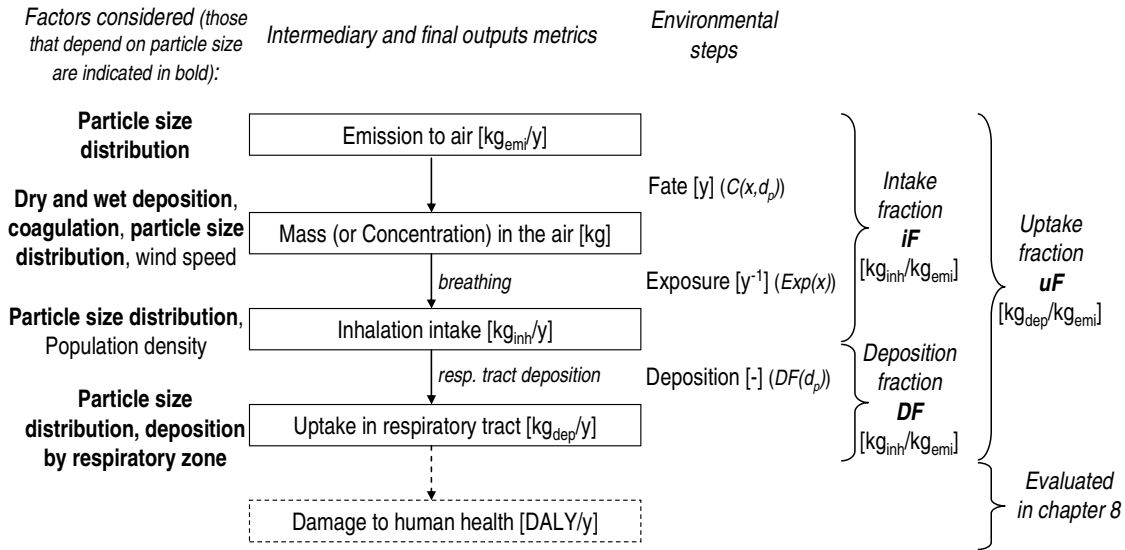


Figure 7-1: Framework to evaluate the uptake fraction.

Environmental modeling

Fate

Note that PM is considered with a density of 1 g/cm³.

a) Emission and initial concentration

I estimate fate assuming a steady-state box model. The initial atmospheric concentration at the point of emission ($x=0$) is a function of the distribution of particle diameters (d_p) in the emission, $C(0, d_p)$, evaluated according to Equation 7-2, where $S(d_p)$ is the constant emission rate of particle d_p , H is the mixing height of the atmosphere (set by default to 1,000 m – Rosenbaum et al. 2008), W is the width chosen for the modeling (set by default to 1,000 m – note that this value is arbitrary and does not influence the uptake fraction because the model considers the air flow as parallel, as explained in detail below), and U is the wind velocity (set by default to 4 m/s).

$$C(0, d_p) = \frac{S(d_p)}{H \times W \times U} \quad (7-2)$$

The fate determines the net concentration $C(x, d_p)$ of particle of size d_p at a distance x downwind from the emission source. The two physical phenomena influencing the concentration considered are dry deposition and wet deposition. Coagulation also influences the concentration but is excluded from this analysis. Further research is needed to address the complex phenomenon of coagulation.

$$C(x, d_p) = C(x - dx, d_p) \times [1 - f_{dep, dry} - f_{dep, wet}] \quad (7-3)$$

where $f_{dep, dry}$ is the fraction of particles d_p that deposit over dx through dry deposition and $f_{dep, wet}$ is the fraction that deposits over dx through wet deposition. The two deposition phenomena as well as the different parameters are described in detail below.

b) Dry deposition

The concentration of particle d_p at a distance x downwind from the source influenced by dry deposition only, $C(x, d_p)_{dry}$, is calculated according to Equation 7-4 using the approach

developed by Nazaroff (2006). The fraction of particles d_p that deposits over dx through dry deposition is given by:

$$f_{dep,dry} = \frac{dC(x, d_p)_{dry}}{C(x - dx, d_p)_{dry}} = 1 - \frac{C(x, d_p)_{dry}}{C(x - dx, d_p)_{dry}} = 1 - \exp\left(-\frac{v_d(d_p) \times dx}{H \times U}\right) \quad (7-4)$$

where $v_d(d_p)$ is the dry deposition velocity for the particle of size d_p (evaluated, by default, as the average of the values provided by Nazaroff (2006) and Seinfeld and Pandis (1998, p. 970)) (Figure 7-2).

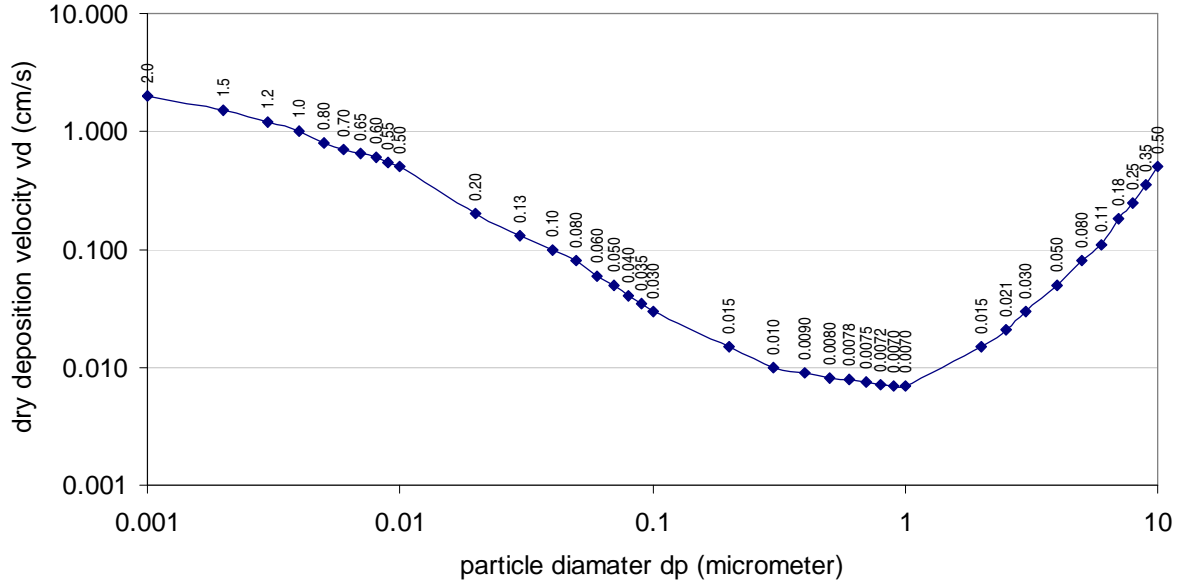


Figure 7-2: Dry deposition velocity v_d (in cm/s) as a function of the particle size d_p (average of the values provided by Nazaroff (2006) and Seinfeld and Pandis (1998, p. 970))

c) Wet deposition

The reduction in concentration that will occur between $x-dx$ and x is $dC(dx, d_p)_{wet}$. It is evaluated assuming that $dC/dt = A \cdot C$ (Seinfeld and Pandis 1998), where A (in s^{-1}) is the wet-precipitation scavenging coefficient, and that $dC(dx, d_p) = (dC/dt) \cdot (dx/U)$, where dx/U is the residence time of the particles within dx . Thus the fraction that deposit over distance dx through wet deposition is given by:

$$f_{dep,wet} = \frac{dC(x, d_p)_{wet}}{C(x - dx, d_p)_{wet}} = 1 - \frac{C(x, d_p)_{wet}}{C(x - dx, d_p)_{wet}} = \frac{C(x - dx, d_p)_{wet} - C(x, d_p)_{wet}}{C(x - dx, d_p)_{wet}} = \Lambda \times (dx/U) \times F_{rain} \quad (7-5)$$

where F_{rain} is the fraction of the time that it is raining. Assuming a rainfall event with an intensity of 1.3 mm/h and an average annual precipitation of 800 mm/y (Jolliet and Hauschild 2005), one gets an average of 600 hours of rain per year, or 7% of the time with rain. Using the formula provided by Seinfeld and Pandis (1998) for precipitation scavenging of particles, and considering a rainfall event that is monodispersed, one finds that:

$$\Lambda = \frac{\pi}{4} \times d_r^2 \times v_t \times E(d_p, d_r) \times N(d_r), \quad (7-6)$$

where d_r is the diameter of a raindrop (set by default to 1 mm — a simplification that would require further analysis), v_t is the vertical velocity of the falling raindrops (set by default to 4 m/s — a simplification that would require further analysis), $E(d_p, d_r)$ is the collision efficiency (Seinfeld and Pandis 1998), and $N(d_r)$ is the concentration of raindrops (set by default to 173 drops/m³ when representing a rainfall event that has an intensity of 1.3 mm/h with the v_t and d_r mentioned above). Note that if v_t is used to calculate $N(d_r)$, A becomes independent of v_t , since A is both multiplied and divided by v_t .

Note that assuming a monodisperse rainfall with droplet diameter of 1 mm can underestimate the effects of precipitation scavenging. Indeed, first, smaller raindrops are present and are more efficient than larger droplets in removing particles. Second, the scavenging model in Equation 7-6 may underestimate what is observed in the field (a possible explanation could be because of the role of condensational growth of hygroscopic fine PM during rainfall events). (Nazaroff 2009)

Exposure

The exposure $Exp(dx)$ (in m³/s) is evaluated as:

$$Exp(dx) = N_{pers}(x, dx) \times BR \quad (7-7)$$

When Equation 7-7 is multiplied with the concentration $C(x, d_p)$ of particles of size d_p at the location x (in kg/m³) (Equation 7-3), I obtain the intake fraction (iF) evaluated according to Equation 7-14 as the cumulative intake fraction downwind from the source:

$$iF [amount_{inhalated} / kg_{emitted}] = \frac{\sum_{x=0}^x \left[\sum_{d_p \min}^{d_p \max} (Exp(dx) \times C(x, d_p)) \right]}{S_{tot}}, \quad (7-8)$$

where the $N_{pers}(x, dx)$ is the number of persons situated between $x-dx$ and x and BR is the breathing rate (13.3 m³/pers·d, the average between 11.3 for female and 15.2 for male – USEPA, 1997). $N_{pers}(x, dx)$ can be expressed as $PD(x) \cdot W \cdot dx$ where $PD(x)$ is the population density at the location x (in pers/m²) and W is the width of the model. Note that the value of W is arbitrary and does not influence the uptake fraction because the model considers the air flow to be parallel. Indeed, Equation 7-8 is independent from W since Equation 7-8 is both multiplied by W , in the term $Exp(dx)$, and divided by W , in the term $C(x, d_p)$. In the present evaluation, a default distance of 1,000 km (representing approximately 3 days at a wind velocity of 4 m/s) has been chosen to represent average conditions. After 1,000 km, it is estimated that, in average geographical conditions, either the ocean or the desert will limit exposure and the successive uptake of PM. This value can be over- or underestimated for specific situations and can be adjusted to better represent specific conditions.

Deposition and uptake

The uptake fraction is the product of the intake fraction (Equation 7-8) and the deposition fraction in the respiratory tract. The uptake fraction (uF) is evaluated according to Equation 7-9 as the cumulative uptake fraction downwind from the source up to a distance x , that is taken to be 1,000 km in the present analysis.

$$uF [amount_{deposited_in_the_resp_tract} / kg_{emitted}] = \frac{\sum_{x=0}^x \left[\sum_{d_p \min}^{d_p \max} (ConvFct(d_p) \times DF(d_p) \times Exp(x) \times C(x, d_p)) \right]}{S_{tot}} \quad (7-9)$$

where $S_{tot} = \sum S(d_p)$ represents the total emission of the particles of interest (in kg/s). $DF(d_p)$ is the fraction of the particles of size d_p inhaled that deposits in the respiratory tract (see Figure 6-2 in section 6.4). The concentration $C(x, d_p)$ is always computed in mass per unit volume. The metric conversion factor $ConvFct(d_p)$ allows one to evaluate the final uptake fraction in terms of two alternative metrics, namely as mass and surface area of particle per unit volume, respectively. For units of mass deposited per mass emitted, $ConvFct(d_p)$ is simply 1 kg/kg; and for surface deposited per mass emitted, $ConvFct(d_p)$ is equal to $S(d_p)$ in m^2/kg — the surface of particles d_p per mass of particle d_p .

This approach shows that for a specific case of PM, the particle size distribution and the deposition mechanisms in the respiratory tract are taken into account when determining the PM uptake fraction. The particle size distribution is captured in the relationship by the two terms $S(d_p)$ and S_{tot} . The deposition mechanisms in the respiratory tract are also functions of d_p which is captured in the term $DF(d_p)$. Equation 7-9 assumes a steady-state deposition in the respiratory tract with a steady-state emission in the environment.

The UFPM tool

The framework with the different phenomena described above is combined with the matrix approach in a tool named “UFPM” (version 1.0) developed in MS Excel. This tool is used to evaluate the uptake fraction in the different zones of the respiratory tract for different particle size distributions, under different weather conditions and with different population density patterns. The UFPM tool can be downloaded from <http://www.impactmodeling.org> or by contacting sebastien.humbert@cal.berkeley.edu. Figure 7-3 presents a schematic for the modeling of the uptake fraction.

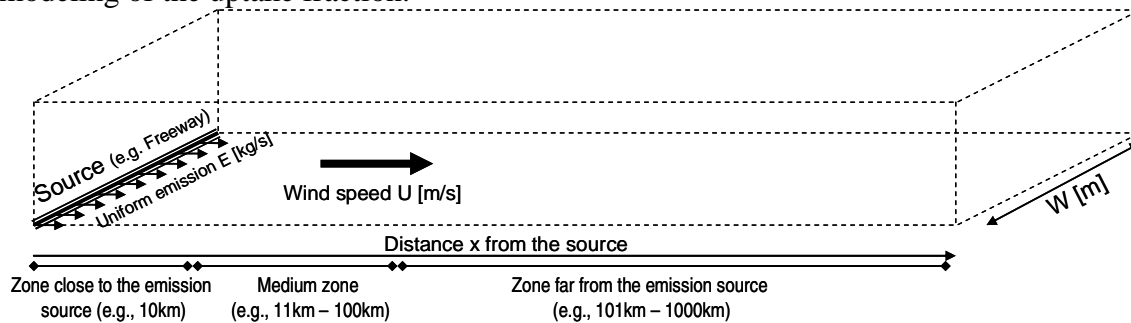


Figure 7-3: Schematic to model the uptake fraction.

7.4. Diesel engine case study

A case study is performed to determine the uptake fraction for tailpipe emissions of $PM_{2.5}$, $PM_{10-2.5}$ and PM_{10} from diesel engines’ private car for different weather conditions and different population densities. Since measurements, reported data, and life cycle inventories are typically expressed using $PM_{2.5}$, $PM_{10-2.5}$ and PM_{10} as boundaries, the present evaluation expresses uptake fraction for $PM_{2.5}$, $PM_{10-2.5}$ and PM_{10} to easily compare and combine our results with other findings and data. However, if needed, the tool provides results for any size range.

Fate

a) Emissions scenario and initial concentrations

Tailpipe emissions from diesel engines are a substantial source of primary $PM_{2.5}$ and $PM_{10-2.5}$. The particle size distribution of emissions is considered after exhaust control systems, but without diesel particle filters. Particle size distributions specific to the different sources are

difficult to estimate (Chang et al. 2004). The particle size distribution for diesel engine emissions used in this chapter to generate the uptake fraction specific to a source is based on Norbeck et al. (1998) and is presented in Figure 7-4.

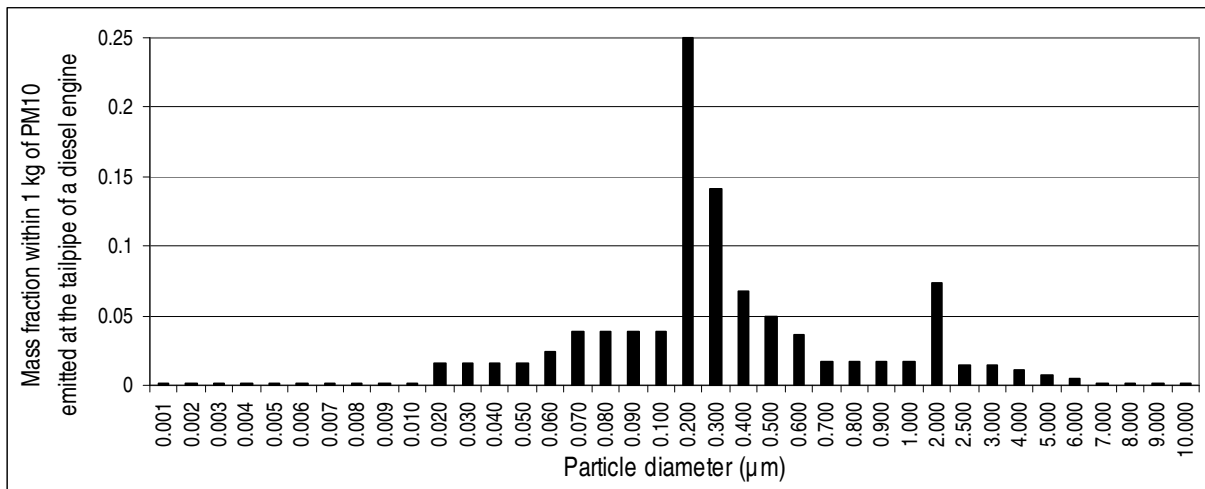


Figure 7-4: Typical particle size distribution for diesel engine emissions, represented for discrete particle sizes (based on Norbeck et al. 1998).

Note that the variability of particle size distribution within the same source can be large. In general, the diameter refers to the aerodynamic diameter. However, in the literature some particle size distributions have been reported as functions of the electrical mobility diameters, which I here consider equivalent to the aerodynamic diameter. The slight uncertainty that may arise from this simplifying assumption is believed to not considerably alter the final results (especially when compared to the variability among the different particle size distribution measurements of same types of sources). Further research may be needed to confirm this assumption.

b) Weather scenarios

The three weather scenarios considered are:

- “dry” season, with the sole removal mechanism from the atmosphere being dry deposition,
- “wet” season, with removal mechanisms from the atmosphere being dry and wet deposition, and
- “mixed” season with intermittent rain, being a mix of dry and wet weather scenarios. This scenario is also used as a proxy for a situation where the weather conditions are unknown. The mixed scenario assumes that wet deposition occurs 7% of the time.

Exposure: population density scenarios

The population density pattern scenarios considered are:

- “high density”: with downtown, suburbs, and countryside population density patterns in the zones close to, a medium distance from, and far from the emission,
- “medium density”: considering the entire area to be countryside, and
- “low density”: considering the entire area to be remote.

“Downtown,” “suburbs,” “countryside” and “remote” population density patterns are assumed to be, respectively, 5,000, 500, 50 and 5 persons/km².

7.5. Results

Fate of particles in the air

The spatial evolution of the mass present in the air at different distances x from the source (expressed as $C(x, d_p)/C(0, d_p)$), for each diameter d_p , are presented in Figure 7-5. Figure 7-5 represents the mass of particles that are at a size d_p (the fraction of the initial amount of particle d_p emitted in the air), as a function of the distance downwind from the emission source. It shows that after a few kilometers, very small and very large particles are no longer observed in significant quantities. After 1,000 km, no particles smaller than $0.01 \mu\text{m}$ or greater than $3 \mu\text{m}$ are observed in the air. However, for particles between 0.1 and $1 \mu\text{m}$, nearly the same amount is observed in the air 1,000 km away and beyond as observed near the source. Therefore, as long as there are people downwind from a source, the uptake fraction will be cumulative, as these observations indicate that the accumulation mode (approximately 0.1 to $1 \mu\text{m}$) continues to contribute to the uptake fraction even after particles travel for thousands of kilometers.

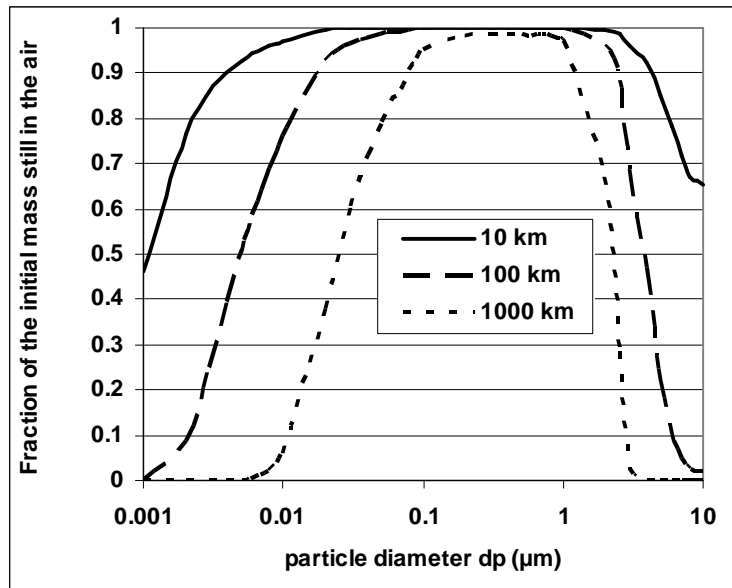


Figure 7-5: Evolution of the “observed” mass in the air of particles of interest, as a function of the distance downwind from emission for each diameters d_p , for “mixed” weather conditions.

Uptake fraction of diesel emissions

Figure 7-6 represents the evolution of the cumulative uptake fraction of diesel emissions, for emissions in medium population density patterns, and for mixed weather conditions. The uptake fraction is evaluated respectively for mass and surface area of particles deposited per kg emitted.

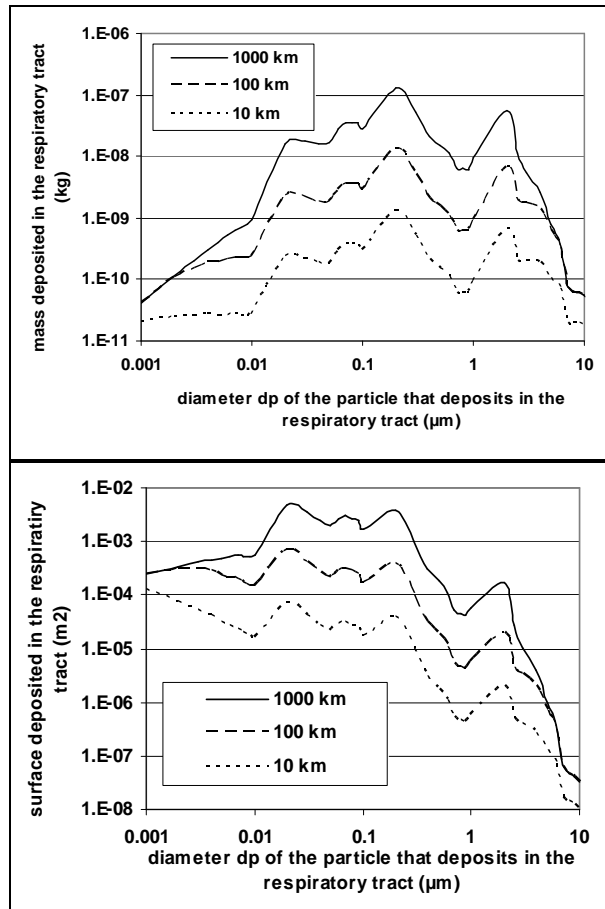


Figure 7-6: Evolution of the cumulative uptake fraction of diesel emissions, for emissions in medium population density patterns, and for “mixed” weather conditions.

Figure 7-6 shows that the mass deposited in the respiratory tract tends to be dominated by particles between 0.01 and 3 μm (note that the decrease in uptake around 1 μm is partly explained by a smaller emission of particles around 1 μm than around 2 μm). The total surface area of particles deposited in the respiratory tract is dominated by particles between 0.01 and 0.3 μm . The two metrics show that beyond 100 km, the accumulation mode (between 0.01 and 0.3 μm) keeps on building (which is consistent with what is observed in Figure 7-5), and very small and very large particles do not accumulate anymore in the respiratory tract. For both metrics, if still present, populations farther than 1,000 km downwind need to be considered to properly evaluate the damage to human health caused by particles emissions.

Note that none of the three population scenarios evaluated in this chapter considers an eventual increase in the density of people far away from the source. Thus the population density scenarios considered do not hold perfectly for cases where cities are situated close to each other (e.g., Kansas City, St. Louis, Indianapolis, Cincinnati, and Columbus, with each city within a few hundred kilometers of another). It would be interesting to evaluate how sensitive the uptake fraction is to whether the PM is emitted up- or downwind from a city. To solve each one of these specific cases, a regionalized approach can be used.

The intake fraction and uptake fraction evaluated with the UFPM tool for $\text{PM}_{2.5}$, $\text{PM}_{10-2.5}$, and PM_{10} are presented in Table 7-1. The intake fraction (iF) and uptake fraction (uF) developed

for unknown population density patterns are calculated as follows: $iF_{unknown} = 0.23 \times iF_{high} + 0.40 \times iF_{medium} + 0.37 \times iF_{low}$, and $uF_{unknown} = 0.23 \times uF_{high} + 0.40 \times uF_{medium} + 0.37 \times uF_{low}$. The fractions 0.23, 0.40, and 0.37 represent the respective fractions of PM emissions occurring within high, medium, and low population density regions in the United States (extrapolated from Greco et al. 2007). The proxy used for these three population densities in the United States is based on the counties with more than 160 pers/km², with 16 to 160 pers/km², and below 16 pers/km², respectively. The ratios between high, medium, and low population density regions can be adapted to match specific situations.

Table 7-1: Intake fraction and uptake fractions of PM_{2.5}, PM_{10-2.5} and PM₁₀, expressed using different exposure metrics, for diesel engine emissions, for mixed weather season, and for different population density patterns.

Population density pattern for the location of emission	Intake fraction [kg _{inhaled} /kg _{emitted}]	Naso-pharyngeal zone	Tracheo-bronchial zone	Alveolar zone	Total respiratory tract	
PM _{2.5}	Uptake fraction [kg _{deposited} /kg _{emitted}]					
	high	1.8E-5	18%	25%	57%	5.4E-6
	medium	1.8E-6	18%	26%	56%	5.6E-7
	low	1.8E-7	18%	26%	56%	5.6E-8
	unknown	5.0E-6				1.5E-6
	Uptake fraction [m ² _{deposited} /kg _{emitted}]					
	high		10%	34%	56%	2.6E-1
	medium		14%	39%	47%	3.6E-2
low		13%	39%	49%	3.4E-3	
unknown					7.6E-2	
PM _{10-2.5} , i.e., PM(2.5<dp≤10)	Uptake fraction [kg _{deposited} /kg _{emitted}]					
	high	2.7E-6	68%	8%	25%	2.1E-6
	medium	2.7E-7	68%	8%	25%	2.1E-7
	low	2.7E-8	68%	8%	25%	2.1E-8
	unknown	7.5E-7				5.7E-7
	Uptake fraction [m ² _{deposited} /kg _{emitted}]					
	high		67%	8%	26%	3.6E-3
	medium		67%	8%	26%	3.6E-4
low		67%	8%	26%	3.6E-5	
unknown					9.7E-4	
PM ₁₀	Uptake fraction [kg _{deposited} /kg _{emitted}]					
	high	1.8E-5	19%	10%	45%	5.3E-6
	medium	1.7E-6	19%	14%	61%	5.5E-7
	low	1.8E-7	19%	13%	58%	5.4E-8
	unknown	4.8E-6				1.5E-6
	Uptake fraction [μm ² _{deposited} /kg _{emitted}]					
	high		25%	34%	45%	2.5E-1
	medium		26%	39%	37%	3.4E-2
low		26%	39%	39%	3.2E-3	
unknown					7.3E-2	

7.6. Discussion

Uptake fraction versus intake fraction

The total uptake fractions of PM_{2.5}, PM_{10-2.5}, and PM₁₀ are 30% (i.e., 0.3 kg_{deposited}/kg_{inhaled}), 76% and 31% of the respective intake fractions. The PM_{10-2.5} fraction is trapped in the respiratory tract more easily than PM_{2.5} because the particle size distribution of diesel PM has its peak between 0.1 μm and 1 μm (Figure 7-4), where the deposition in the respiratory tract is the lowest (Figure 6-2). The value for PM₁₀ appears close to the value of PM_{2.5} because in the particle size distribution used in this chapter more than 95% of the PM₁₀ is actually below PM_{2.5} and thus PM₁₀ nearly measures PM_{2.5}. When combining the typical ambient

urban concentration (Seinfeld and Pandis, 1998, p.430) with the total deposition curve in the respiratory tract (Figure 6-2), one also gets a value of approximately $0.3 \text{ kg}_{\text{deposited}}/\text{kg}_{\text{inhaled}}$.

PM_{10-2.5} versus PM_{2.5}

When evaluated per mass deposited, the uptake fractions of PM_{10-2.5} are approximately 25% to 40% of the uptake fractions of PM_{2.5}. However, when evaluated per surface deposited, the uptake fractions of PM_{10-2.5} are more than two orders of magnitude lower than the uptake fractions of PM_{2.5}. These observations suggest that if the adverse health effects are related to the overall mass deposited, then the human health damage for PM_{10-2.5} could be of the same order magnitude as the human health damage for PM_{2.5}. However, if the adverse health effects are best related to surface area of particles deposited in the respiratory tract rather than mass deposited in the respiratory tract, then adverse health effects from PM_{10-2.5} are virtually negligible compared to those of PM_{2.5}.

Deposition in the different parts of the respiratory tract

For diesel engine emissions, the fractions of the surface of PM_{2.5} that deposit in the nasopharyngeal, the tracheobronchial, and the alveolar zones, are, respectively, typically 10%-14%, 35%-39% and 47%-56% (Table 7-1). When evaluated by mass, the fractions are typically 18%, 26% and 56% for the three zones respectively (Table 7-1). Whether the uptake fractions for PM_{10-2.5}, are expressed per mass or surface area of particles, the fractions of PM_{10-2.5} that deposit in the nasopharyngeal, the tracheobronchial, and the alveolar zones, are, respectively, about 67%, 8% and 26% (Table 7-1). This observation is consistent with Figure 6-2 which indicates that large particles will deposit predominantly in the nasopharyngeal zone.

Comparison with values found in Chapter 4

The intake fraction obtained for PM_{2.5} emitted in high population density areas, $1.8 \times 10^{-5} \text{ kg}_{\text{inh}}/\text{kg}_{\text{emi}}$, is within a factor of two of the one obtained in Chapter 4 which is $3.0 \times 10^{-5} \text{ kg}_{\text{inh}}/\text{kg}_{\text{emi}}$. The intake fraction obtained for PM_{2.5} emitted in low population density areas is a factor of two lower than the one obtained in Chapter 4 which is $8.4 \times 10^{-8} \text{ kg}_{\text{inh}}/\text{kg}_{\text{emi}}$ for 1 pers/km², therefore approximately $4.2 \times 10^{-7} \text{ kg}_{\text{inh}}/\text{kg}_{\text{emi}}$ for 5 pers/km². Considering the variability of input parameters, such as population density around the zone of emission, urban mixing height and wind speed, a factor of two difference can be considered small.

7.7. Sensitivity and limitations

Sensitivity analysis

Table 7-2 presents the main input parameters in the model and their influence on the final uptake fraction. The parameters that are sensitive and which have a high range of possible values include the wind speed (U), the mixing height (H), the population density (PD), and the distance downwind of the emission where the population is present (x).

Table 7-2: Main input parameters in the model and their influence on final uptake fraction.

Parameters	Value used by default	Range	Influence on the uptake fraction (if the parameter is multiplied by 2) (mass based)
Fate:			
Wind speed (U)	4 m/s	1 – 10	increase in speed decreases uptake fraction (app \times 0.6)
Mixing height (H)	1,000 m	300 – 2,000	increase in height decreases uptake fraction (app \times 0.5)
Rain intensity (R_{in})	1.3 mm/h	0.1 – 10	increase in intensity decreases uptake fraction (app \times 0.85)
Raindrop diameter (d_r)	1 mm	0.3 – 3	increase in drop size increases uptake fraction (app \times 1.2)
Raindrop fall velocity (v_r)	4 m/s	2 – 8	does not influence uptake fraction
Dry deposition velocity (v_d)	0.007 and 2 cm/s (for d_p = 1 and 0.001 μ m)		increase in dry deposition velocity decreases uptake fraction (app \times 0.97)
Exposure:			
Population density (PD)	5, 50, 500, and 5,000 pers/km ² respectively	1 – 20,000	increase in population density increases uptake fraction (app \times 2)
Breathing rate (BR)	13.3 m ³ /(pers·d)	11-20	increase in breathing rate increases uptake fraction (exactly \times 2)
Modeling:			
Horizontal increment (dx)	1,000 m	500 – 2,000	increase in dx increases uptake fraction (max \times 1.04)
Distance downwind where the population is present (x)	1,000 km	100-4,000	increase in x increases uptake fraction (app \times 1.8)

Limitations of the model

One of the main limitations of the model is that wind is considered to be continuous and unidirectional. Furthermore, phenomena like coagulation, homogeneous nucleation, heterogeneous condensation, and the possible effects of van der Waals forces, electronic interactions, and laminar shear are not considered. The influence of buildings on the indoor fate and exposure to PM from outdoor origin (Riley et al. 2002, Liu and Nazaroff 2003) has not been considered in the present modeling. The possible influence of the actual shape (particles are not smooth spheres) and composition of PM on its fate is also not taken into account. The fact that secondary PM formation can shift the size distribution of primary PM emissions is not considered in the present analysis. Present results are based on only one emission study. Finally, a similar analysis could be applied to number of particles and secondary PM in order to generate intake fractions consistent with those for primary PM. Further research should be carried out to address these issues.

7.8. Conclusions

This chapter proposes a consistent and feasible implementation of air pollutant dynamics for the evaluation of exposure to particulate matter. It introduces several ideas, including (i) that for the case of particles, the exposure metric should not be expressed as the intake fraction (the mass inhaled over the mass emitted), but as the uptake fraction (the amount deposited in the respiratory tract over the amount emitted); (ii) that the uptake fraction can easily be expressed as surface area or number of particles deposited (per mass emitted) instead of simply as mass deposited, to better integrate surface area or number of particles should they appear to be a better metric to measure adverse health effects than mass; (iii) that PM_{10-2.5} can be evaluated consistently with PM_{2.5} and not simply considered as negligible; and (iv) that the total uptake fraction of PM_{2.5} and PM_{10-2.5} should be evaluated, taking into account the particle size distribution of the emission and its subsequent differential behavior in the atmosphere as one moves farther from the source.

The results show that particles within the accumulation mode (\sim 0.1 – 1 μ m) will stay in the air for a long time ($>$ 3 days) and even cities situated hundreds of kilometers downwind from the source will substantially contribute to the overall uptake fraction. When evaluated by mass deposited in the respiratory tract, compared to particles smaller than 2.5 μ m, exposure to

particles greater than 2.5 μm is not negligible, but if evaluated by surface area of particles deposited in the respiratory tract, exposure to particles greater than 2.5 μm can be considered negligible. The uptake fraction for particles is determined for different weather conditions, for different population density patterns, and for different sources. For the particle size distributions assessed (ambient PM distribution from Seinfeld and Pandis (1998) and diesel tailpipe emissions from Norbeck et al. (1998)), the ratio of uptake fraction to intake fraction for $\text{PM}_{2.5}$ is found to be 0.3.

These uptake fractions can be combined with appropriate effect factors to generate the associated characterization factors, also known as human damage factors. These factors can be used in life-cycle assessment of human health effects caused by particles, as well as for exposure and risk assessments. Since adverse health effects associated with an overall system are often found to be dominated by particle emissions (Chapters 5), such an approach can help reduce the uncertainties and improve the reliability of risk assessments, as well as life-cycle assessment results.

7.9. Acknowledgments

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8. Effect and characterization factors for primary particulate matter: Reevaluation incorporating air pollutant dynamics

In this chapter, I develop new effect factors for particulate matter (PM) by considering air pollutant dynamics in combination with (i) the uptake fractions obtained in Chapter 7 applied to primary PM for different conditions, (ii) the updated PM effect factors obtained in Chapter 4, and (iii) dose-response and severity estimates using the uptake fraction concept. I use these newly developed effect factors with the uptake fractions calculated in Chapter 7 to obtain new characterization factors for PM that include the influence of air pollutant dynamics.

8.1. Summary

PM is recognized as a significant source of adverse health effects. While evidence in the literature indicates that estimates of health damage caused by PM should incorporate the surface area and number of particles, current life-cycle impact assessment methods only evaluate PM damage based on overall mass intake. I propose herein a method to address this issue. I use ambient PM size distributions and the consequences of the differential deposition in the respiratory tract to reevaluate the effect factors for particles based, respectively, on mass and surface area of particles deposited in the respiratory tract instead of only mass inhaled. Combining these uptake-based effect factors with fate and exposure factors (expressed as uptake fraction in the respiratory tract), I derive characterization factors, also known as human damage factors, for $PM_{2.5}$, $PM_{10-2.5}$ and PM_{10} for diesel engine tailpipe emissions and for different population density patterns. I find that characterization factors of $PM_{2.5}$ vary by three orders of magnitude, influenced first by variation in population density patterns, then by the metric (mass or surface area of particles) with which the effects are associated and lastly by the zone within the respiratory tract with which the effects are associated. I use the example of a diesel car to illustrate the influence of these factors on human health. The method presented here can be used as an example to refine life-cycle assessments that show PM as a dominant impact among all pollutant emissions. The specific characterization factors can be applied as a sensitivity study to the many studies that find diesel PM emissions to be a dominant impact.

8.2. Introduction

This chapter incorporates the influence of size distribution in the evaluation of effect factors and characterization factors for PM.

As presented in Chapters 4, 5 and 6, though PM seems to be one of the most significant source of adverse health effects, characterization factors of PM within life-cycle assessment is currently somewhat limited with respect to its reliability.

In life-cycle impact assessment modeling, neither the particle size distribution nor the actual shape or composition of particles is taken into account for the evaluation of PM characterization factors, also known as human damage factors. However, the exposure and health assessment community suggests exploring whether these two parameters can play an important role in determining the relative characterization factors for PM. Indeed, risks for health can vary depending on the PM composition (Franklin et al. 2008), increase with decreasing size of particles (Peters et al. 1997) and may be better correlated with particle surface area than mass or number of particles (Froines 2006, Oberdörster et al. 2005, Borm et al. 2007).

As explained in Chapter 4, in life-cycle impact assessment methods, the characterization factor of PM₁₀ is often derived from the PM_{2.5} characterization factor, assuming that the entire effect of PM₁₀ is due to particles smaller than 2.5 µm and using an estimate of the mass ratio between PM_{2.5}, PM_{10-2.5} and PM₁₀. The common assumption that PM greater than 2.5 µm has no adverse health effect is subject to debate (Perez et al. 2009, Andersen et al. 2008), and further investigation should be undertaken to better capture potential effects. In the present chapter, the characterization factors for PM smaller or greater than 2.5 µm are evaluated using a consistent fate-and-exposure modeling framework for particles up to 10 µm.

In their review of research on PM within life-cycle impact assessment, Potting et al. (2007) suggested the need for consistency in evaluating fate, exposure, and effects of PM. Building on the work of Potting et al. (2007), Chapter 4 reviewed models evaluating PM and recommended parameters for PM fate and exposure. One of the recommendations in Chapter 4 for further research to better address PM in life-cycle assessment is to consider particle size distribution in the modeling. This raises the question of what is the influence of the particle size distribution on the fate, exposure, and effect factors? Chapter 6 suggests addressing this question using the concept of uptake fraction, the fraction of emissions that deposits in different zones of the respiratory tract. A method is developed in Chapter 7 to calculate the fate, exposure and uptake of PM in the respiratory tract considering the influence of particle size distribution. Evaluating effect factors and characterization factors considering the influence of particle size distribution has not been done. To do so, one needs to evaluate exposure of ambient PM in terms of deposition in the respiratory tract instead of intake through inhalation.

In life-cycle assessment, the total damage score (*DS*) for a given substance *i* is estimated as the product of the total emission (*S*) and a characterization factor (*CF*):

$$DS_i = S_i \times CF_i \quad (8-1)$$

For a substance *i*, the characterization factor can be expressed as the product of an intake fraction (*iF*) (Bennett et al. 2002) and an effect factor based on intake (EF_i^{iF}) (Rosenbaum et al. 2007):

$$CF_i = iF_i \times EF_i^{iF} \quad (8-2)$$

For the specific case of PM, Chapter 6 suggests using the metric of the uptake fraction (*uF*) instead of the intake fraction and an effect factor based on uptake. The intake fraction by inhalation measures the fraction of the emissions that is inhaled by an entire population, and the uptake fraction extends this concept to measure the fraction of emissions that deposits in the respiratory tract⁶. For most substances, shape and composition are unique and the uptake fraction is directly proportional to the intake fraction. However for PM, Chapter 6 suggests that the uptake fraction is a better metric than the intake fraction because only particles that deposit in the respiratory tract have potential adverse health effects, and for two identical intake fractions of PM, the uptake fractions and therefore the adverse health effects can vary with particle size distribution.

Furthermore, the uptake fraction can be divided into uptake in different zones of the respiratory tract (nasopharyngeal, tracheobronchial and alveolar), which have different deposition rates depending on particle size distribution, and likely lead to different adverse health

⁶ The reason for using the term "uptake fraction" instead of "deposited fraction" is that the latter is commonly used to refer to the fraction of pollutant removed from the atmosphere by physical-chemical phenomena.

effects. Nel (2005) suggests that smaller particles caused more serious adverse health effects than coarse particles and that they penetrate deeper in the lungs. Mayer et al. (2001) suggest that the particles deposited into the nasopharyngeal region are eliminated by natural defense systems, whereas particles penetrating the tracheobronchial and the alveolar systems are less effectively removed by natural defenses and can cause more adverse health effects.

The objectives of this chapter are: (i) to present a method to calculate the PM effect and characterization factors using deposition in the respiratory tract instead of intake, as well as using surface area of particles instead of solely particle mass; (ii) to discuss the influence on the PM effect and characterization factors due to making these changes; and (iii) to discuss whether or not the effect of particles greater than 2.5 μm can be neglected.

The chapter is divided into three parts. First the epidemiology-based effect factors for primary PM are reevaluated and attributed to uptake instead of intake, expressing the effect per mass, per surface area of particles deposited in the respiratory tract. Second, these effect factors are multiplied by the PM uptake fractions calculated in Chapter 7 from diesel engine tailpipe emissions for different population patterns to estimate the associated characterization factors. Finally, a case study is performed to show the influence of the updated characterization factors on actual life-cycle assessment results.

This chapter focuses on the evaluation of the influence of the size of primary PM, considering PM as spheres with a density of 1 g/cm^3 . The influence of the actual shape and composition is outside of the scope of this chapter and needs to be addressed in further investigations.

8.3. Methods

To make the uptake approach operational requires that I evaluate the effect factor in terms of uptake rather than intake. I calculate an uptake-based effect factor that corresponds to the conditions of the epidemiological study from which the intake-based effect factor is derived and then multiply this effect factor by the uptake fraction to yield the characterization factor.

Uptake-based effect factor

The uptake-based effect factor ($EF_i^{total uF}$) can be related to the intake-based effect factor (EF_i^{iF}) through the fraction of the intake that is deposited in the respiratory tract ($F_i^{total uptake}$):

$$EF_i^{total uF} = EF_{i,epidemiological}^{iF} / F_{i,epidemiological}^{total uptake} \quad (8-3)$$

where EF is in units of disability adjusted life years (DALY – Murray and Lopez 1996) per amount taken in (iF) or taken up (uF), and F is in units of amount taken up per amount taken in, with “amount” referring to either the mass (in kg) or the surface area (in m^2) of particles. Note that the superscripts “iF” and “uF” are used to specify whether the effect factor is expressed in DALY per amount of substance intake or per amount of substance uptake, respectively.

In Chapter 4, I reviewed dose-response values derived from epidemiological studies of PM and severity factors typically used in human health damage studies and found an intake-based effect factor for ambient PM_{10} of 82 DALY/ $\text{kg}_{\text{inhaled}}$, dominated by premature mortality and chronic bronchitis.

Fraction of inhaled particles deposited in the respiratory tract for epidemiological studies

To determine the fraction of inhaled particles that are deposited in the respiratory tract ($F_i^{total uptake}$) while being consistent with the intake-based effect factor derived from

epidemiological studies, the ambient aerosol distribution should ideally be the particle distribution at the time that adverse health effects were observed. However, the intake-based effect factor of 82 DALY/kg_{inhaled} for ambient PM₁₀ is based on a combination of different studies, of which none provide the associated particle size distribution. Thus, assumptions must be made regarding the typical ambient particle size distribution. I use here the urban aerosol mass distribution suggested by Seinfeld and Pandis (1998) (Figure 8-1).

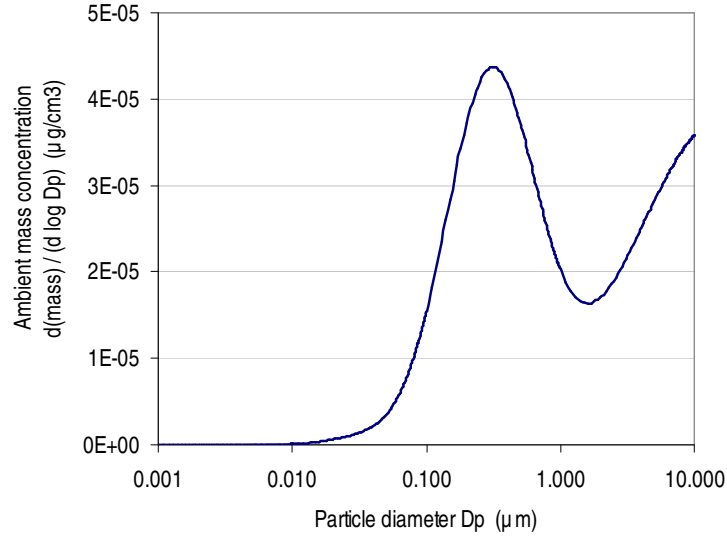


Figure 8-1: Typical urban aerosol mass distribution (generated using the data from Seinfeld and Pandis 1998).

This particle distribution in an urban environment (Figure 8-1) is then multiplied by the total lung deposition curves (Figure 6-2 in Chapter 6). This multiplication yields the fraction of inhaled particles deposited in each of the three zones of the respiratory tract (nasopharyngeal, tracheobronchial, alveolar) by mass or surface area of particles.

Total uptake-based assessment framework for characterizing the impact of PM

Total uptake fractions can be multiplied by effect factors calculated in the present chapter to generate the associated characterization factors (CF , in DALY per amount emitted) of PM of size i , as described in Equation 8-2:

$$CF_i = uF_i^{total} \times EF_i^{total uF} \quad (8-4)$$

Influence of the deposition zone in the respiratory tract

In addition to the total uptake fraction in the respiratory tract, I explore the possibility of separating this total uptake fraction into uptake fractions for three different respiratory tract zones (nasopharyngeal, tracheobronchial, alveolar) with corresponding effect factors:

$$CF_i = uF_i^{total} \times EF_i^{total uF} = uF_i^{nas} \times EF_i^{nas uF} + uF_i^{tra} \times EF_i^{tra uF} + uF_i^{alv} \times EF_i^{alv uF} \quad (8-5)$$

I start from the default case in which all deposited particles have a similar effect and then explore two possible scenarios based on studies suggesting differing health effects by zone (Nel 2005, Mayer et al. 2001): (i) only PM deposited in the tracheobronchial and alveolar zones contribute to the effect, and (ii) only PM deposited in the alveolar zone contributes to the effect. In these cases the effect factors can be determined using Equation 8-3 as a function of the fraction

deposited in the nasopharyngeal ($F_i^{nas\ uptake}$), tracheobronchial ($F_i^{tra\ uptake}$) and alveolar zones ($F_i^{alv\ uptake}$). The values $F_i^{nas\ uptake}$, $F_i^{tra\ uptake}$, $F_i^{alv\ uptake}$, and $F_i^{total\ uptake}$ are found by multiplying the particle distribution function of Figure 8-1 with the deposition curves from Figure 6-2.

Application to diesel PM emissions using uptake fraction for diesel particulates

I evaluate here the characterization factors of diesel PM emissions, using the uptake fractions from Chapter 7, which account for the specific particle size distribution for diesel engine PM emissions (Norbeck et al. 1998), different weather conditions, and different population density patterns (high, medium, low, and unknown population density at the location of emission).

8.4. Results

This section aims at attributing the effects to uptake instead of intake.

Fraction of ambient aerosol deposited in the respiratory tract for epidemiological conditions

Table 8-1 presents the fractions of inhaled ambient aerosol deposited in the nasopharyngeal zone ($F_i^{nas\ uptake}$), the tracheobronchial zone ($F_i^{tra\ uptake}$) and the alveolar zone ($F_i^{alv\ uptake}$); the total fraction of inhaled ambient aerosol deposited in the respiratory tract ($F_i^{total\ uptake}$); and the fraction of inhaled ambient aerosols that is inhaled and then exhaled, by mass (in kg) or surface area (in m^2) of particles inhaled.

Table 8-1: Fractions of ambient aerosols inhaled that is deposited in the respiratory tract or exhaled, with ambient particle size distribution functions based on Seinfeld and Pandis (1998), expressed per mass or surface area of particles inhaled.

	Fraction deposited in the following zones of the respiratory tract				Fraction inhaled that is exhaled
	Naso-pharyngeal	Tracheo-bronchial	Alveolar	Total respiratory tract	
	per $kg_{inhalated}$				
Mass based [kg_{dep}/kg_{inh}]	0.22	0.061	0.15	0.42	0.58
Surface based [m^2_{dep}/kg_{inh}]	$9.8E+02$	$2.2E+03$	$3.7E+03$	$6.9E+03$	$1.2E+04$
Number based [$\#/kg_{inh}$]	$3.2E+17$	$9.9E+17$	$4.1E+17$	$1.7E+18$	$5.4E+17$
	per $m^2_{inhalated}$				
Mass based [kg_{dep}/m^2_{inh}]	$1.2E-05$	$3.3E-06$	$7.8E-06$	$2.3E-05$	$3.1E-05$
Surface based [m^2_{dep}/m^2_{inh}]	0.053	0.12	0.20	0.37	0.63
Number based [$\#/m^2_{inh}$]	$1.7E+13$	$5.3E+13$	$2.2E+13$	$9.2E+13$	$2.9E+13$

As a matter of comparison, using particle size distributions measured for Vienna (range measured goes from 0.01 μm to 16 μm — Horvath et al. 1996), and combining these values with the total lung deposition curve (Figure 6-2), I obtain 0.41 $kg_{deposited}/kg_{inhalated}$ and 0.44 $kg_{deposited}/kg_{inhalated}$ for urban and suburban conditions, respectively. These values are close to the value of 0.42 $kg_{deposited}/kg_{inhalated}$ obtained using data from Seinfeld and Pandis (1998).

Uptake effect factor expressed per mass, surface area or number of particles

Assuming first that the adverse health effects caused by PM_{10} are equal for each respiratory tract deposition zone, combining the intake-based effect factor for PM_{10} of 82 DALY/ $kg_{inhalated}$ with the deposited fractions of Table 8-1 provides the uptake-based effect factors of, respectively, 200 DALY/ $kg_{deposited}$ and 0.012 DALY/ $m^2_{deposited}$. Based on the same approach, uptake-based effect factors can be derived assuming that adverse health effects are only caused by PM_{10} deposition in the tracheobronchial and alveolar zones or solely in the alveolar zone. Table 8-2 lists these different uptake-based effect factors, expressed per mass or surface area of particles deposited in the adverse zone(s) of the respiratory tract.

Table 8-2: Uptake-based effect factors for ambient aerosols, with ambient particle size distribution functions based on Seinfeld and Pandis (1998), expressed per mass or surface area of particles deposited in the respiratory tract.

	Assuming equal contribution to adverse health effects from all zones (nasopharyngeal, tracheobronchial, alveolar) of the respiratory tract	Assuming that only particle deposited in the tracheobronchial and alveolar zones contribute to adverse health effects	Assuming that only particle deposited in the alveolar zone contribute to adverse health effects	Units
Effect factors expressed per mass deposited	200	390	550	[DALY/kg _{uptake in the adverse zone}]
Effect factors expressed per surface area deposited	0.012	0.014	0.022	[DALY/m ² _{uptake in the adverse zone}]

For example, if only particles deposited in the alveolar zone cause adverse health effects, then the effect factor of particles is entirely due to the fraction deposited into the alveolar zone and is thus 82 DALY divided by 3,700 m² deposited in the alveolar zone, representing 0.022 DALY per m² deposited in the alveolar zone.

Further investigation should be undertaken to identify how the mechanisms involved in uptake by the body differ depending on the zone of deposition in the respiratory tract as well as how composition varies with size. Such findings would help determine which effect factor evaluated in Table 8-2 is optimal for capturing the influence of the actual uptake mechanisms on the adverse health effects caused by PM.

Effect factors of PM_{10-2.5}

Figure 8-2 shows which fraction of the effect factor derived from epidemiological studies can be attributable to PM_{2.5} and to PM_{10-2.5} for different modeling approaches. The interpretation of Figure 8-2 is the following: if the sizes of the gray and of the crosshatched bars are the same, then the effects from ambient PM are distributed equally between PM_{2.5} and PM_{10-2.5}. Therefore, if adverse health effects are better represented by mass deposited in the respiratory tract, then the effect factor of PM_{10-2.5} represents between half and the same value as the effect factor of PM_{2.5}. However, if effects are better represented by surface area deposited in the respiratory tract, between 1% and 4% of the effect of ambient particles can be attributable to PM_{10-2.5}, indicating that the effect factor of PM_{10-2.5} is between 25 and 100 times lower than the effect factor of PM_{2.5}. I observe that the metric (mass or surface area of particles) has more influence on how the effects are attributable to fractions smaller and greater than 2.5 μm than the zone of the respiratory tract that is considered to actually cause adverse health effects. This analysis does not consider potential differences in severities between PM_{2.5} and PM_{10-2.5} caused endpoints. Because studies suggest that PM smaller and greater than 2.5 μm cause different endpoints (Andersen et al. 2008, Perez et al. 2009), further research should aim at quantifying these differences in severities.

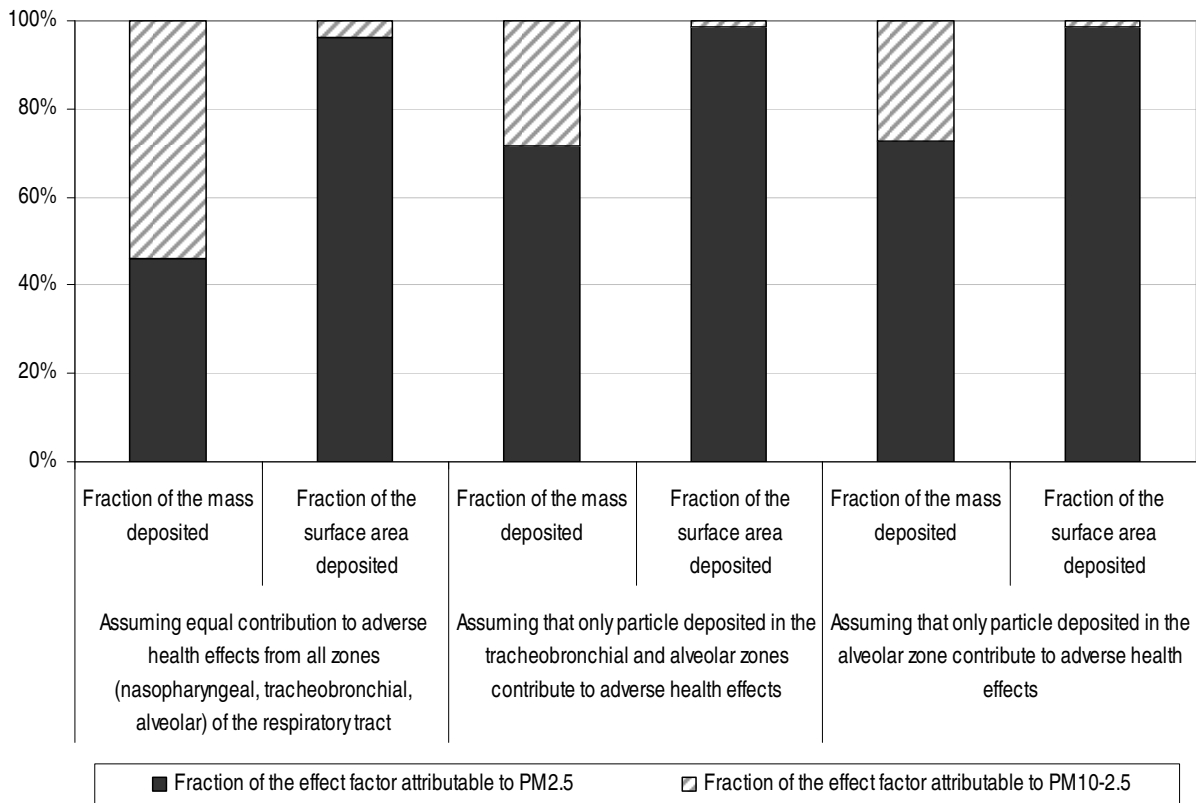


Figure 8-2: Fraction of the effect factor from epidemiological studies attributable to PM_{2.5} and PM_{10-2.5} for different modeling approaches.

Characterization factors for diesel particulate

Combining the uptake fractions calculated in Chapter 7 (Table 7-1) for diesel engine tailpipe PM emissions with the uptake-based effect factors shown in Table 8-2 gives the associated characterization factors presented in Table 8-3. Example: The characterization factor for PM_{2.5}, emitted in a high population density area, assuming equal contribution to adverse health effects from all zones, and using mass as proxy to extrapolate the adverse health effects (i.e., first value of the Table 8-3) is 5.4×10^{-6} kg_{deposited}/kg_{emitted} (Table 7-1) multiplied by 200 DALY/kg_{uptake in the adverse zone = deposited}, which gives a characterization factor of 1.1×10^{-3} DALY/kg_{emitted}.

Table 8-3: Characterization factors for diesel engine tailpipe PM emissions for the different scenarios studied, under mixed weather conditions.

Population density pattern at emission	Assuming equal contribution to adverse health effects from all zones (nasopharyngeal, tracheobronchial, alveolar) of the respiratory tract		Assuming that only particle deposited in the tracheobronchial and alveolar zones contribute to adverse health effects		Assuming that only particle deposited in the alveolar zone contribute to adverse health effects	
	Metric used as proxy to extrapolate adverse health effects					
	mass	surface	mass	surface	mass	surface
	Characterization factor of PM_{2.5} [DALY/kg_{emi}]					
high	1.1E-3	3.1E-3	1.7E-3	3.3E-3	1.7E-3	3.2E-3
medium	1.1E-4	4.3E-4	1.8E-4	4.3E-4	1.7E-4	3.7E-4
low	1.1E-5	4.1E-5	1.8E-5	4.2E-5	1.7E-5	3.7E-5
unknown	3.0E-4	9.1E-4	4.8E-4	9.4E-4	4.6E-4	9.0E-4
	Characterization factor of PM_{10-2.5} [DALY/kg_{emi}]					
high	4.2E-4	4.3E-5	2.7E-4	1.7E-5	2.9E-4	2.1E-5
medium	4.2E-5	4.3E-6	2.7E-5	1.7E-6	2.9E-5	2.1E-6
low	4.2E-6	4.3E-7	2.7E-6	1.7E-7	2.9E-6	2.1E-7
unknown	1.1E-4	1.2E-5	7.4E-5	4.7E-6	7.9E-5	5.6E-6
	Characterization factor of PM₁₀ [DALY/kg_{emi}]					
high	1.1E-3	3.0E-3	1.1E-3	2.8E-3	1.3E-3	2.5E-3
medium	1.1E-4	4.1E-4	1.6E-4	3.6E-4	1.8E-4	2.8E-4
low	1.1E-5	3.8E-5	1.5E-5	3.5E-5	1.7E-5	2.7E-5
unknown	3.0E-4	8.8E-4	3.3E-4	7.9E-4	3.8E-4	6.9E-4

^a “mixed” weather conditions (Chapter 7) are used as a proxy for “unknown weather conditions.”

The value highlighted in bold and gray represents the case that is typical in life-cycle impact assessment methods (i.e., an emission in a region with an unknown population density, using mass as a metric for adverse health effects and assuming no differences in sensitivity to adverse health effects among respiratory tract zones).

Influence of the population density

There is considerable variation among different population density patterns (up to two orders of magnitude). When effects are considered proportional to mass or surface area uptake, the characterization factor varies by one order of magnitude between emissions in low versus medium population density areas and between emissions in medium versus high population density areas.

Influence of the uptake zone in the respiratory tract

Table 8-3 shows that characterization factors can vary by a factor of two based on where particles deposit in the respiratory tract and which zones cause adverse health effects. A higher fraction of the PM_{10-2.5} is trapped in the nasopharyngeal zone compared to PM_{2.5}. Thus if the nasopharyngeal zone is found to be less sensitive to adverse health effect than the zones deeper in the respiratory tract, the characterization factor of PM_{10-2.5} will be lower than if all zones are assumed to be equally sensitive to particles.

Characterization factors of PM_{10-2.5} for diesel particulate

Table 8-3 indicates that the characterization factors of PM_{10-2.5} relative to PM_{2.5} are typically 15%-40% and 1% when effects are considered proportional to mass and surface area of particles deposited, respectively. This suggests that if the effects of PM are considered correlated to the surface area of particles, the effects of PM_{10-2.5} can be neglected. However, if mass deposited is the best indicator for effects, the effects of PM_{10-2.5} can be comparable to the effects of PM_{2.5}. Note that effects from PM_{10-2.5} is still strongly debated and further research is needed.

Characterization factors of PM₁₀ for diesel particulate

The characterization factors of PM_{2.5} are significantly greater than those of PM_{10-2.5} but approximately equivalent to those of PM₁₀ because in the data used for diesel engine emissions 96% of the mass of PM₁₀ is in fact PM_{2.5} (Norbeck et al. 1998).

8.5. Case study

To apply the results above, I present a case study here using personal transportation with a diesel engine car. Selected inventory data are presented in Table 8-4 for the average European diesel car fleet in 2005, based on Ecoinvent v2.01 (Frischknecht 2005, Ecoinvent 2009). Detailed computations can be found in the section 8.9.

Table 8-4: Selected inventory data for a diesel engine car, per km driven (average European fleet in Europe in 2005, occupancy is 1) (Ecoinvent v2.01 — Frischknecht 2005, Ecoinvent 2009).

Unit	diesel consumption kg	Pollutant emitted						
		CO ₂ g	CO mg	NH ₃ mg	NO _x mg	SO ₂ mg	PM _{2.5} mg	PM _{10-2.5} mg
Tailpipe	0.061	190	160	5.1	700	6.1	58	13
Life cycle, excluding tailpipe ^a	-	80	280	2.8	260	360	29	33

^a Car life cycle excluding tailpipe refers to diesel production and delivery; car manufacturing, maintenance and end-of-life; and road construction and maintenance.

Figure 8-3 shows the influence of the characterization factors presented in Table 8-3 for PM_{2.5} and PM_{10-2.5} on the total impact of the life-cycle assessment of a diesel car, evaluated per km driven. The life-cycle impact assessment method used is the IMPACT 2002+ v2.1 method (Jolliet et al. 2003). The results using the default characterization factors of PM_{2.5} and PM_{10-2.5} are presented in Figure 8-3, along with the results obtained using the characterization factors of PM_{2.5} and PM_{10-2.5} developed in this chapter that take into account the population density pattern at the emission locations. The four cases consider tailpipe emissions for areas that have high, medium, low, and unknown population densities. The PM impacts shown are calculated from characterization factors (Table 8-3) based on surface area deposited, with error bars representing the variation in human health damage score if mass (lower value) deposited was instead used as the basis for PM characterization factors.

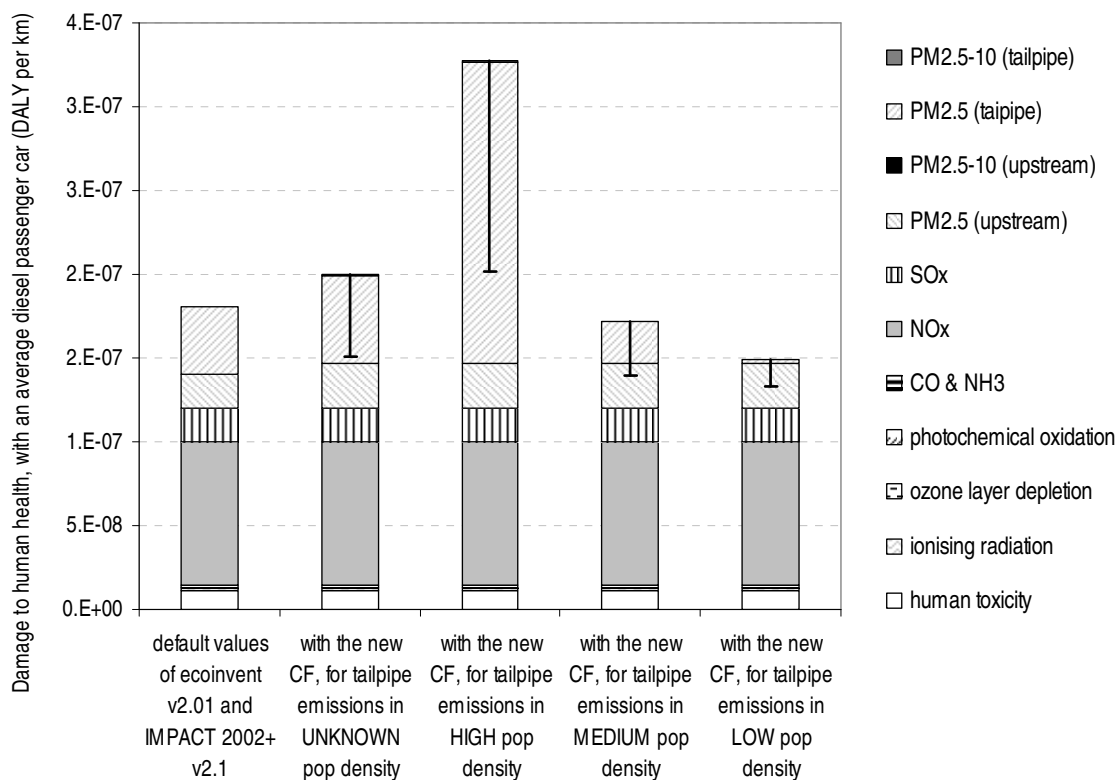


Figure 8-3: For the life-cycle assessment of a diesel engine car, evaluated per km driven, the contribution of each pollutant to human health damage and the influence of the type of PM characterization factor. PM characterization factors are based on using the surface area deposition in the respiratory tract, with error bars representing the mass basis (lower value).

The deposition metric used to evaluate adverse health effects and the location of emissions of the particles influences the final results of the life-cycle assessment. If mass is used, the overall results are in the same range as when using the default values provided by IMPACT 2002+. Finally, I find that for the surface area metric, the impacts caused by PM_{10-2.5} are negligible compared to the impacts of PM_{2.5} (more than two orders of magnitude lower).

The purpose of Figure 8-3 is to illustrate the variation of the impacts on human health caused by variations in the characterization factor of PM, depending on the deposition metric used to evaluate adverse health effects, and where the emissions occur. Impacts of other pollutants (e.g., NO_x) will also vary depending on the location of the emissions. However, these are not evaluated in the present chapter and no conclusion regarding the variation of pollutants other than PM should be drawn from this example.

This example shows that when particles dominate the human health damage caused by pollutants, a clearly defined metric for adverse human health effects and the location of particle emission can improve the accuracy and quality of final life-cycle assessment results.

8.6. Outlook

As demonstrated by the variation in impacts depending on different assumptions, I find that more research is needed to evaluate which of the zones in the respiratory tract is most likely

to cause adverse health effects as well as whether these effects are more correlated to mass, surface area or the number (not evaluated here) of particles deposited in the respiratory tract.

I recommend applying the uptake fraction concept to secondary particles, extending the present assessment to number of particles, other studies on diesel, to sources of PM emissions other than diesel, and better addressing actual shape (i.e., opposite to the simplification made in Chapters 6, 7 and 8, PM are not smooth spheres) and composition of particles.

The method introduced in this chapter and the resulting characterization factors of PM_{2.5} and PM_{10-2.5} — and PM₁₀ if attention is paid to avoid double counting — can be directly used in the categories related to the adverse health effects caused by particles within damage-orientated life-cycle impact assessment methods. Based on the findings of Froines (2006), Oberdörster et al. (2005) and Borm et al. (2007), I suggest that using characterization factors that consider the surface area deposited in the respiratory tract can be a proxy for adverse health effects as good as mass of particles. The characterization factors based on mass or surface of particles deposited can be used as uncertainty analysis.

However, the characterization factors based on Chapters 6, 7 and 8 are exploratory. They are not yet robust enough to be used as a replacement of characterization factors found in Chapter 4 in life-cycle assessments aiming at giving recommendations for policy or industry. The developments using mass or surface of particles in Chapter 6, 7 and 8 serve as an illustration of the way that alternatives could be implemented in the event that health studies demonstrated the importance of considering such an approach.

Because life-cycle assessment studies are often found to be dominated by particle emissions (Jolliet et al. 2006, Chapter 5), better understanding and modeling characterization factors of PM can help reduce the uncertainties associated with the different scenarios and improve life-cycle assessment results related to human health damage.

8.7. Acknowledgments

I wish to thank Professor William Nazaroff from the University of California, Berkeley, for his invaluable feedback contributing to improve this work, as well as Holly Fox and Naomi Lubick for the editing.

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8.9. Supporting information for this chapter

UFPM tool

The UFPM tool is available at <http://www.impactmodeling.org> or by contacting sebastien.humbert@cal.berkeley.edu.

Case study

Table 8-5 presents the impact scores for the scenarios of Figure 8-3 (in DALY/km).

Table 8-5: Impact scores for the scenarios of Figure 8-3 (in DALY/km) (CF stands for “characterization factors”).

Contributor to damage to human health	default values of ecoinvent v2.01 and IMPACT 2002+ v2.1	with the new CF, for tailpipe emissions in UNKNO WN pop density	with the new CF, for tailpipe emissions in HIGH pop density	with the new CF, for tailpipe emissions in MEDIU M pop density	with the new CF, for tailpipe emissions in LOW pop density	Source
human toxicity	1.2E-08	1.2E-08	1.2E-08	1.2E-08	1.2E-08	Ecoinvent 2.01 (Frishknecht 2005 for life-cycle inventory and IMPACT 2002+ (Jolliet et al. 2003) for impact assessment); Original values, given in points, are multiplied by 1.6 to correct occupancy (Ecoinvent data are provided in p-km and it assumes 1.6 person/car) and using the IMPACT 2002+ conversion factor of 0.0071 DALY/point (Jolliet et al. 2003); CF for CO, NH ₃ , NO _x and SO _x are respectively 7.3E-7, 8.5E-5, 8.9E-5 and 5.5E-5 DALY/kg (Jolliet et al. 2003)
ionising radiation	1.2E-09	1.2E-09	1.2E-09	1.2E-09	1.2E-09	
ozone layer depletion	4.2E-11	4.2E-11	4.2E-11	4.2E-11	4.2E-11	
photochemical oxidation	3.4E-10	3.4E-10	3.4E-10	3.4E-10	3.4E-10	
CO and NH ₃	9.9E-10	9.9E-10	9.9E-10	9.9E-10	9.9E-10	
NO _x	8.5E-08	8.5E-08	8.5E-08	8.5E-08	8.5E-08	
SO _x	2.0E-08	2.0E-08	2.0E-08	2.0E-08	2.0E-08	
Using the surface area of particle as a proxy to extrapolate adverse health effects (main results in Figure 8-3)						<p><i>For the first column:</i> Ecoinvent 2.01 (Frishknecht 2005 for life-cycle inventory and IMPACT 2002+ (Jolliet et al. 2003) for impact assessment); Original values, given in points, are multiplied by 1.6 to correct occupancy (Ecoinvent data are provided in p-km and it assumes 1.6 person/car) and using the IMPACT 2002+ conversion factor of 0.0071 DALY/point (Jolliet et al. 2003); CF for PM_{2.5} and PM_{10-2.5} are respectively 7.0E-4 and 0 DALY/kg (Jolliet et al. 2003)</p> <p><i>For the other columns:</i> See Table 8-6</p>
PM _{2.5} (upstream)	2.0E-08	2.6E-08	2.6E-08	2.6E-08	2.6E-08	
PM _{2.5-10} (upstream)	0.0E+00	4.0E-10	4.0E-10	4.0E-10	4.0E-10	
PM _{2.5} (tailpipe)	4.1E-08	5.3E-08	1.8E-07	2.5E-08	2.4E-09	
PM _{2.5-10} (tailpipe)	0.0E+00	1.6E-10	5.6E-10	5.6E-11	5.6E-12	
Using the mass of particle as a proxy to extrapolate adverse health effects (“error bars” in Figure 8-3)						
PM _{2.5} (upstream)	2.0E-08	8.7E-09	8.7E-09	8.7E-09	8.7E-09	
PM _{2.5-10} (upstream)	0.0E+00	3.6E-09	3.6E-09	3.6E-09	3.6E-09	
PM _{2.5} (tailpipe)	4.1E-08	1.7E-08	6.4E-08	6.4E-09	6.4E-10	
PM _{2.5-10} (tailpipe)	0.0E+00	1.4E-09	5.5E-09	5.5E-10	5.5E-11	

Table 8-6 presents the emissions inventory (from Table 8-4) and characterization factors (from Table 8-3) used to calculate the impact scores related to PM_{2.5} and PM_{10-2.5} presented in Table 8-5. The impact score presented in Table 8-5 is the multiplication of the emissions with the respective characterization factors.

Table 8-6: Emissions and characterization factors (CF) used to calculate the impact scores related to PM_{2.5} and PM_{10-2.5} in Table 8-5.

Contributor to damage to human health	Emission (in mg/km) (Table 8-4)	CF (DALY/kg) (default values in IMPACT 2002+ — Jolliet et al. 2003)	CF (DALY/kg), for unknown population density area (Table 8-3)	CF (DALY/kg), for high population density area (Table 8-3)	CF (DALY/kg), for medium population density area (Table 8-3)	CF (DALY/kg), for low population density area (Table 8-3)
Using the surface area of particle as a proxy to extrapolate adverse health effects						
PM2.5 (upstream)	29	7.0E-4	9.1E-4	Upstream emissions are assumed to occur in an area with “unknown” population density		
PM2.5-10 (upstream)	33	0.0E-4	1.2E-5			
PM2.5 (tailpipe)	58	7.0E-4	9.1E-4	3.1E-3	4.3E-4	4.1E-5
PM2.5-10 (tailpipe)	13	0.0E-4	1.2E-5	4.3E-5	4.3E-6	4.3E-7
Using the mass of particle as a proxy to extrapolate adverse health effects						
PM2.5 (upstream)	29	n/a	3.0E-4	Upstream emissions are assumed to occur in an area with “unknown” population density		
PM2.5-10 (upstream)	33	n/a	1.1E-4			
PM2.5 (tailpipe)	58	n/a	3.0E-4	1.1E-3	1.1E-4	1.1E-5
PM2.5-10 (tailpipe)	13	n/a	1.1E-4	4.2E-4	4.2E-5	4.2E-6

Annual total DALYs from diesel-based road transport in the United States

Table 8-5 shows that damage to human health of diesel is approximately 2.0E-7 DALY/km driven. Because 0.061 kg of diesel was consumed for one km (Table 8-4), 2.0E-7 DALY/km driven represents a burden of 3.3E-6 DALY/kg of diesel consumed. Annual U.S. diesel consumption for road transport is approximately 6E+18 Joules (2007 values — NATS 2009). Considering that one kg of diesel contains approximately 43 MJ/kg (Ecoinvent 2009), annual U.S. diesel consumption for road transport is approximately 1.4E+11 kg. I therefore obtain an annual damage to human health from U.S. diesel-based road transport of approximately 460,000 DALYs or approximately 6% of current DALYs related to outdoor pollution (Table 5-7). As a matter of comparison, assuming that 10 DALYs represent one premature death (Table 4-4), the damage to human health from U.S. diesel-based road transport represents approximately 46,000 premature deaths per year. These values are important and suggest that air pollution-related policy should consider diesel with attention.

8.9.1. References used in the supporting information of this chapter

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9. Life-cycle assessment of coal fly ash disposal: Influence of regionalization and allocation for different alternatives

The different regionalized characterization factors developed in this dissertation are applied to the case of coal fly ash disposal. The influence of regionalization on life-cycle assessment results is evaluated. Furthermore, impact allocation is addressed for different alternatives.

9.1. Summary

Life-cycle assessment is used to evaluate coal fly ash disposal and the influence of regionalization and allocation. With differences in transportation excluded, we find that displacing Portland cement by fly ash is beneficial for the environment and provides 30 to 100 times more benefits in terms of greenhouse gases than displacing inert materials such as sand, gravel, flowable fill, road base, mineral filler, or aggregate. The use of fly ash to displace cement is environmentally advantageous if used regionally and transported only by truck, and used anywhere within the United States as long as it is transported by train. If fly ash displaces low-grade materials (i.e., inert materials) like crushed rock or sand, then it should be used within a radius of no more than approximately 50 km, by truck, further than a landfill or lagoon for fly ash or from where the low-grade material originates, whichever distance is the longest (the range increases to 200 km for train transport). United States annual fly ash production contributes less than 0.02% to United States greenhouse gas emissions if disposed of, but can contribute to a reduction of United States greenhouse gas emissions by as much as 1%, achieved by avoided emissions, if used to displace Portland cement. Regionalization slightly alters the results, but does not change the overall conclusions. However, if fly ash is considered as a co-product instead of a waste, the benefits of using fly ash can be debated, thus a switch from being a waste to being a co-product is examined using economic allocation and consequential life-cycle assessment. An economic allocation of less than 1% of the impacts from the coal-fired power plant to the fly ash would outweigh the potential benefits provided by the displaced Portland cement. Consequential life-cycle assessment indicates that if the re-use of fly ash permits the overall coal-based electricity generation to increase by 3% compared to what it would be without fly ash reuse, the additional damage from the increase in coal-based electricity generation (because, on a per kWh basis, coal is the most polluting option to produce electricity) outweighs the potential benefits provided by the displaced Portland cement. Therefore, it is important that using fly ash does not increase the competitive advantage and therefore production of coal-based electricity.

9.2. Introduction and objectives

Coal fly ash

Coal fly ash is a fine powder recovered from gases created by combustion in coal-fired power plants. In 2006, more than 70 million tonnes of coal fly ash were produced in the United States of which approximately 20 million tonnes were incorporated into cement (ACAA 2007). Traditionally, fly ash has been dumped in landfills or storage lagoons. However, much of it can be recovered and used to displace cement in various applications, such as soil stabilization, brick making, paving, and structural fills. In the United States, coal fly ash is mainly produced in the East and the Midwest. Table 9-1 shows the different applications for United States coal fly ash in 2006, in millions of metric tonnes per year.

Table 9-1: Disposal and use of United States coal fly ash for 2006 (ACAA 2007).

Application	Amount (million t/y)	(%)
Cement/concrete products/grout	15	21%
Cement/raw feed for clinker	4.2	5.7%
Flowable fill	0.11	0.2%
Structural fills/embankments	7.2	9.9%
Road base/subbase/pavement	0.38	0.5%
Soil modification/stabilization	0.65	0.9%
Mineral filler in asphalt	0.027	0.1%
Mining applications	0.94	1.3%
Waste stabilization/solidification	2.6	3.6%
Agriculture	0.081	0.1%
Aggregate	0.27	0.4%
Miscellaneous/other	1.0	1.4%
Total used	32	45%
Disposed in landfill or storage lagoons	40	55%
Total of coal fly ash generated	72	100%

Fly ash consists mostly of silica, alumina and iron, and is a pozzolan, i.e., a substance containing aluminous and silicious material that will, in the presence of water and lime, react with the calcium in the lime to produce calcium silicate hydrates (cementitious compounds). When mixed with lime and water it forms a compound similar to Portland cement. The spherical shape of the particles reduces internal friction and thereby increases the concrete's consistency and mobility, thus permitting longer pumping distances (Mehta and Monteiro 2006). Improved workability means less water is needed, resulting in less segregation of the mixture. Fly ash to be used in cement concrete must meet the requirements of ASTM C618 (1994).

Two classes of fly ash are defined: Class F and C. Fly ash that is produced from the burning of anthracite or bituminous coal is typically pozzolanic and is referred to as a Class F fly ash if it meets the chemical composition and physical requirements specified in ASTM C618. Fly ash that is produced from the burning of lignite or subbituminous coal, in addition to having pozzolanic properties, also has some self-cementing properties (i.e., the ability to harden and gain strength in the presence of water alone). When this fly ash meets the chemical composition and physical requirements outlined in ASTM C618, it is referred to as a Class C fly ash. Most Class C fly ashes have self-cementing properties. In the United States Class F fly ash is generally produced from Eastern coal. Class C fly ash is generally produced from Western coal. The latter is more commonly used for structural concrete. Indeed, concrete with Class C fly ash generally develops strength much faster than concrete with Class F fly ash because Class C fly ash has a higher calcium content. Typical amounts of fly ash in concrete range from 15% to 25% by weight of the total cementitious materials, but as much as 60% has been used (Mehta and Monteiro 2006, Marceau et al. 2002, TFHRC 2005, ToolBase Services 2005). Blended Portland cements containing fly ash from coal-fired power plants and granulated slag from the blast-furnace iron industry provide interesting examples of industrial ecology because they offer a solution to reduce the environmental impact of several industries (Mehta and Monteiro 2006, Marceau et al. 2002, TFHRC 2005).

Economic and structural approach

Fly ash is a viable substitute for Portland cement in concrete (Marceau et al. 2002). Several studies acknowledge the engineering and economic advantages of incorporating fly ash into concrete (Rostami and Brendley 2003, Majling and Roy 1993, Ayoko et al. 2005, Marceau et al. 2002, Proctor 2001, Aguirre González 2005). Quantification of the economic and structural advantages and disadvantages of the different uses of fly ash is outside of the scope of this

chapter, which focuses on the environmental burdens and benefits of the different alternatives for the disposal of fly ash.

Environmental approach

Alternative uses of fly ash were studied by Babbitt and Lindner (2007, 2008), Theis and Gardner (1990) (disposal), Mroueh et al. (2001) (pavement), Nisbet et al. (2002), Aguirre González (2005) and O'Brien et al. (2009). Babbitt and Lindner (2007, 2008) evaluated the life-cycle inventory of six beneficial uses for fly ash: concrete production, structural fills, soil amendments, road construction, blasting grit and roofing granules, and wallboard. They showed the beneficial use of fly ash to yield reductions in raw material requirements and various emissions. Mroueh et al. (2001) performed a life-cycle assessment on the possibility of using fly ash as a pavement material. Their results show that when cement is substituted with fly ash, energy consumption and emissions are lowered for most applications. However, compared to other materials such as sand or aggregate, fly ash does not appear to provide substantial benefits when used as a pavement structure. Though Carroll et al. (1998) mentioned the issue of allocating part of the coal power plant impacts to the fly ash, they did not quantify it and provided results for life-cycle assessment of concrete considering that fly ash has no impacts. O'Brien et al. (2009) evaluated the influence of transportation distance on embodied greenhouse gas emissions and water consumption in fly ash concrete considering that fly ash is a waste and therefore has no impacts as such.

Fly ash can be transported over long distances throughout United States. Previous chapters indicate that impacts on human health can vary significantly depending on where the pollutants are emitted within the United States. However, no life-cycle assessment has been performed on the alternative uses of fly ash considering the influence of regionalization for the damage on human health, as well as how allocating the impacts of the coal power plant would change the results.

Objectives of this chapter

The objectives of this chapter are to quantify the environmental burdens and benefits of the different alternatives to the disposal or use of fly ash and evaluate how the results change when considering the influence of regionalization on human health damage, as well as the influence of allocation of the impacts of the coal-fired power plants. The potential environmental benefits and burdens associated with the entire United States annual fly ash production are also evaluated.

9.3. Method

Life-cycle assessment

First a life-cycle assessment (ISO 2006a, 2006b) of the different alternatives to dispose of fly ash was done using inventory and impact assessment values that are independent from geography (i.e., generic values for United States conditions). Then a life-cycle assessment was performed for the same functional unit, but with geographic differentiation taken into account. Finally, life-cycle assessment results were generated considering allocation of parts of the impacts from the coal-fired power plant to the fly ash.

Goal and scope

The functional unit is the disposal of one tonne of fly ash in the United States. The main results of this chapter apply to fly ash as a waste produced by coal-fired power plants. In that case, no impacts from the coal-fired power plant are allocated to fly ash. A sensitivity analysis is performed and the consequences discussed for the case where fly ash is considered as a co-

product having a commercial value and being treated by the coal-fired electricity industry as a source of income. In that case, based on ISO 14044 (ISO 2006b), economic allocation should be used to allocate part of the impacts of the coal-fired power plant to the fly ash co-product. The burdens and benefits of disposing of the entire United States fly ash production for different alternatives is also presented. The different alternatives to dispose of or use fly ash studied in this chapter are: 1) landfilling or lagooning, 2) cement production and/or concrete products, 3) structural fills or embankments, 4) stabilization of waste materials, 5) road base or subbase materials, 6) flowable fill and grouting mixes, and 7) mineral filler in asphalt paving. The rate of substitution typically specified is a minimum of 1 to 1 — one kg of fly ash to one kg of cement (USDOT 2009). The system definition and boundaries for these different alternatives are presented in Figure 9-1. The net impacts of the different alternatives should identify the more environmentally suitable alternatives.

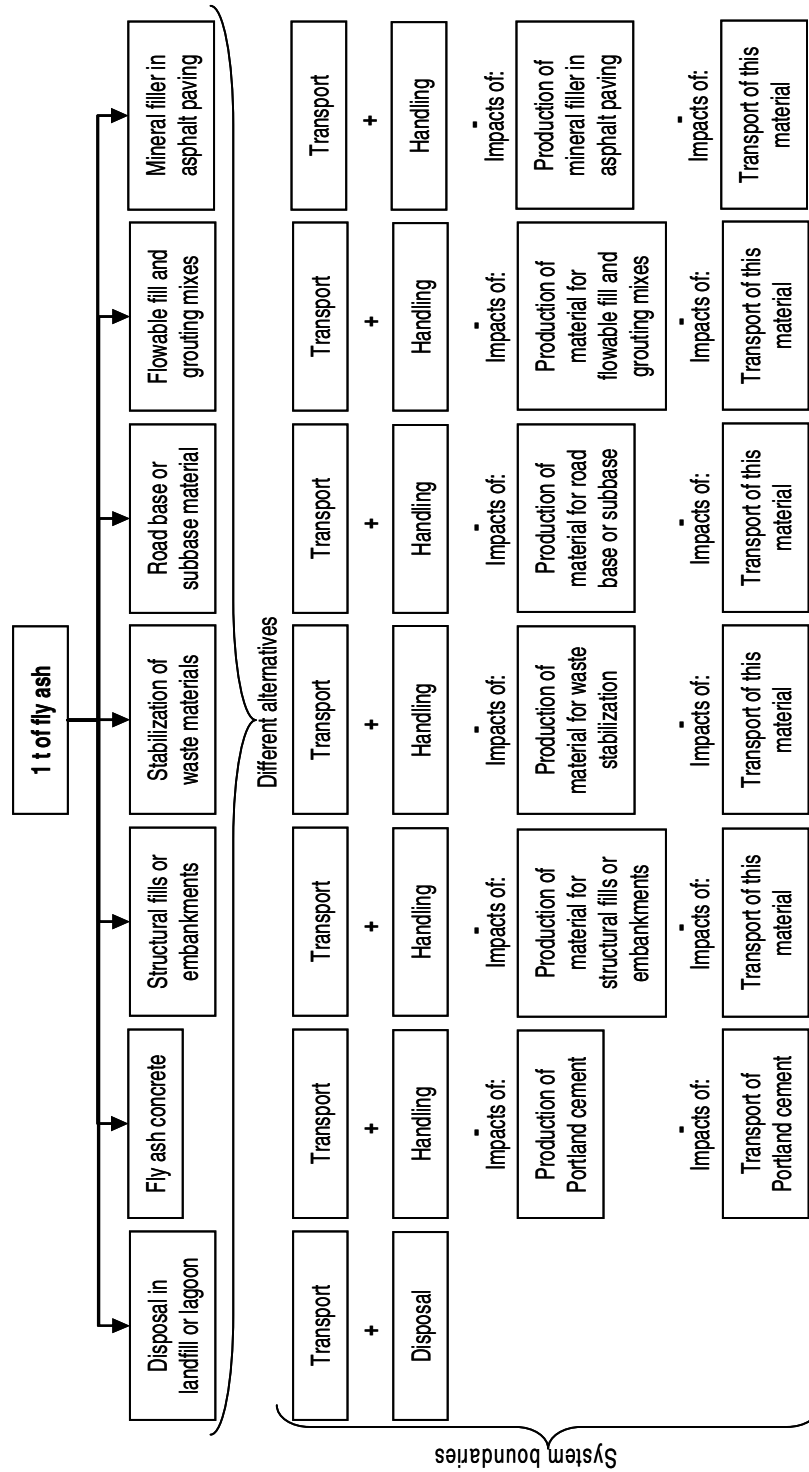


Figure 9-1: Alternatives for the disposal or use of fly ash.

Reference flows

The reference flows (ISO 2006a, 2006b, section 1.1.1) used in this chapter are shown in Table 9-2. The last column indicates assumptions that differ fromecoinvent regarding the “sub-compartment” (or “archetype”) where emissions of pollutants occur. The density of fly ash during transportation is assumed to be 2,500 kg/m³ (ProAsh 2008).

Table 9-2: Reference flows used in this chapter.

Phase of the life cycle	Flow	Value	Unit	Source and Comment
Disposal in landfill or lagoon				
Transport of fly ash	Truck 40t	50	km	The landfill and lagoon is assumed to be close to the power plant. <i>Regionalization:</i> Assumed to be in a low population density area.
Disposal of fly ash	Inert material landfill, infrastructure	1	t	(i.e., construction and closure). <i>Regionalization:</i> Assumed to be in a low population density area.
	Inert material landfill, operation	1	t	Dominated by diesel consumption (0.75 l/t). <i>Regionalization:</i> Assumed to be in a low population density area.
Fly ash concrete				
Transport of fly ash	Truck 40t	50	km	
	Train	1000	km	This distance can vary a lot. <i>Regionalization:</i> Assumed to be 90% in a low and 10% in a high population density area.
	Truck 40t	100	km	<i>Regionalization:</i> Assumed to be in a high population density area.
Handling of fly ash	Front loader	2	t	Transfer from truck to train and from train to truck. <i>Regionalization:</i> Assumed to be in a low population density area.
Portland cement	Portland cement	-1	t	
Transport of Portland cement	Truck 40t	-100	km	This distance can vary a lot.
Structural fills or embankments				
Transport of fly ash	Truck 40t	50	km	
	Train	1000	km	This distance can vary a lot. <i>Regionalization:</i> Assumed to be 90% in a low and 10% in a high population density area.
	Truck 40t	100	km	
Handling of fly ash	Front Loader	2	t	Transfer from truck to train and from train to truck. <i>Regionalization:</i> Assumed to be in a low population density area.
Structural fill material	Crushed gravel	-1	t	
Transport of structural fill material	Truck 40t	-100	km	This distance can vary a lot.
Stabilization of waste materials				
Transport of fly ash	Truck 40t	50	km	
	Train	1000	km	This distance can vary a lot. <i>Regionalization:</i> Assumed to be 90% in a low and 10% in a high population density area.
	Truck 40t	100	km	<i>Regionalization:</i> Assumed to be in a low population density area.
Handling of fly ash	Front Loader	2	t	Transfer from truck to train and from train to truck. <i>Regionalization:</i> Assumed to be in a low population density area.
Portland cement	Portland cement	-1	t	If is estimated that the fly ash is replacing Portland cement as stabilizing material for waste materials (especially for ashes or slag from municipal incinerator).
Transport of Portland cement	Truck 40t	-100	km	This distance can vary a lot.
Road base or subbase materials				
Transport of fly ash	Truck 40t	50	km	
	Train	1000	km	This distance can vary a lot. <i>Regionalization:</i> Assumed to be 90% in a low and 10% in a high population density area.
	Truck 40t	100	km	
Handling of fly ash	Front Loader	2	t	Transfer from truck to train and from train to truck. <i>Regionalization:</i> Assumed to be in a low population density area.
Production of road base or subbase material	Crushed gravel	-1	t	

Phase of the life cycle	Flow	Value	Unit	Source and Comment
Transport of road base or subbase material	Truck 40t	-100	km	This distance can vary a lot.
Flowable fill and grouting mixes				
Transport of fly ash	Truck 40t	50	km	This distance can vary a lot. <i>Regionalization:</i> Assumed to be 90% in a low and 10% in a high population density area.
	Train	1000	km	
	Truck 40t	100	km	
Handling of fly ash	Front Loader	2	t	Transfer from truck to train and from train to truck. <i>Regionalization:</i> Assumed to be in a low population density area.
Material for flowable fill and grouting mixes	Sand	-1	t	
Transport of material for flowable fill and grouting mixes	Truck 40t	-100	km	This distance can vary a lot.
Mineral filler in asphalt paving				
Transport of fly ash	Truck 40t	50	km	This distance can vary a lot. <i>Regionalization:</i> Assumed to be 90% in a low and 10% in a high population density area.
	Train	1000	km	
	Truck 40t	100	km	
Handling of fly ash	Front Loader	2	t	Transfer from truck to train and from train to truck. <i>Regionalization:</i> Assumed to be in a low population density area.
Mineral filler in asphalt paving	Sand	-1	t	
Transport of mineral filler in asphalt paving	Truck 40t	-100	km	This distance can vary a lot.
<i>Note:</i> Details about distances: The distance of 1000 km by train is taken as a mid value between the two extreme case of (a) no use of train (i.e., 0 km) and (b) 2000 km by train - which correspond approximately to the distance to go from the center of the United States to a coast. The distance of 50 km for the disposal of fly ash in a landfill is based on the rounded distance between the Battlefield Golf Club fly ash landfill, in Centerville, VA, and the Dominion Virginia Power coal-burning plant in Deep Creek, VA. The distance of 50 km for the distance between the coal-fired power plant and the train station is taken equal to 50 km too to be consistent with the distance assumed to the landfill. This avoids implementing variation caused by assumptions and methodological issues. The distance of 100 km for the distance from the train to the location where the fly ash is used (concrete plant, etc.) is based on the one order of magnitude rounded distance between the Stockton, CA rail yard and San Francisco, CA. The distance of 100 km for the distance to deliver Portland cement is based on the one order of magnitude rounded distance between the Cupertino, CA cement plant and San Francisco, CA. The distances to deliver other materials are also taken as equal to 100 km to be consistent with the 100 km taken for the cement plant. This avoids implementing variation caused by assumptions and methodological issues.				

Heidrich et al. (2005) estimated that collection of fly ash at the power stations results in emissions of 0.006 kg CO_{2-eq} per tonne of fly ash. However, fly ash will be produced at the power stations for air quality reasons, and thus collected regardless of whether it is used in concrete, and so this impact value should not be included in the fly ash emissions factor (O'Brien et al. 2009).

Life-cycle inventory

Life-cycle inventory is mainly based on the ecoinvent database (Frischknecht 2005) adapted to North America for the electricity mix used in the different sub-processes. Inventory data are compiled using ecoinvent, the available literature and using information gathered directly from the parties active in the fly ash concrete life-cycle. The emissions of the different pollutants (especially PM_{2.5}, PM_{10-2.5}, NO_x and SO₂) are allocated to zones with different population density (i.e., high, low and unknown population density zone) to facilitate the evaluation of regionalization on the final results.

The foreground processes are the processes that are the focus of our analysis – usually those that are directly affected by any decision based on a life-cycle assessment study. The background processes represent other economic activities that provide materials or energy to the foreground processes (Sikdar and Diwekar 1999). The process “train, electricity, average load” as well as the electricity mixes other than the one for North America are used in the sensitivity

analysis considering the effect of regionalization (see below). The default electricity mix used in the different processes is the one for North America (United States, Canada and Mexico). Transportation distances can vary greatly depending on the specific case studied. If the results of the present chapter are used to analyze an alternative where transportation distances would differ significantly from the distances used in the present chapter, results of the present chapter need to be adapted. The inventory database ecoinvent (Frischknecht 2005) allows one to differentiate, in a convenient way, among emissions in different sub-compartments (i.e., emissions in high, low or unknown population density areas).

To increase the confidence in the data used for truck and train transportation, data provided by ecoinvent are compared to the data reported by Facanha and Horvath (2006) for PM₁₀ and NO_x emissions. The absolute emission factors, in g/(t·km), reported by Facanha and Horvath (2006), both for tailpipe emissions and for life cycle emissions, are on average half the factors reported by ecoinvent. The ratios of tailpipe emissions over life-cycle emissions reported by Facanha and Horvath (2006) represent between 75% and 110% of the value reported by ecoinvent. Because the values of Facanha and Horvath (2006) are consistently lower than the values of ecoinvent, a change in the values will not affect the ranking among the scenarios. Furthermore, considering that the difference among the scenarios providing high and low environmental benefits is significantly higher than a factor of two (see Figure 9-2 below), the use of the data from Facanha and Horvath (2006) instead of ecoinvent would not change the ranking among the scenarios. For consistency among the sources of data and especially because the data reported by Facanha and Horvath (2006) do not differentiate between PM_{2.5} and PM_{10-2.5}, I decided to use the data reported by ecoinvent in the main analysis. Indeed, in Chapter 8 I report characterization factors for PM_{10-2.5} two orders of magnitude lower than those for PM_{2.5}, making the distinction between PM_{2.5} and PM_{10-2.5} of importance in the evaluation of the damage to human health.

Life-cycle impact assessment

The life-cycle impact assessment used to evaluate the life-cycle inventory is a combination of the results found in Chapter 4 for respiratory impacts associated with inorganics, and IMPACT 2002+ (Jolliet et al. 2003, Humbert et al. 2009) for the other impact categories related to human health. Regionalization is considered in the impact assessment. Results are presented for the damage categories human health, ecosystems quality, climate change, and resources. The evaluation of the life-cycle inventory is performed using the software SimaPro (PRé 2006). Damage to human health considers all emissions other than the fly ash dust itself. The assessment of the potential damage to human health associated with occupational exposure to fly ash dust during handling of fly ash, or during fly ash concrete crushing, are outside the scope of this chapter but should be also considered in further studies.

Impact assessment scores for regionalization consider damage to human health from toxicity and respiratory effects from inorganics. Regionalization follows the method developed in Chapter 4 and is discussed in more detail in the next sections.

Unit processes

A list of the processes used in this chapter as “foreground” processes, as well as the different electricity mixes used as “background” processes for all these unit processes is shown in Table 9-3.

Table 9-3: Impact score of the different unit processes used in this chapter.

Unit processes (life-cycle inventory taken from ecoinvent v1.3 (Frischknecht 2005) with electricity mix adapted to North America)	Unit (“per ...”)	Human health (DALY)		Eco-system quality (PDF· m ² ·y) ^a	Climate change (kg CO ₂ -eq) ^a	Resources (MJ primary non- renewable) ^a	Type of regiona- lization (for operation)
		without regiona- lization	with regiona- lization				
Truck 40t, average load	t·km	1.9E-07	6.8E-08	0.080	0.16	2.8	low pop
Truck 40t, average load	t·km	1.9E-07	4.1E-07	0.080	0.16	2.8	high pop
Truck 40t, average load	t·km	1.9E-07	1.8E-07	0.080	0.16	2.8	-
Train, diesel, average load	t·km	7.8E-08	3.5E-08	0.011	0.05	0.7	90% low 10% high
Train, electric, average load (with North American mix)	t·km	2.9E-08	2.2E-08	0.006	0.04	0.7	-
Inert material landfill, infrastructure	t	5.7E-06	2.7E-06	2.100	4.60	170.0	low pop
Inert material landfill, operation	t	4.8E-06	6.5E-07	0.390	2.60	39.0	low pop
Loader	m ³	9.9E-07	1.3E-07	0.078	0.51	7.7	low pop
Portland cement, at plant	t	1.9E-04	1.6E-04	34.000	762.00	3260.0	-
included released from clinker:	t				587.00		
Gravel, crushed, at mine	t	5.0E-06	2.7E-06	1.200	4.25	120.0	low pop
Sand, at mine	t	3.5E-06	1.2E-06	0.620	2.40	52.0	low pop
Electricity (North America mix), at grid	kWh	3.5E-07	2.0E-07	0.160	0.68	11.00	-
Electricity (United States mix), at grid	kWh	3.7E-07	1.9E-07	0.160	0.73	12.0	replacing North America mix for sensitivity study
Electricity (Canada mix), at grid	kWh	1.7E-07	1.2E-07	0.120	0.29	5.5	
Electricity (Mexico mix), at grid	kWh	3.8E-07	6.9E-07	0.150	0.71	12.0	
Electricity (UCTE (i.e., Western Europe) mix), at grid	kWh	3.1E-07	2.4E-07	0.130	0.51	12.0	
Electricity (California mix), at grid	kWh	1.2E-07	2.1E-07	0.110	0.39	9.6	
Electricity (natural gas), at grid	kWh	1.2E-07	2.3E-07 ^b	0.105	0.68	13.0	
Electricity (coal), at grid	kWh	6.0E-07	1.1E-07 ^b	0.210	1.10	13.0	
Electricity (hydro), at grid	kWh	2.0E-08	1.9E-08	0.090	0.01	0.1	

^a With IMPACT 2002+ (Jolliet et al. 2003), 100-year time horizon; ^b Note that the results for coal electricity are surprisingly lower than electricity from other sources of energy. This is mainly a result of the fact that, in ecoinvent v1.3 (Frischknecht 2005), on a per kWh basis, electricity produced from natural gas has higher impacts on human health than electricity produced from coal. The main source of impacts from natural gas-based electricity is caused by SO₂ and aromatic compounds emitted at the extraction. The impact caused by SO₂ and aromatic compounds emitted at the extraction is higher than the impacts caused by stack emissions of the coal-power plant. This surprising result calls for more research to identify (i) whether the life-cycle inventory is accurate, and (ii) improvement of regionalization scheme within life-cycle inventory database. Indeed, currently, in ecoinvent, it is not possible to qualify an emission of remote, but only rural, and therefore stack emissions of coal power plant have the same characterization factor as emissions during extraction of natural gas. Should the natural gas extraction be in remote location (e.g., ocean), the contribution of natural gas extraction to human health damage would be overestimated. Further analysis of the database would be needed to strengthen the results presented in Table 9-3 and used in this chapter.

Each of the unit process of Table 9-3 represents the impacts associated with the different reference flows of Table 9-2. The impacts per unit process of Table 9-3 are multiplied with the reference flows of Table 9-2 to obtain the impact scores presented in the results.

Table 9-4 shows an example of detailed computation to obtain the climate change score (Figure 9-4) for the scenario of using fly ash in concrete (second bar in Figure 9-4).

Table 9-4: Example of detailed computation to generate the impact score.

Life cycle stage	Reference flows (Table 9-2) (per tonne of fly ash)	Impact score of the unit process (Table 9-3) (kg CO _{2-eq})	Damage score (multiplication of the reference flows column and the unit process impact score column) (kg CO _{2-eq} per tonne of fly ash)
Truck (50 km)	1 t × 50 km = 50 t·km	0.16 per t·km	8.0
Train (1000 km)	1 t × 1000 km = 1000 t·km	0.05 per t·km	50
Truck (100 km)	1 t × 100 km = 100 t·km	0.16 per t·km	16
Loader	2 t	0.051 per t	0.10
Landfill, infra.	-		
Landfill, operation	-		
Portland cement	-1 t	(762-587) = 175 per t	-175
Released from clinker	-1 t	587 per t	-587
Crushed gravel	-		
Sand	-		
Truck (100 km)	1 t × -100 km = -100 t·km	0.16 per t·km	-16
Net impact			-704

The same approach is used to generate the results for all other damage scores (bars in the different figures) presented in this chapter.

9.4. Results and discussion

The reference flows of Table 9-2 are multiplied with the impacts per unit process of Table 9-3 to obtain the impact score indicated Figure 9-2, Figure 9-3, Figure 9-4 and Figure 9-5.

Impact assessment results without regionalization

The burdens and benefits of the disposal of one tonne of fly ash are shown for human health (in DALY, Figure 9-2), ecosystem quality (in PDF·m²·y, Figure 9-3), climate change (in kg CO_{2-eq}, Figure 9-4) and primary non-renewable energy (resource) (upper heating value, in MJ, Figure 9-5).

The disability adjusted life years metric (DALY — Murray and Lopez 1996) measures the damage to human health associated with pollutants emitted to the environment, accounting for both mortality (years of life lost due to premature death) and morbidity. The PDF·m²·y, potentially disappeared fraction (PDF) of species over a certain area during a certain time, is a measure of the damage to ecosystem quality. The CO_{2-eq} is a measure of the greenhouse potential of different greenhouse gases, based on the IPCC Global Warming Potentials for a 100-year time horizon (IPCC 2007). The unit for damage to resources measures the amount of primary energy (upper heating value, in MJ — Jolliet et al. 2003) that is non-renewable (i.e., from fossil energy as well as from nuclear energy, but not from renewable energy) and extracted from the ground. More explanation of these units can be found in Jolliet et al. (2003) and Humbert et al. (2009).

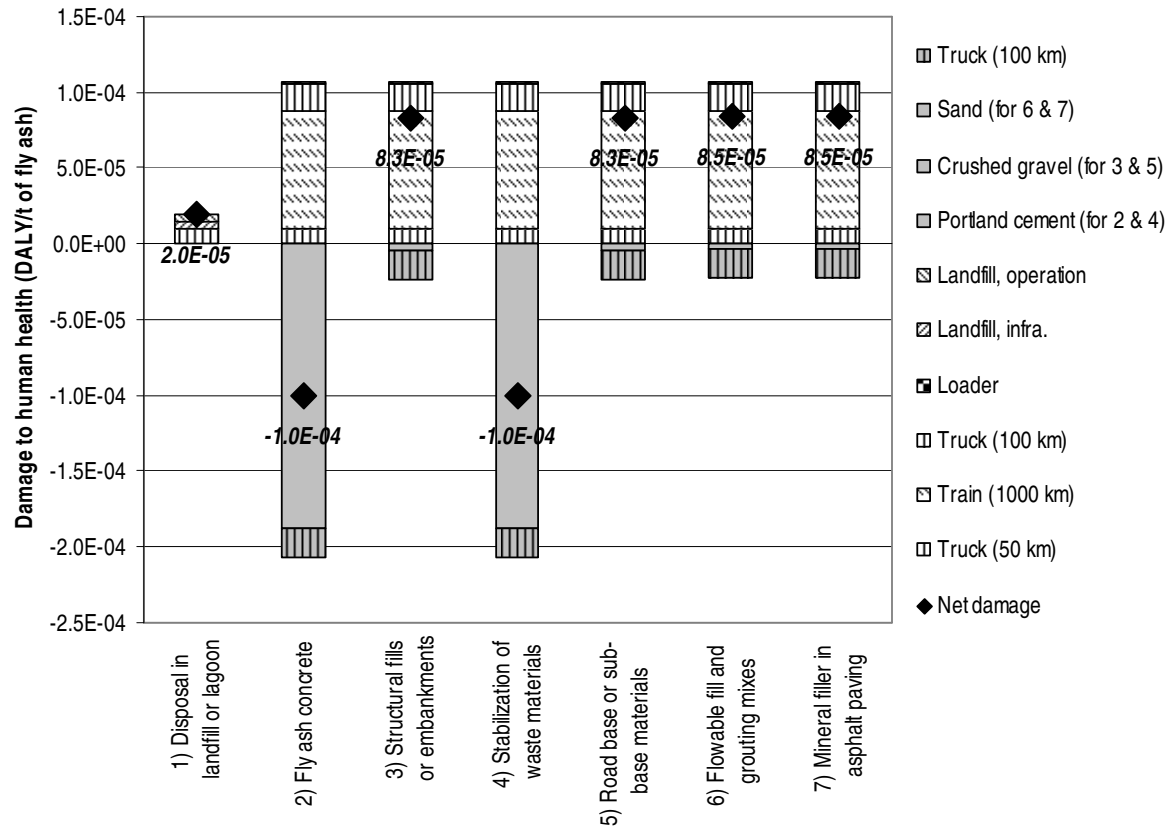


Figure 9-2: Damage to human health (other than from fly ash dust) for the disposal of one tonne of fly ash (net damages are given with the dot and associated value).

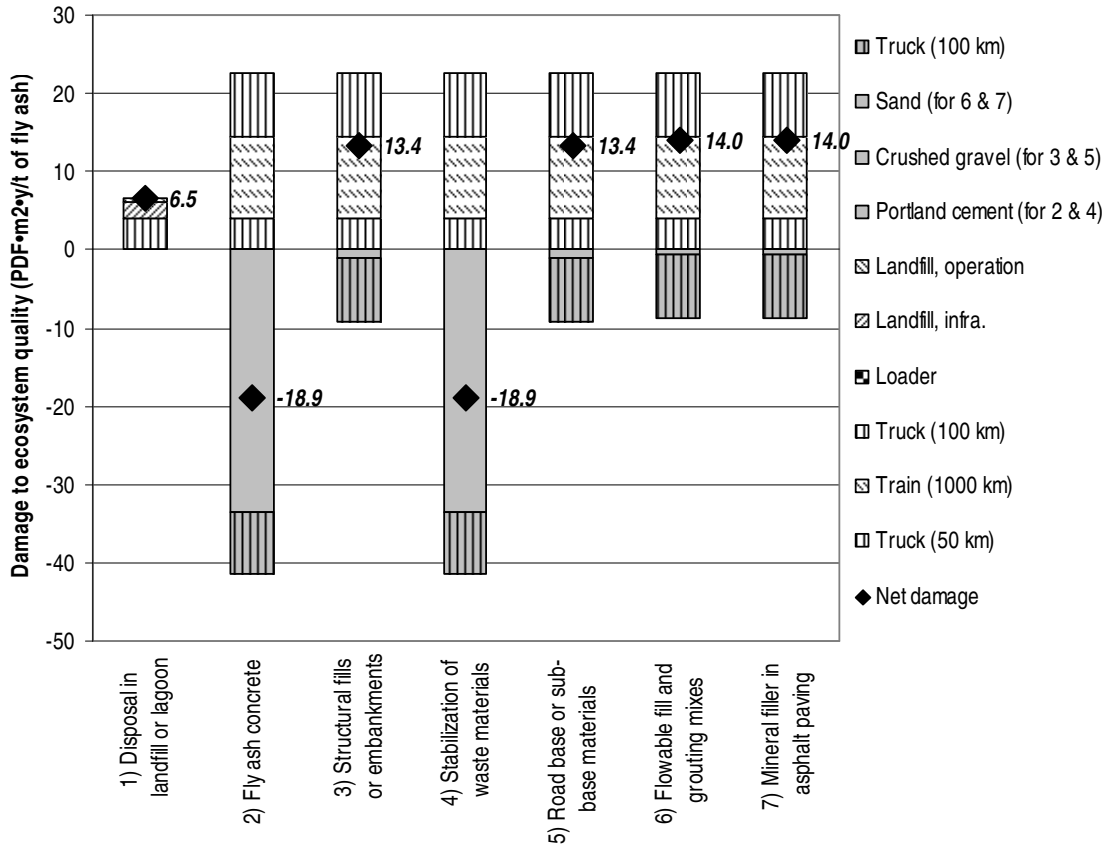


Figure 9-3: Damage to ecosystem quality for the disposal of one tonne of fly ash (net damages are given with the dot and associated value).

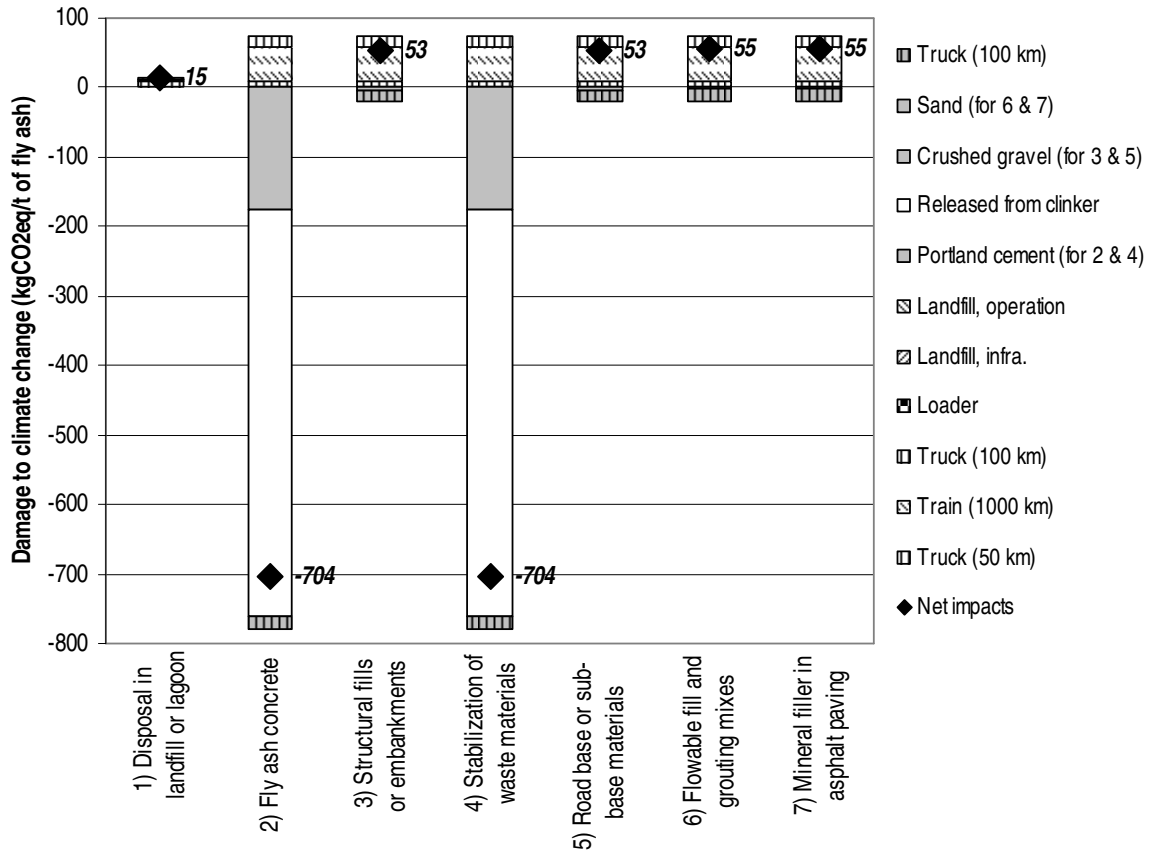


Figure 9-4: Damage to climate for the disposal of one tonne of fly ash (net damages are given with the dot and associated value).

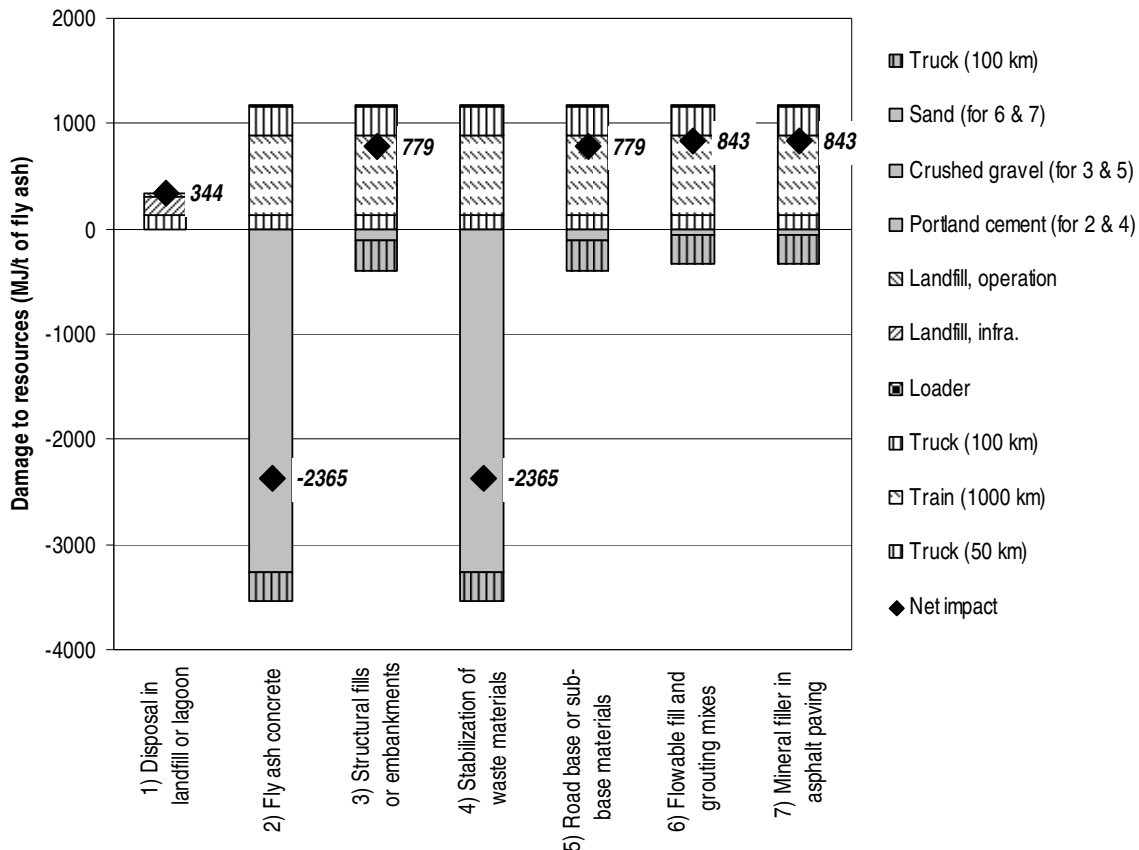


Figure 9-5: Damage to resources for the disposal of one tonne of fly ash (net damages are given with the dot and associated value).

Discussion

The four damage indicators show approximately the same trend. It appears that practices that enable the displacement of Portland cement offer significant advantages over any other use because of avoided emissions. Indeed, the production of Portland cement is very energy intensive (Table 9-3). When fly ash is used instead of being disposed in a landfill or lagoon, it avoids the impacts of disposing this fly ash in this landfill or lagoon and the impacts of the material it replaces. For the use to be a better alternative than the disposal in landfill or lagoon, the net impacts of the use (i.e., burdens minus benefits, represented by a dot with its value on Figure 9-2, Figure 9-3, Figure 9-4 and Figure 9-5) should be lower than the impacts caused by the disposal in landfill or lagoon.

Significance of transportation

It appears that apart from the case where fly ash displaces Portland cement, the burdens from transportation outweigh the benefits associated with the displacement of materials that fly ash displaces as soon as the additional transportation requirements exceed 50 km by truck or 200 km by train (see Figure 9-2, Figure 9-3, Figure 9-4 and Figure 9-5). Thus fly ash should be, as much as possible, used to displace Portland cement. If, for technical reasons, fly ash cannot be used to displace Portland cement but only low-grade materials (i.e., inert materials) like crushed rock or sand, then it should not be transported more than approximately 50 km by truck (or 200 km by train) further than a landfill or lagoon for fly ash, or from where the low-grade material originates, whichever distance is the longest. Indeed, if the fly ash needs to travel a longer

distance, it becomes environmentally advantageous to simply dispose of it in the landfill or the lagoon and not displace a low-grade material. Depending on the damage used as an indicator, it appears that it is environmentally advantageous to use fly ash to displace cement as long as the transportation does not exceed 400 to 5,000 km by truck or 2,000 to 20,000 km by train, with damage to ecosystem quality and climate change giving, respectively, the shorter and longer break-even distances (see Figure 9-2, Figure 9-3, Figure 9-4 and Figure 9-5). These distances are the distances that are traveled in addition to the distance that the displaced cement would have traveled. Thus, the use of fly ash to displace cement is environmentally advantageous if used regionally when transported only by truck, and used wherever possible nationally, as long as it is transported by train.

Impact assessment results with regionalization

Human health and ecosystems quality

As indicated in Table 9-3 using regionalization tends to reduce the damage to human health by a factor of two to ten for all processes except for transportation by truck in a zone of high population density, and electricity production in Mexico, in California and from natural gas. The fact that regionalization tends to reduce the damage to human health for most processes is correlated with the fact that I decided to have the processes actually occur in low population density areas to represent the fact that fly ash is transported over medium to long distances and thus in low population density areas. I also assumed that power plants and cement plants are situated outside urban areas. For truck transport in an unknown population density area, no significant difference is observed since these processes can occur as much in high as in low population density areas. Thus, no specific distinction is made for direct emissions. For trucks driving in high population density areas, damage to human health is approximately a factor of two higher when considering regionalization. The results for damage to human health without and with using regionalization are presented in Figure 9-6.

The reference flows of Table 9-2 are multiplied with the impacts per unit process of Table 9-3 for “with regionalization” to obtain the impact score shown in Figure 9-6.

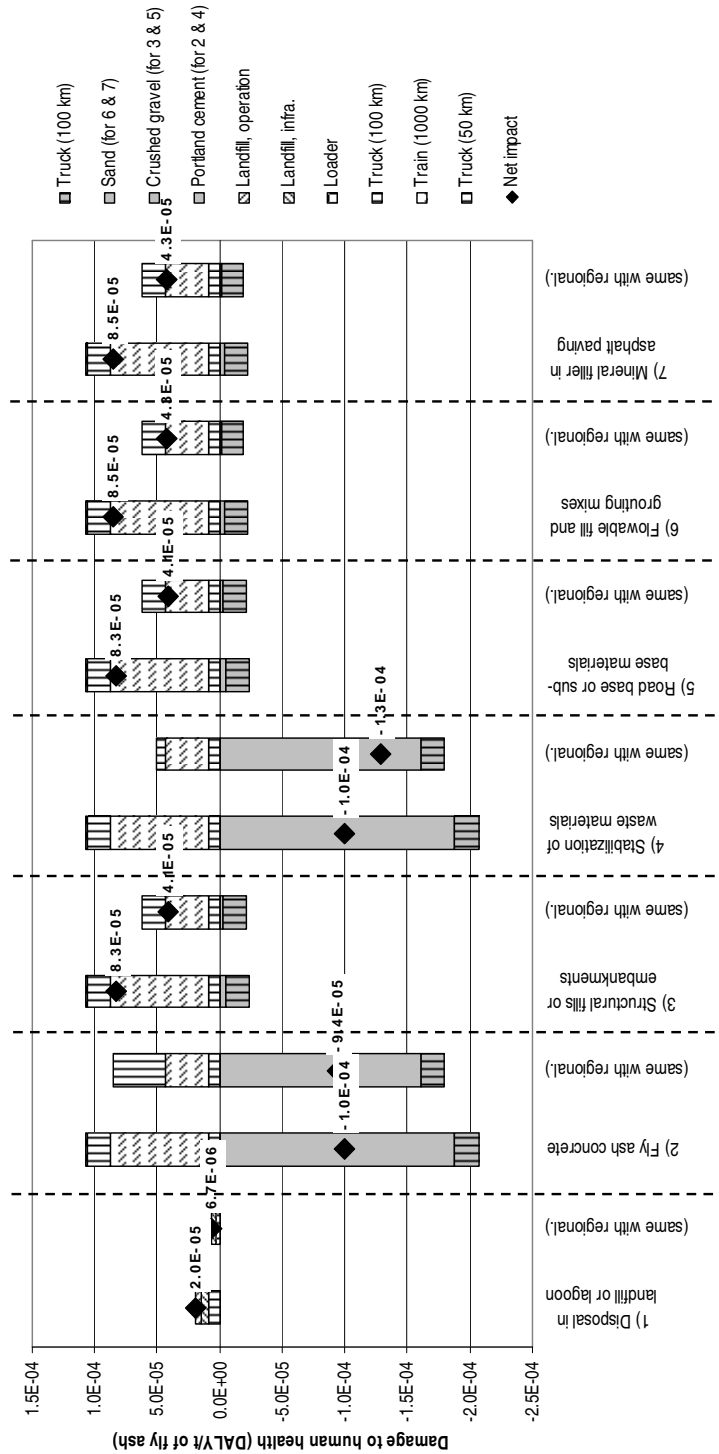


Figure 9-6: Damage to human health (other than from fly ash dust), without and with using regionalization.

Regionalization reduces overall impacts: indeed, most of the processes are situated in areas of low population density. Regionalization reduces results dominated by transportation by a factor of two (Figure 9-6). However, results dominated by Portland cement production do not follow the same trend because of the dominance from credits from the Portland cement

production avoided. Using damage to human health as an indicator, when performing regionalization, the break-even distance, required to keep Portland cement displaced by fly ash to be an advantageous use, is reduced by approximately 15% (from 950 km to 800 km)⁷ when done by truck, but is increased by approximately 80% (from 2,000 km to 3,700 km) when done by train (calculated in both cases without and with regionalization, respectively). However, these values vary greatly depending on the assumption for the type of population density pattern where foreground processes take place. Considering the range in the characterization factors of particles calculated in Chapters 4 and 8, differences of less than a factor of two should not be considered significant.

Climate change and resources consumption

Regionalization does not change the impact assessment related to climate change and resources consumption as these damage categories are global. However, regionalization leads to a change in the energy mix, energy efficiency, transportation distances or transportation mode, thus modifying the inventory of greenhouse gases emissions and energy use. Therefore, regionalization can change the overall results for climate change and resources consumption through a change in the inventory instead of through a change in the impact assessment.

Results presented in this chapter are not intended to reflect specific conditions of any fly ash producer or transporter but an average United States situation. If one wants to adapt these results to specific conditions, attention should be paid to transportation distances, the energy efficiency of transportation modes, and cement, gravel or sand production. Concerning the energy mix, and especially the one used for the electricity mix, several philosophies exist to tackle this issue; a discussion about which to adopt is outside of the scope of this chapter (e.g., whether one should use regional electricity mixes or not). This chapter follows the approach that, since electricity grids are, or could be, integrated, only the overall North American electricity mix matters and hence is used in the analysis.

However, as a sensitivity study, the results of Figure 9-4 are reproduced using different regional mixes as well as with a different mix of fuels and with different modes of transportation presented in Table 9-5. Table 9-5 presents the impact score results for the reference flows (or unit process) used in the sensitivity analysis.

This calculation is performed with SimaPro (PRé 2007). This software permits one to change the background electricity mix to all ecoinvent (Frischknecht 2005) processes at once.

Table 9-5: Damage to climate (in kg CO_{2-eq} per unit), using different electricity mix for all background processes.

Damage to climate, using the below mix for all the background processes:	Portland cement	Truck 40t, low pop	Truck 40t, high pop	Truck 40t, average pop	Train (diesel)	Inert material landfill, infrastructure	Inert material landfill, operation	Train, electricity, average load
Unit:	per t	per t-km	per t-km	per t-km	per t-km	per t	per t	per t-km
Electricity (North America mix)	7.6E+02	1.6E-01	1.6E-01	1.6E-01	4.9E-02	4.6E+00	2.5E+00	
Electricity (United States mix)	7.6E+02	1.6E-01	1.6E-01	1.6E-01	4.9E-02	4.6E+00	2.5E+00	
Electricity (Canada mix)	7.5E+02	1.6E-01	1.6E-01	1.6E-01	4.9E-02	4.6E+00	2.5E+00	
Electricity (Mexico mix)	7.6E+02	1.6E-01	1.6E-01	1.6E-01	4.9E-02	4.6E+00	2.5E+00	
Electricity (UCTE (i.e., Western Europe) mix)	7.6E+02	1.6E-01	1.6E-01	1.6E-01	4.9E-02	4.6E+00	2.6E+00	3.5E-02
Electricity (California mix)	7.5E+02	1.6E-01	1.6E-01	1.6E-01	4.9E-02	4.6E+00	2.5E+00	
Electricity (natural gas)	7.6E+02	1.6E-01	1.6E-01	1.6E-01	4.9E-02	4.6E+00	2.5E+00	
Electricity (coal)	7.8E+02	1.6E-01	1.6E-01	1.6E-01	4.9E-02	4.6E+00	2.5E+00	6.6E-02
Electricity (hydro)	7.4E+02	1.6E-01	1.6E-01	1.6E-01	4.9E-02	4.6E+00	2.5E+00	1.0E-02

⁷ These values are simply found by calculating how many km one can do to have the dot of Figure 9-6 reaching the value of disposal in landfill or lagoon, using the values in Table 9-3.

Figure 9-7 presents the results of this sensitivity analysis for the damage to climate. Figure 9-7 is generated in the same way as Figure 9-4, with the difference that unit processes of Table 9-5 are used to replace the corresponding unit processes of Table 9-3. Each bar in Figure 9-7 correspond to the results generated using one line of Table 9-5 — the first bar of each scenarios in Figure 9-7 is generated using the results of the first line in Table 9-5, etc.

One can observe that the electricity mix used does not change the absolute results significantly. The use of electric train instead of diesel train to transport fly ash over long distances reduces the overall impacts only if the electricity is considered to come mainly from hydro energy. The damage to climate for scenarios displacing Portland cement is not very sensitive to either the energy mix or to the transportation mode because it is dominated by the CO₂ released by the clinker itself. The relative results (i.e., which are the alternatives providing the highest environmental benefits) do not change significantly. Furthermore, considering the robustness of the results, calculated by default for the United States context but not changing significantly with the energy mix, it is possible to say that the results of this chapter are also valid for other regions of the world.

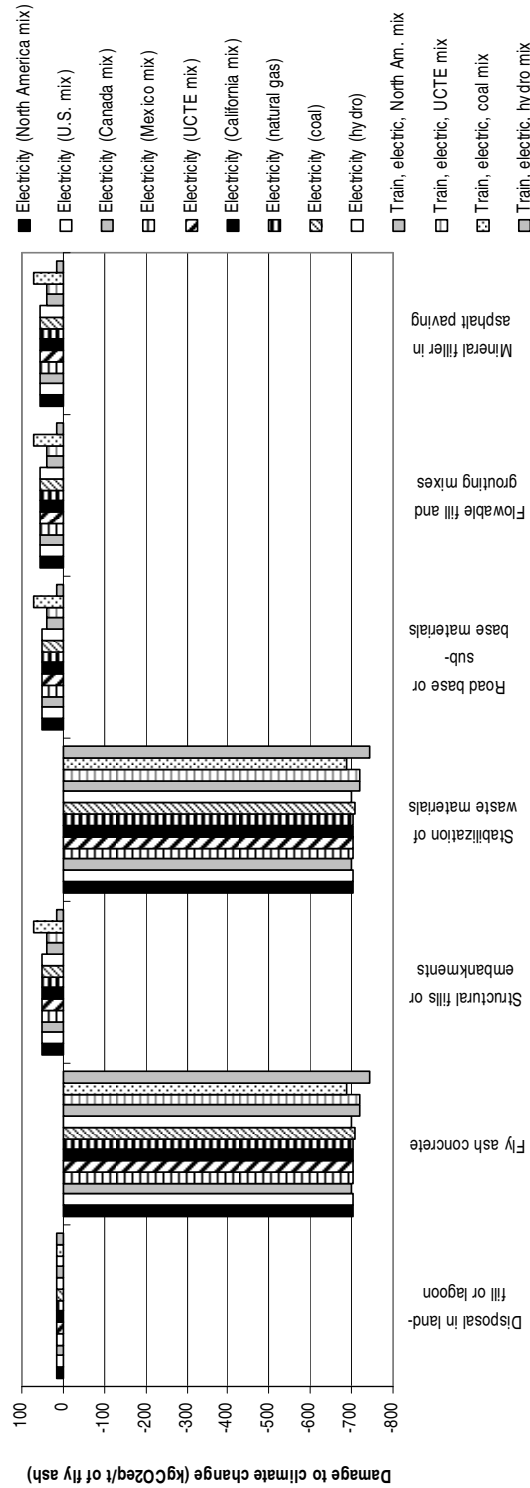


Figure 9-7: Damage to climate when using different electricity mixes as well as electric trains instead of diesel trains.

Because the difference among alternatives providing low and high benefits (represented in Figure 9-2 to Figure 9-5) is so significant, regionalization does not change the observations and ranking obtained without using regionalization.

Overall burden and benefit associated with United States fly ash production

Fly ash is often portrayed as a potential environmental problem when disposed of in landfills or lagoons, but actually has high potential benefits when used properly. This section aims at quantifying the overall potential greenhouse gas burdens and benefits associated with United States fly ash production. This section only evaluates greenhouse gases, but Figure 9-2 to Figure 9-5 show that other scores for damage categories are correlated with greenhouse gas emissions. Using the environmental burdens and benefits per tonne of fly ash calculated in this chapter, it is possible to extrapolate the results to the entire United States annual production of fly ash to get an idea of the overall potential for savings.

The annual United States greenhouse gas emissions are approximately 7 billion tonnes of CO_{2-eq} (2007 value – USEPA 2009). The annual production of fly ash is approximately 70 million tonnes. Thus, if all the fly ash is dumped into landfills or storage lagoons, (which represents an emissions burden of approximately 0.015 tonne CO_{2-eq} per tonne of fly ash (Figure 9-4)), the overall annual burden is approximately 1 million tonnes of CO_{2-eq}, which is less than 0.02% of United States annual greenhouse gas emissions. However, if the majority of the 70 million tonnes of fly ash produced annually is used to displace energy and CO₂-intensive materials, like Portland cement, the avoided CO_{2-eq} emissions can reach 0.7 tonne CO_{2-eq} per tonne of fly ash (Figure 9-4) which represents an annual saving of approximately 50 million tonnes of CO_{2-eq} or about 0.7% of United States annual greenhouse gas emissions (assuming transportation distances indicated in Table 9-2). Hence, while the disposal of fly ash does not appear, from a greenhouse gas perspective, to be an issue (i.e., 0.02% of United States greenhouse gas emissions), a wise use of fly ash has considerable potential to reduce overall United States greenhouse gas emissions (i.e., as much as 1% of the overall emissions).

Allocation of parts of the impacts from the coal-fired power plant to the fly ash co-product

In the coal-based electricity industry, fly ash is increasingly seen as a co-product and not as a waste. This co-product has an increasing market value⁸ and generates a new source of revenue for the coal-based electricity industry. In that case, common practice in life-cycle assessment is to allocate part of the impacts from the coal power plant to the fly ash co-product. Allocation of impacts to co-products is an important methodological issue (Guinée et al. 2004, Frischknecht 2000). Co-products and waste should be differentiated. If fly ash is considered as a waste (i.e., the basis for the results presented in this chapter), then no impacts from the coal-fired power plant should be allocated to the fly ash (ISO 2006b). However, because fly ash has a market value and provides an income to the coal-fired power plant, it can be considered as a co-product instead of as a waste. In that case, part of the impacts of the coal-fired power plant should be allocated to the fly ash. Two approaches can be distinguished: (a) the influence on the system boundary expansion (the economic allocation approach), and (b) the assessment of the consequences of fly ash valorization on the electricity production market (consequential life-cycle assessment approach). The economic allocation approach is commonly applied in the field of life-cycle assessment and is suggested by the ISO 14044 standard (ISO 2006b). Economic allocation is usually considered a good descriptor when distributing the burdens among co-products according to their responsibilities, in generating these burdens by assuming that price or revenues are a good descriptor of these responsibilities. The consequential approach aims at assessing the consequences of an economic gain when valorizing fly ash, by evaluating what is the maximum competitive advantage that the income from fly ash can provide to the coal-based electricity generation industry to keep the use of fly ash environmentally sound.

⁸ Price of fly ash on the market varies a lot. Prices around 40 USD/t are common (average prices from different sellers' websites).

Economic allocation

The detailed computations and data sources for the economic allocation are presented in Table 9-6.

Table 9-6: Detailed computations and data sources for the economic allocation.

Parameter	Value	Unit	Comment	Unit	Allocation equivalent
Fly ash price paid to the power plant	10	USD/t	Parameter to be varied		
Electricity price paid to the power plant	0.04	USD/kWh	United States average net income in 2006 is 0.013 USD/kWh (EEI 2007)		
Coal price paid by the power plant	35	USD/t of coal	(EIA 2008)		
Electricity production	2400	kWh/t of coal	German average (Frischknecht 2005)		
US coal consumption, in 2006	1,050,000,000	t/y	(EIA 2008)		
US fly ash production, in 2006	72,000,000	t/y	(ACAA 2007)		
Fly ash production	0.069	t/t of coal	United States ratio		
Amount of fly ash produced per kWh	0.029	kg fly ash/kWh			
Amount of coal needed to produce one tonne of fly ash	15	t of coal/t of fly ash			
Amount of kWh produced per tonne of fly ash	35,000	kWh/t of fly ash			
Income from electricity production	96	USD/t of coal			
Income from fly ash production	0.69	USD/t of coal	Fly ash price paid to the power plant (USD/t fly ash) multiplied by the fly ash production (t fly ash/t coal)		
Amount of cement produced per tonne of coal	5.9	t of cement/t of coal	Equivalent energy (assuming 19.1 MJ per kg of coal - Jolliet et al. 2003)		
Comparison between fly ash and Portland cement production:					
Parameter	Value	Unit	Value	Units	Allocation to be equivalent
Allocation factor (ratio between income from fly ash and from electricity production)	0.71%	Production of 1 tonne of Portland cement			
Allocated damage to human health per tonne of fly ash	0.00015	DALY/t of fly ash	0.00019	DALY/t of Portland cement	
Allocated damage to ecosystems quality per tonne of fly ash	30	PDF-m2-y/t of fly ash	33	PDF-m2-y/t of Portland cement	
Allocated damage to climate per tonne of fly ash	280	kgCO2/t of fly ash	760	kgCO2/t of Portland cement	
Allocated damage to resources per tonne of fly ash	3,300	MJ/t of fly ash	3,300	MJ/t of Portland cement	
Allocation factor	100%				
Allocated damage to human health per tonne of fly ash	0.020	DALY/t of fly ash	0.00019	DALY/t of Portland cement	0.92%
Allocated damage to ecosystems quality per tonne of fly ash	4,200	PDF-m2-y/t of fly ash	33	PDF-m2-y/t of Portland cement	0.79%
Allocated damage to climate per tonne of fly ash	39,000	kgCO2/t of fly ash	760	kgCO2/t of Portland cement	1.95%
Allocated damage to resources per tonne of fly ash	470,000	MJ/t of fly ash	3,300	MJ/t of Portland cement	0.70%

On average, in the United States, 0.07 tonne of fly ash is produced per tonne of coal burned. The gross income from electricity generation in 2006 was approximately 4 cents/kWh (the net income was 1.3 cents/kWh) (EEI 2007). Using this rate, the net income from electricity generation represents 100 USD per tonne of coal burned. On average, one tonne of fly ash is produced per 15 tonnes of coal burned, which produce 35,000 kWh, representing approximately 40,000 kg of CO_{2-eq} (using the value for electricity from coal-fired power plants of 1.12 kg of CO₂ per kWh shown in Table 9-3). Figure 9-8 presents the damage to climate, in kg of CO_{2-eq} per tonne of fly ash within the concrete, including both “real” impacts (mainly shipping) and allocated impacts (from the coal-fired power plant). Both Portland cement and fly ash are considered delivered to the concrete plant (i.e., using the delivery distances and mode indicated in Table 9-2 and impacts per unit processes indicated in Table 9-3).

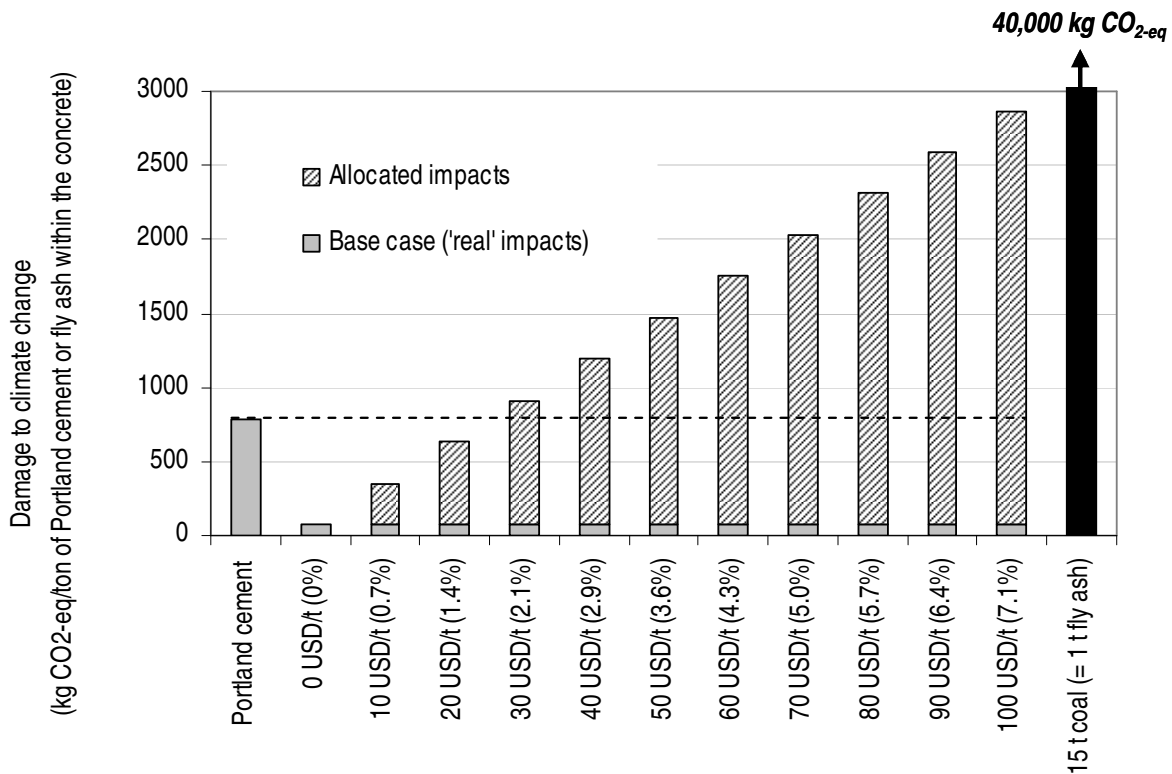


Figure 9-8: Damage to climate, in kg of CO_{2-eq} per tonne of fly ash within the concrete, including both “real” impacts and allocated impacts.

Using the damage scores presented in Figure 9-3 per kWh for coal-based electricity as well as per tonne of Portland cement production, environmental burdens from fly ash remain lower than the impacts of producing Portland cement only if the economic allocation factor remains less than 1% (or 2% if only greenhouse gas emissions are taken as the indicator) (Figure 9-8). This corresponds to a price paid to the power plant for fly ash below 10 USD per tonne of fly ash (25 USD if only CO_{2-eq} is taken as an indicator). Above these prices, when using economic allocation, concrete based on pure Portland cement is to be preferred to the one using fly ash. Note that the market price of Portland cement is significantly higher than 25 USD (for example, it was 124 USD per tonne in California in 2007 – PCA 2009).

Influence on the electricity production market

If fly ash is considered a co-product that drives the electricity industry to slightly increase the share of coal-based electricity generation, then, in a model for studying this large-scale change, the fly ash should carry this extra burden. Quantifying the influence of the fly ash market on the electricity market is beyond the scope of this chapter. However, it is possible to quantify the marginal increase in coal-based electricity generation that fly ash generated income could afford to have and not outweigh the potential environmental benefits from displacing Portland cement. The marginal increase can be expressed in amount of additional kWh produced, as a fraction of the current coal-based electricity production or as a fraction of the share of the electricity mix that is coming from coal. Detailed computation is presented in Table 9-7. The electricity replaced by coal is taken as the weighted average of the United States electricity mix (EIA 2008). If the switch for fly ash from a waste (that needs to be disposed of) to a co-product (that generates a certain income for the electricity industry) leads to an increase in the damage intensity (in damage/kWh) of the average electricity mix of United States by 2%, then the use of fly ash causes more environmental burdens than benefits (using CO_{2-eq} as indicator for the burdens). This corresponds to a 3% increase in the coal-based electricity generation, or 1.5% of the share of coal within the United States electricity mix.

Table 9-7: Evaluation of the benefits created by displacing Portland cement with fly ash compared to the damage from United States (U.S.) electricity production (total and only coal).

Electricity production	Value	Unit	Source		
U.S. electricity production	4.1E+12	kWh/an	(EIA 2008)		
U.S. coal electricity production	2.1E+12	kWh/an	(EIA 2008)		
U.S. fly ash production	7.2E+07	t/y	(ACAA 2007)		
	Human health (DALY)	Ecosystem quality (PDF·m ² ·y)	Climate change (kg CO _{2-eq})	Resources (MJ primary non-renewable)	Unit
Damage from U.S. electricity mix without coal (<i>using values from Table 9-3</i>) (weighted average of the 2006 U.S. non-coal electricity mix – EIA 2008)	2.6E-07	1.1E-01	3.0E-01	1.1E+01	per kWh
Damage from U.S. coal-based electricity (<i>using values from Table 9-3</i>)	1.1E-07	2.1E-01	1.1E+00	1.3E+01	per kWh
Additional damage from U.S. coal-based electricity compared to U.S. mix without coal (<i>difference between the two above</i>)	-1.5E-07	9.7E-02	8.2E-01	2.4E+00	per kWh
Damage from U.S. electricity production	7.6E+05	6.6E+11	3.0E+12	5.0E+13	per y
Damage from U.S. coal-based electricity production	2.4E+05	4.4E+11	2.4E+12	2.8E+13	per y
Benefits from displacing Portland cement with the U.S. fly ash production	1.2E+04	2.4E+09	5.5E+10	2.3E+11	per y
Fraction of the U.S. electricity production	1.5%	0.36%	1.9%	0.47%	
Fraction of the U.S. coal-based electricity production	4.9%	0.55%	2.3%	0.83%	
Number of coal kWh so that the extra damage from coal-based electricity equals damage from Portland cement displacement	-7.8E+10	2.5E+10	6.7E+10	9.8E+10	kWh/y
Fraction of the current coal-based electricity production	-3.7%	1.2%	3.2%	4.7%	

The results of this chapter are valid for cases where the use of fly ash does not significantly change the overall life or strength of the structure compared to the case with no fly ash use. If the life or strength of the structure happens to be reduced (or increased) by using fly ash, the present results should be modified accordingly.

9.5. Conclusion and recommendations

Fly ash should be, as much as possible, used to displace Portland cement. Indeed, if used to displace cement, fly ash is environmentally advantageous, if used regionally when transported

only by truck, and when used nationally, when transported by train. If fly ash is used to displace low-grade materials like crushed rocks or sand, then it should be used within a distance of 50 km (by truck) further than a landfill for fly ash, or from where the low-grade material originates, whichever is the furthest distance. Indeed, if the fly ash needs to be transported further, then it becomes more environmentally advantageous to simply dispose of it in the landfill and not displace a low-grade material.

The use of regionalized, instead of generic, values for the inventory and impact assessment changes the final results for damage to human health by no more than a factor of two, and does not change the damage to climate significantly. Regionalization or non-regionalization does not change the main conclusions drawn: the type of use for fly ash that is most advantageous is the same (i.e., to displace cement) and the approximate break-even distances of transport that can be afforded to keep the use of fly ash advantageous compared to disposal in landfill or lagoon are comparable (i.e., by less than a factor of two, in the order of 900 km by truck and 3,000 km by train, when using damage to human health as indicator). Furthermore, considering the robustness of these results, they can be considered as valid for other regions of the world.

This chapter also shows that when used to displace part of Portland cement, fly ash can provide several environmental benefits, especially for climate change. However, the economic system associated with fly ash should be set up in a way that will avoid any incentive to increase the coal-based share of the electricity mix. Indeed, even a small (less than 2%) allocation of the impacts from the coal-fired power plant to the fly ash production, or an increase by 3% of the coal-based electricity permitted by fly ash selling, will outweigh the benefits from using fly ash to displace Portland cement. To stay environmentally sound, the valorization of fly ash should be made in a way that does not increase the competitive advantage of coal-based electricity generation on the electricity market. A wise use of overall United States fly ash production can contribute to reducing the overall United States greenhouse gas reduction by as much as 1%.

Finally, because of lack of data it was not possible herein to quantify the potential additional energy and associated impacts required to prepare fly ash for shipping. Further research would be necessary to explore this issue. It is also important to notice that issues associated with occupational exposure to fly ash dust during the handling of fly ash or fly ash concrete crushing as well as potential issues related to heavy metals or radioactivity (Hvistendahl 2007) are outside the scope of this chapter. They should however also be considered and require further investigation.

9.6. Acknowledgments

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10. Regionalized life-cycle assessment of coal fly ash concrete

Chapter 9 identified that the best use of coal fly ash is to use it in concrete to replace Portland cement. In Chapter 10, I analyze coal fly ash concrete in more detail, with a special attention to the influence of regionalization.

10.1. Summary

Concrete is one of the most common man-made materials in the world. Most of its environmental impacts are due to the energy-intensive production of Portland cement. Using coal fly ash for a binder and substituting it for a certain fraction of the Portland cement can help reduce the environmental impacts of concrete. However, sometimes fly ash needs to be shipped over long distances while Portland cement is generally produced locally. The goal of this chapter is to use a regionalized impact assessment approach to quantify the burdens and benefits and identify the key parameters and trade-offs of using coal fly ash as a substitute for a certain fraction of Portland cement in concrete. This chapter focuses on the differential impacts on human health throughout the western United States.

Life-cycle assessment is used to evaluate fly ash concrete in San Francisco, California, with fly ash coming from the states of Washington and Colorado. The life-cycle inventory is a combination ofecoinvent, data specific to the United States cement production industry, and data gathered directly from a concrete batching plant (Bode Gravel in San Francisco). A geographically differentiated life-cycle impact assessment method is used for the impact categories related to human toxicity and respiratory effects caused by inorganics. Other impact categories are directly adapted from the IMPACT 2002+ method. The influence of regionalization on the results for human health damage is evaluated.

Ready-mix concrete is associated with 130 to 240 kg CO_{2-eq} per ton, depending on the fly ash content, the overall binder content, and the shipping distances for the coarse and fine aggregates. The impact of concrete is reduced almost linearly with the fly ash content: for each percent of Portland cement replaced by fly ash, the environmental impacts of concrete are reduced by 0.5% to 0.9%. Key parameters are the shipping distance and mode of transportation used for the supply of fly ash, and the location and the energy mix used for the production of Portland cement. Regionalization reduces by a factor of two to three the results obtained with generic values, mainly because of the net reduction of the impacts caused by boats' emissions during shipping of aggregates and slag, as well as diesel train emissions during the shipping of fly ash. In several cases, the damage to human health is the limiting factor when calculating the break-even distance between fly ash shipping and Portland cement production.

The use of fly ash as a substitution material for part of the Portland cement can reduce the environmental impact of concrete. However, CO_{2-eq} emissions are a poor indicator when looking at overall environmental impacts of concrete because damage to climate is the least limiting factor when evaluating break-even distances among potential alternatives to supply binder. When human health is the limiting factor for break-even distances of supply, the use of regionalization can increase the reliability of the results since results based on CO_{2-eq} emissions would over estimate the overall environmental impacts difference between Portland cement and fly ash based concrete.

10.2. *Introduction and objectives*

Concrete

Concrete is one of the most common materials in the world, with more than 15 billion tonnes used per year (Mehta and Monteiro 2006). Most of its environmental impacts are due to the very energy intensive production of Portland cement, responsible for 4% of the world's CO₂ emissions (IEA 2007). A concrete mix typically consists, by volume, of 10% to 15% binder (mainly Portland cement), 60% to 75% aggregate, 15% to 20% water and 5% to 8% air pockets (Marceau et al. 2002).

Cement

The most common binder used in concrete is Portland cement. In 2007, the world's cement production was 2.6 billion tonnes (USGS 2008). In 2006, the United States (U.S.) consumed 127 million tonnes of cement and produced 100 million tonnes (USGS 2008). A viable alternative binder to Portland cement is fly ash.

Fly ash and fly ash concrete

Coal fly ash, with a global rate of production of more than 500 million tonnes a year, constitutes one of the largest industrial waste products in the world (Feuerborn 2005). With proper quality control, large amounts of fly ash can be incorporated into concrete, either in the form of blended Portland cement or as mineral admixtures (Chapter 9). Typical amounts of fly ash in concrete range from 15% to 25% by weight of the total cementitious materials (Marceau et al. 2002), but as much as 50% can be used in so called “high volume fly ash concrete” (Crouch et al. 2007). The rate of substitution typically specified is a minimum of 1 to 1 — one kg of fly ash to 1 kg of cement (USDOT 2009).

The life-cycle assessment literature on concrete, cement, and fly ash is extensive (Nisbet et al. 2002, Aguirre González 2005, Parrott 2002, Young et al. 2002, Carroll et al. 1998, Theis and Gardner 1990, Mroueh et al. 2001, Nisbet et al. 2002, Carroll et al. 1998, Babbitt and Lindner 2007, 2008, O'Brien et al. 2009, Babbitt and Lindner 2007, 2008). However, rare are the studies considering the impacts associated with the shipping of fly ash, which in some cases is shipped over longer distances than cement, or the benefits from diverting fly ash from landfills. Finally, no life-cycle assessment has taken into consideration the influence of regionalization, especially for damage to human health, arising from the differences among locations of pollutant emissions. Only one geographically differentiated life-cycle assessment has been performed on the different alternatives for the disposal and use of fly ash, including the impacts avoided by diverting fly ash from landfills, and it is described in Chapter 9. Chapter 9 identified that the displacement of Portland cement provides a substantial environmental benefit. The key parameter when using fly ash is the transportation mode and distances over which the fly ash is shipped. These key parameters become important for cases when fly ash is not produced at the same location as its potential use in concrete. For example, in the United States, fly ash is mainly produced in the Midwest and the East. However, some states, like California, where fly ash is extensively used in concrete do not produce fly ash. Thus fly ash needs to be shipped from thousands of km away. Chapter 9 showed that another key parameter is whether or not some of the impacts of the coal power plant are allocated to the fly ash. In that case, results show that with economic allocation, the use of fly ash becomes as much “polluting” as the Portland cement for prices between 10 and 25 USD per tonne of fly ash.

The objectives of this chapter are to evaluate the influence of geographically differentiated emissions and related impacts associated with fly ash concrete, and to perform a

sensitivity study regarding distance and mode of transportation for cases, like California, where fly ash is not produced locally, but shipped over long distances.

10.3. Method

Goal and scope

The functional unit evaluated is one tonne of concrete at the ready-mix plant, ready to be delivered to the job site. Two cases are evaluated: a generic United States ready-mixed concrete plant, and a ready-mixed concrete plant situated in San Francisco, California. This specific plant is Bode Gravel and produces 1,000 yd³ per working day (approximately 765 m³/d). The aggregate comes from the Vancouver area, British Columbia, Canada, by barge. The Portland cement is produced by Hansen Cement and comes from Cupertino, California, by truck. The fly ash is of class C, shipped from the states of Washington and Colorado by rail, and then by truck from the rail yard to the concrete plant. When fly ash is used, no special additive needs to be used (Bode Gravel, San Francisco, personal communication, summer 2006). Figure 10-1 shows the system considered for the production of fly ash concrete. Each process contains the entire up-stream processes needed to produce this process. For example, the process Portland cement production also contains mining, transport, clinker production, energy production, etc.

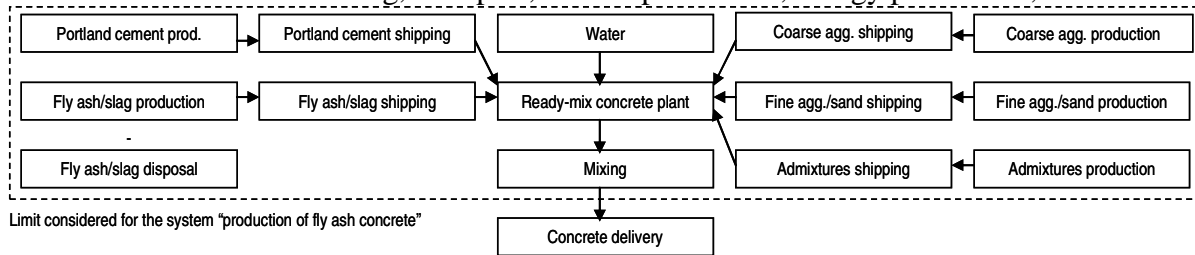


Figure 10-1: Production of fly ash concrete.

Life-cycle inventory

Data are gathered from the ecoinvent database (Frischknecht 2005) adapted to United States conditions by changing the background electricity mixes, taken from the available literature (e.g., USGS), and gathered directly from the manufacturers (Bode Gravel, San Francisco). Because of its regionalized inventory (i.e., it indicates whether emissions occur in an area of high or low population density), ecoinvent data are selected whenever possible. Close attention is paid to energy and CO₂, along with PM_{2.5}, PM_{10-2.5}, NO_x, and SO₂. Indeed, these pollutants appear to be the dominating elementary flows in the evaluation of the damage to human health (see Chapter 5, Joliet et al. 2006). Consideration is given to these pollutants to evaluate the influence of the location of emissions on final results. The impacts avoided (i.e., the impacts caused by landfilling or lagooning that will not occur because fly ash is diverted from disposal) are taken directly from Chapter 9.

The composition of the different concrete mixes studied is shown in Table 10-1. Composition for concrete with different amounts of fly ash is presented for typical concrete produced in a typical United States plant as well as at the Bode Gravel plant in San Francisco, California. Furthermore, five types of specific concrete produced by Bode Gravel are also presented. These five types of concrete are used as example of concrete produced in San Francisco, California and include a type of concrete that is sold as “green concrete” slag (a by-product from steel production). In 2006, 127 million tonnes of Portland cement were used in the United States, of which 100 million tonnes were manufactured in the United States and 27

million tonnes were imported (USGS 2008). The production of imported cement is assumed to have the same impacts as the cement produced in the United States. Only the transportation distances are adapted and increased. The two cases “0% fly ash” and “100% fly ash” represent the two extreme cases, in between which typical fly ash concrete is situated. Typical concrete on the market contains 15% fly ash (Bode Gravel, San Francisco, CA, personal communication). Concrete with 50% fly ash can be considered realistic for most of the applications and is the largest fraction that can be achieved on a large scale since annual fly ash production (70 million tonnes — ACAA 2007) represents approximately half of the annual use of binder (Portland cement and fly ash used in concrete) (147 million tonnes — ACAA 2007).

Table 10-1: Concrete composition used in this chapter (in kg per metric tonne of ready-mix concrete).

type of concrete:	Generic U.S. ready-mix concrete plant				Bode Gravel (San Francisco, CA) ready-mix concrete plant								
	typical concrete				typical concrete			specific concrete					
	0% fly ash	15% fly ash	50% fly ash	100% fly ash	0% fly ash	15% fly ash	100% fly ash	Basic (15% fly ash)	Slab (15% fly ash)	Shotcrete (16% fly ash)	Caltrans (25% fly ash)	“Green” (50% slag)	
Coarse aggregate	430	430	430	430									
Aggregate from Sechelt, Canada					430	430	430	396	448	224	433	445	
Sand (fine agg.)	320	320	320	320									
Sand from Sechelt, Canada					320	320	320	247	132	357	149	287	
Sand from Angel Island, California								174	188	171	210	0	
Portland cement	180	153	90		180	153		99	141	142	105	99	
Fly ash		27	90	180		27	180	17	25	27	35	0	
Slag												99	
Water	70	70	70	70	70	70	70	67	66	79	68	69	
Pozzoloth 200 N (admixture)	0.54	0.54	0.54	0.54	0.54	0.54	0.54	0	0.45	0.32	0	1.1	

Reference flows

In order to be able to evaluate the impact score associated with the different scenarios studied, the reference flows (ISO 2006a, 2006b, section 1.1.1) needed by the different scenarios to fulfill the functional unit need to be quantified. The reference flows used in this chapter are shown in Table 10-2. Each section corresponds to one line in Table 10-1.

Table 10-2: Reference flows used in this chapter.

Phase of the life cycle	Intermediary flow	Value	Unit	Source and Comment
Fly ash (1 t) (~2,500 kg/m³)				
Transport of fly ash (for generic U.S. ready-mix concrete plant)	Train	1,000	km	<i>Regionalization:</i> Assumed to be 90% in a low and 10% in a high population density area.
	Truck 40t	100	km	<i>Regionalization:</i> Assumed to be in a high population density area.
Transport of fly ash (to San Francisco, CA)	Train	2,000	km	From Washington and Colorado. <i>Regionalization:</i> Assumed to be 90% in a low and 10% in a high population density area.
	Truck 40t	130	km	From Stockton, CA. <i>Regionalization:</i> Assumed to be in a high population density area.
Disposal of fly ash in landfill or lagoon (based on Chapter 9)	Truck 40t	-50	km	Assumption. The landfill and lagoon is close to the power plant. <i>Regionalization:</i> Assumed to be in a low population density area.
	Inert material landfill, infrastructure	-1	t	(i.e., construction and closure). <i>Regionalization:</i> Assumed to be in a low population density area.
	Inert material landfill, operation	-1	t	Dominated by diesel consumption (0.75 l/t). <i>Regionalization:</i> Assumed to be in a low population density area.
Slag (1 t)				
Transport of fly ash (to San Francisco, CA)	Boat	10,000	km	From Asia. <i>Regionalization:</i> Assumed to be 99% in a low and 1% in a high population density area. Note that emissions in a low population density area should actually be in a remote area. However, ecoinvent does not contain any emission

Phase of the life cycle	Intermediary flow	Value	Unit	Source and Comment
				compartment that fit for remote area. This asks for further development in the life-cycle inventory databases.
	Truck 40t	130	km	From Stockton, CA. <i>Regionalization:</i> Assumed to be in a high population density area.
Disposal of slag (disposal of fly ash used as proxy)	Truck 40t	-50	km	Assumption. The landfill and lagoon is close to the power plant. <i>Regionalization:</i> Assumed to be in a low population density area.
	Inert material landfill, infrastructure	-1	t	(i.e., construction and closure). <i>Regionalization:</i> Assumed to be in a low population density area.
	Inert material landfill, operation	-1	t	Dominated by diesel consumption (0.75 l/t). <i>Regionalization:</i> Assumed to be in a low population density area.
Portland cement (1 t) (2,360 kg/m³)				
Production of Portland cement	Portland cement	1	t	<i>Regionalization:</i> Assumed to be in a medium population density area (using characterization factor for “unknown” conditions as a proxy).
Transport of Portland cement (for generic U.S. ready-mix concrete plant)	Boat	1,139	km	<i>Regionalization:</i> Assumed to be 99% in a low and 1% in a high population density area. (see Table 10-4)
	Train	101	km	<i>Regionalization:</i> Assumed to be 90% in a low and 10% in a high population density area. (see Table 10-4)
	Truck 40t	218	km	<i>Regionalization:</i> Assumed to be in a high population density area. (see Table 10-4)
Transport of Portland cement (to San Francisco, CA)	Truck 40t	70	km	From Cupertino, CA. <i>Regionalization:</i> Assumed to be in a high population density area.
Coarse aggregate (1 t)				
Production of coarse aggregate	Crushed gravel	1	t	<i>Regionalization:</i> Assumed to be in a low population density area.
Transport of coarse aggregate (for generic U.S. ready-mix concrete plant)	Boat	100	km	<i>Regionalization:</i> Assumed to be 99% in a low and 1% in a high population density area.
	Truck 40t	100	km	<i>Regionalization:</i> Assumed to be in a high population density area.
Transport of coarse aggregate (Sechelt aggregate — from Vancouver, BC, Canada) (to San Francisco, CA)	Boat	1,500	km	<i>Regionalization:</i> Assumed to be 99% in a low and 1% in a high population density area.
	Truck 40t	0.25	km	From the harbor to the plant, which is situated in the harbor. <i>Regionalization:</i> Assumed to be in a high population density area.
Fine aggregate (1 t)				
Production of fine aggregate	Sand	1	t	<i>Regionalization:</i> Assumed to be in a low population density area.
Transport of fine aggregate (for generic U.S. ready-mix concrete plant)	Boat	100	km	<i>Regionalization:</i> Assumed to be 99% in a low and 1% in a high population density area.
	Truck 40t	100	km	<i>Regionalization:</i> Assumed to be in a high population density area.
Transport of fine aggregate (Sechelt sand — from Vancouver, BC, Canada — and Angel Island sand — from San Francisco Bay) (to San Francisco, CA)	Boat	1,500	km	For Sechelt sand. <i>Regionalization:</i> Assumed to be 99% in a low and 1% in a high population density area.
	Truck 40t	0.25	km	From the harbor to the plant, which is situated in the harbor. For Sechelt sand. <i>Regionalization:</i> Assumed to be in a high population density area.
	Boat	10	km	For Angel Island sand. <i>Regionalization:</i> Assumed to be 100% in a high population density area.
	Truck 40t	0.1	km	For Angel Island sand. <i>Regionalization:</i> Assumed to be in a high population density area.
Admixture (1 kg)				
Production of admixture	Pozzolith 200 N	1	kg	Approximated as (based on information available on packaging): 20% Triethanolamine, 80% Chemicals organic
Transport of admixture	Truck 40t	50	km	<i>Regionalization:</i> Assumed to be in a high population density area.
Ready-mix plant (per t of concrete)				
Infrastructure	Concrete mixing plant	2.1E-8	plant	Capacity: 20,000,000 m ³ concrete per lifespan
Operation	Electricity	1.85	kWh	ecoinvent v2.01 (4.36 kWh/m ³ , 2,380 kg/m ³)
	Other processes	1	t	<i>Regionalization:</i> Assumed to be in a high population density area.

The calculated weighted average travel distance of one tonne of imported cement transported to a United States terminal is approximately to 5,300 km by boat, 150 km by train

and 74 km by truck, using distances and amounts of cement imported from the different countries in 2004 (Table 10-3).

Table 10-3: Computation of transportation distances for import cement from origin to United States terminal.

Country of import	Total amount imported (t/y) (USGS 2008)	Partition per mode of transportation (assumption)			Estimated distance (in km) per mode of transport			
		Truck (t/y)	Railroad (t/y)	Barge and boat (t/y)	Truck (km)	Railroad (km)	Barge and boat (km)	
Canada	5,700	2,850	2,850		500	1,000		
China	2,000			2,000			10,000	
Colombia	2,000			2,000			2,000	
Greece	2,000			2,000			10,000	
Korea	1,600			1,600			10,000	
Mexico	1,400	700	700		500	1,000		
Sweden	1,000			1,000			10,000	
Taiwan	1,000			1,000			10,000	
Thailand	2,800			2,800			10,000	
Venezuela	2,500			2,500			2,000	
Others	2,000			2,000			7,000	
Total import	24,000	3,550	3,550	16,900				
<i>Weighted average distance (in km) for 1 t of imported cement to United States terminal</i>					<i>74</i>	<i>148</i>	<i>5,292</i>	

The calculated weighted average distance that one tonne of cement transported from the cement plant (United States and foreign combined) to a United States cement consumer is approximately 1,100 km by boat, 100 km by train and 220 km by truck.

Table 10-4: Computation of transportation distances from a cement plant to a U.S. cement consumer.

	Total amount shipped (t/y) (USGS 2008)	Partition per mode of transportation (USGS 2008, except for foreign, which is based on Table 10-3)			Estimated distance (in km) per mode of transport			Estimated distance (in km) per mode of transport		
		Truck (t/y)	Railroad (t/y)	Barge and boat (t/y)	Truck (km)	Railroad (km)	Barge and boat (km)	Truck (km)	Railroad (km)	Barge and boat (km)
U.S. plant production	92,000									
From U.S. plant to U.S. terminal (2004)	28,000	5,500	14,000	8,500	200	500	500	39	250	152
Shipment from foreign country to U.S. terminal	24,000	3,550	3,550	16,900				74	148	5,292
From U.S. plant to consumer (2004)	64,000	62,000	1,800	200	200	500	500	194	14	2
From U.S. terminal to U.S. consumer (2004)	52,000	50,000	500	1,500	200	500	500	192	5	14
Total shipped to U.S. consumer	116,000									
<i>Weighted average distance for 1 t of cement, if U.S. cement</i>								<i>205</i>	<i>87</i>	<i>52</i>
<i>Weighted average distance for 1 t of cement, if foreign cement</i>								<i>266</i>	<i>153</i>	<i>5,306</i>
<i>Weighted average distance for 1 t of cement, if unknown cement</i>								<i>218</i>	<i>101</i>	<i>1,139</i>

To strengthen the confidence in the data used for truck and train transportation, data reported by Facanha and Horvath (2006) for PM₁₀ and NO_x emissions are compared to the data provided byecoinvent. The analysis performed in Chapter 8 shows that the difference between the two sources is not significant. For consistency among the sources of data and especially because the data reported by Facanha and Horvath (2006) do not differentiate between PM_{2.5} and PM_{10-2.5}, I decided to use the data reported byecoinvent in the main analysis.

Life-cycle impact assessment

The life-cycle impact assessment used to evaluate the life-cycle inventory is a combination of the characterization factors calculated in Chapter 4 for respiratory impacts associated with inorganics and IMPACT 2002+ (Jolliet et al. 2003, Humbert et al. 2009) for

other impact categories. Results are presented for human health, ecosystems quality, climate change, and resources. Regionalization is considered both in the inventory and in the impact assessment. The evaluation of the life-cycle inventory is performed using the software SimaPro (PRé 2006). The damage to human health evaluation takes into account all emissions other than the fly ash dust itself. Potential damage to human health associated with occupational exposure to fly ash dust during handling of fly ash or during fly ash concrete crushing are outside the scope of this chapter but should also be considered in further studies.

Regionalization of life-cycle assessment

A detailed discussion of the issue of regionalization of life-cycle assessment is outside of the scope of this chapter (see Sedlbauer et al. (2007) and Chapters 3 and 5 for more discussion on this issue). Several parameters can change when taking into account the differences among regions. This section discusses the parameters specific to this chapter. Whether the use of continental or local electricity grid mix is the correct approach in life-cycle assessment is a methodological issue and is outside the scope of this chapter (see Lesage et al. (2008) for more discussion on this issue). If it is decided to adapt the electricity mix used in the life-cycle assessment to the regional grid, then the inventory associated with the production of electricity changes when the region changes. The type of truck (size and efficiency) or train (size, efficiency, diesel or electric) can change when changing regions, and thus the emissions associated with transportation can change. The different alternatives to the disposal of fly ash if it is not used in concrete can also change when changing regions (see Chapter 9 for different alternatives to disposal of fly ash). Finally, the overall amount of emissions can vary because of changes in transportation distances.

In the present case study the overall type and amount of emissions caused by a change in the transportation distances, the type of train (diesel or electric) and the production of Portland cement are considered to change in the inventory when accounting for regionalization. Because varying the electricity mix does not change significantly the results in the case of fly ash concrete (see Chapter 9), I decided not to address the influence of changing the electricity mix in this chapter.

In the life-cycle impact assessment, regionalization influences the evaluation of the damage to human health and ecosystems quality since these two categories are, at least partly, influenced by local conditions. However, the evaluation of climate change and resources are not affected by regionalization as these two categories are global. In this specific case study, the main point of accounting for regionalization is the evaluation of the damage to human health caused by substantial variations in the population distribution patterns within the western United States. Regionalization is performed using the characterization factors developed in Chapter 4 for respiratory effects of inorganics.

Unit processes

A list of the main unit processes described in this chapter as “foreground” processes, as well as the electricity mix used as “background” process for all these unit processes is shown in Table 10-5. The default electricity mix used in the different processes is the North American mix (United States, Canada and Mexico). One of the advantages of using ecoinvent (Frischknecht 2005) is that this inventory database allows one to perform regionalization using the archetype-based approach (Chapters 3 and 5), i.e., to differentiate conveniently among emissions in high or low population density areas. The last column of Table 10-5 indicates an assumption made that is different from ecoinvent regarding the archetype where emissions of pollutants occur. Values for regionalization take into consideration the effect for human toxicity and respiratory inorganics

for human health. Three unit processes are created for this chapter. The production of Portland cement in the United States is created using values for fuel consumption specific to the United States (van Oss 2004). The resulting process of Portland cement manufacturing in the United States is 10% to 66% more burdensome (depending on the impact category evaluated) than the Portland cement process defined in ecoinvent (adapted to Portland cement that is 100% based on clinker). Indeed, the production of United States cement requires three times more coal and petroleum coke than the production of its European counterpart. Note that uncertainties in these values exist, mainly arising from assumptions about the energy content of the coal. The two other processes created are Pozzoloth 200 N (admixture) and an electricity mix for North America (see Table 10-5 for summary results).

Table 10-5: Impact score of the different unit processes used in this chapter (if not otherwise referenced, values are based on ecoinvent v2 – Frischknecht 2005).

Unit process	Unit (“per ...”)	Human health (DALY)		Eco-system quality (PDF·m ² ·y)	Climate change (kg CO ₂ -eq)	Resources (MJ primary non-renewable)	Type of regionalization (for operation)
		without regionalization	with regionalization				
Truck 40t, average load	t·km	1.9E-7	6.8E-8	0.080	0.16	2.8	low pop
Truck 40t, average load	t·km	1.9E-7	4.1E-7	0.080	0.16	2.8	high pop
Truck 40t, average load	t·km	1.9E-7	1.8E-7	0.080	0.16	2.8	-
Train, diesel, average load	t·km	7.8E-8	3.5E-8	0.011	0.05	0.7	90% low 10% high
Train, electric, average load	t·km	2.9E-8	2.2E-8	0.006	0.04	0.7	-
Boat, barge, on the river or along the coast	t·km	5.4E-8	6.5E-9	0.009	0.04	0.6	-
Boat, transoceanic	t·km	2.3E-8	2.1E-9	0.002	0.01	0.2	-
Inert material landfill, infrastructure	t	5.7E-6	2.7E-6	2.100	4.60	170.0	low pop
Inert material landfill, operation	t	4.8E-6	6.5E-7	0.390	2.60	39.0	low pop
Portland cement, at plant, “unknown” pop area (ecoinvent values, 100% clinker)	t	2.2E-4	1.9E-4	39.000	910.00	3,800.0	medium pop
Portland cement, at plant, low pop area (ecoinvent values, 100% clinker)	t	2.2E-4	7.8E-5	39.000	910.00	3,800.0	low pop
Portland cement, at plant, “unknown” pop area (U.S. values, 100% clinker, recalculated for this chapter, using ecoinvent data)	t	2.4E-4	2.3E-4	47.000	1,060.00	6,300.0	medium pop
Portland cement, at plant, low pop area (U.S. values, 100% clinker, recalculated for this chapter, using ecoinvent data)	t	2.4E-4	8.8E-5	47.000	1,060.00	6,300.0	low pop
CO ₂ released from clinker (100% clinker) included in the value above:					650.00		
Gravel, crushed, at mine	t	5.0E-6	2.7E-6	1.200	4.30	120.0	low pop
Sand, at mine	t	3.5E-6	1.2E-6	0.620	2.40	52.0	low pop
Water	m ³	2.2E-7	1.7E-7	0.300	0.36	5.5	-
Admixture, Pozzoloth 200 N (created for this chapter, using ecoinvent data)	kg	1.5E-6	3.2E-6	0.250	2.10	74.0	-
Concrete mixing plant (ready-mix), infrastructure	plant	3.9E+0	3.4E+0	2.0E+6	3.1E+6	4.3E+7	-
Concrete mixing plant (ready-mix), operation (other than electricity)	t	2.0E-6	2.1E-6	0.280	1.60	25.0	high pop
Electricity (North America mix), at grid (created for this chapter, using ecoinvent data)	kWh	3.5E-7	2.0E-7 ^a	0.160	0.67	11.0	-
Electricity (U.S. mix), at grid (created for this chapter, using ecoinvent data)	kWh	3.7E-7	1.9E-7 ^a	0.160	0.73	12.0	-
Electricity (coal mix), at grid (created for this chapter, using ecoinvent data)	kWh	6.0E-7	1.1E-7 ^a	0.210	1.10	13.0	-
Electricity (non-coal mix), at grid (created for this chapter, using ecoinvent data)	kWh	n/a	2.6E-7 ^a	0.110	0.30	11.0	-

^a See explanation in footnote of Table 9-3.

10.4. Results

Impact assessment

The burdens and benefits of the production of one tonne of fly ash concrete are shown in Figure 10-2, Figure 10-3, Figure 10-4 and Figure 10-5, for, respectively, human health (in DALY – Murray and Lopez 1996 – using regionalization of damage, i.e., having the characterization factors of pollutants differentiated depending on whether they are emitted in a high, medium or low population density area), ecosystem quality (in PDF·m²·y), climate change (in kg CO₂-eq) and resources (in MJ of primary non-renewable energy). Results are presented for a generic ready-mixed concrete plant in the United States and for the specific case of Bode Gravel ready-mixed concrete plant in San Francisco.

Values of Figure 10-2, Figure 10-3, Figure 10-4 and Figure 10-5 are obtained by multiplying the reference flows of Table 10-2 with the impacts per unit process indicated in Table 10-5. An example of detailed computation is shown in section 10.8.

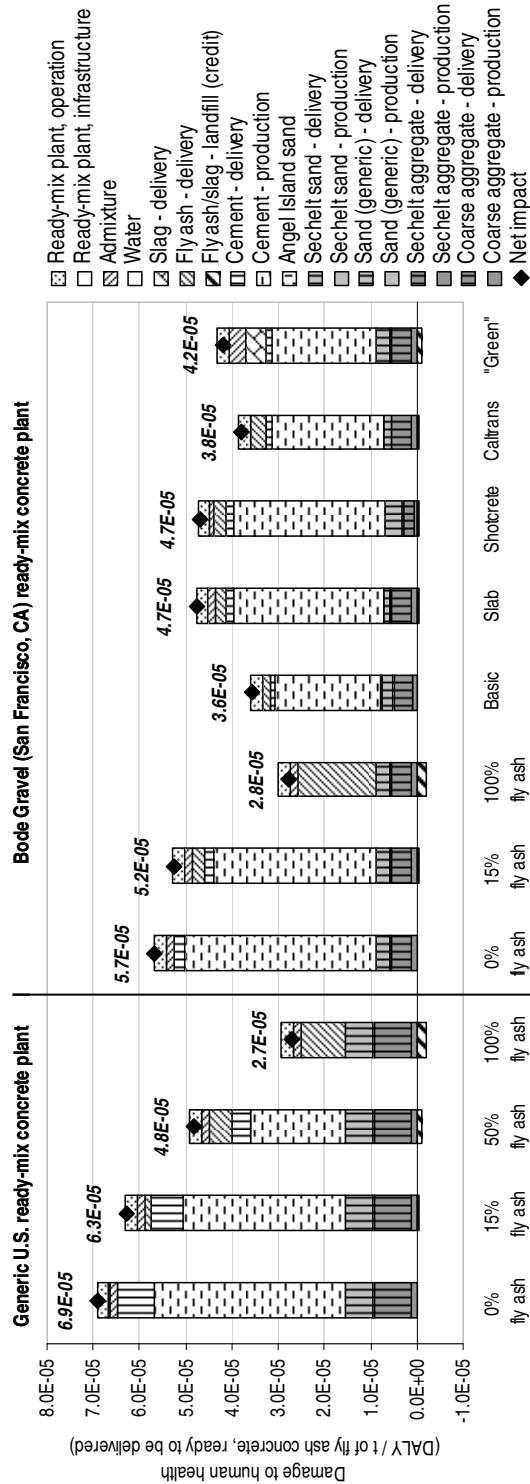


Figure 10-2: Damage to human health of one metric tonne of concrete, ready to be delivered, for a generic (i.e., average) U.S. ready-mix plant, as well as for the specific case of San Francisco

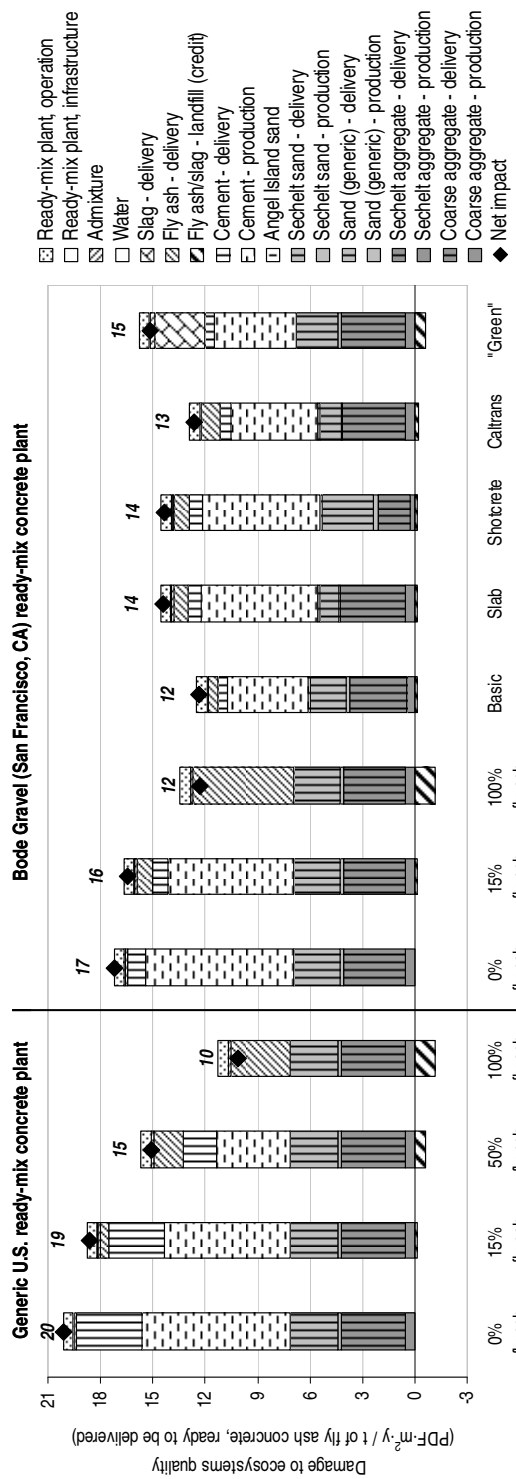


Figure 10-3: Damage to ecosystem quality of one metric tonne of concrete, ready to be delivered, for a generic (i.e., average) U.S. ready-mix plant, as well as for the specific case of San Francisco

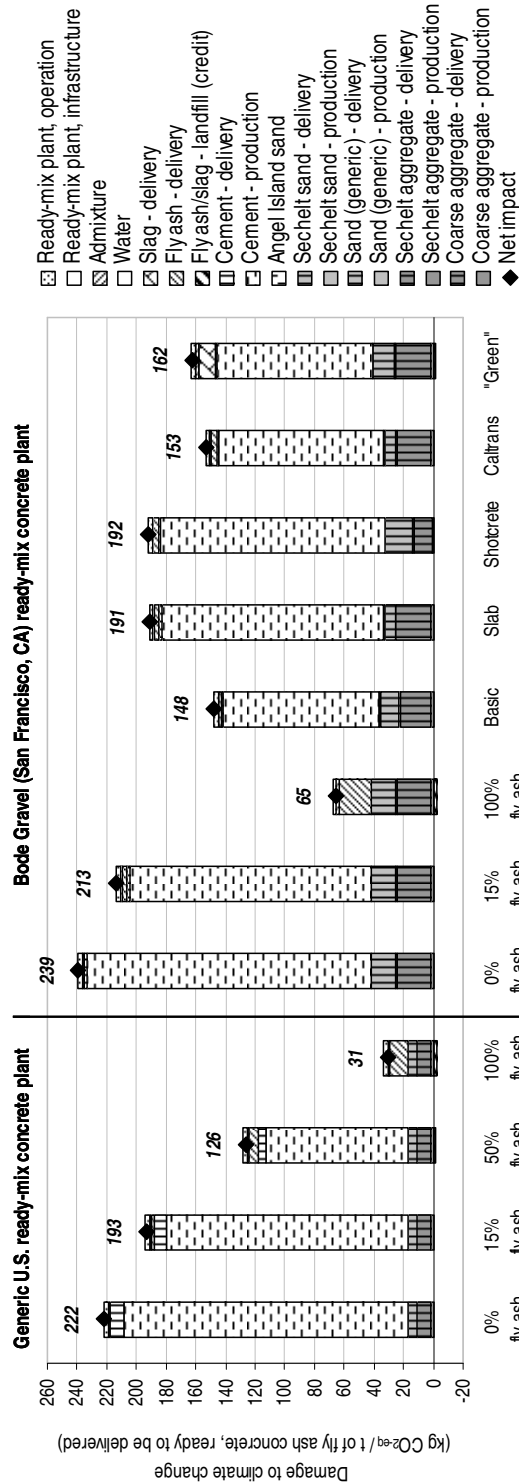


Figure 10-4: Damage to climate of one metric tonne of concrete, ready to be delivered, for a generic (i.e., average) U.S. ready-mix plant, as well as for the specific case of San Francisco

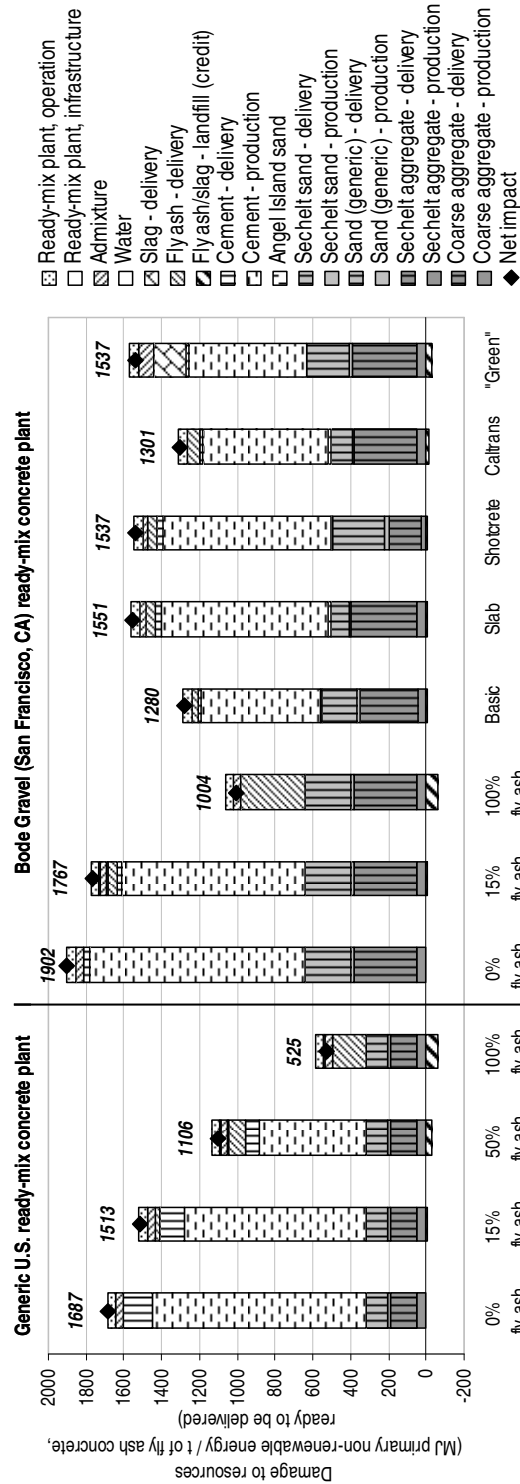


Figure 10-5: Damage to resources of one metric tonne of concrete, ready to be delivered, for a generic (i.e., average) U.S. ready-mix plant, as well as for the specific case of San Francisco

Discussion

The four damage indicators show a similar trend. One tonne of pure Portland cement concrete, ready to be delivered to the job site, emits approximately 220 to 240 kg CO_{2-eq} and consumes approximately 1,700 to 1,900 MJ of primary non-renewable energy. Commercial concrete (having between 15% and 25% of fly ash by volume of binder) emits between approximately 150 and 210 kg CO_{2-eq} and consumes approximately 1,300 to 1,800 MJ of primary non-renewable energy per tonne of concrete. As a matter of comparison, values provided by other sources are: 110 kg CO_{2-eq} per tonne of concrete (ecoinvent v2.01 - Frischknecht 2005); 102, 124, 145, 107 kg CO_{2-eq} per tonne of concrete (for 3,000, 4,000 and 5,000 psi pure Portland cement, and 4,000 psi containing 15% fly ash and 85% Portland cement, respectively) considering cement and aggregate production and transportation and concrete plant operation (Caroll et al. 1998), i.e., 0.87% reduction in CO_{2-eq} emissions per 1% added fly ash. Regarding energy consumption, other studies provide the following values: 1,690 MJ per tonne of concrete (Nisbet et al. 2002), 1,400 to 2,000 MJ per tonne of concrete (Alcorn and Baird 1996), 1,600 to 2,000 MJ per tonne of concrete (Buchanan and Honey 1994), 1,200 to 2,000 MJ per tonne of concrete (FEMP 2001), 2,000 to 8,800 MJ per tonne of concrete (Lawson 1995). Values for CO_{2-eq} emissions of the present chapter are on higher than in other studies. However, values for energy consumption of the present chapter are in the same range as the values obtained by other studies. Further analysis as well as access to the background data of the other studies would be needed to understand the reason for this non-correlated result. Damage to human health is dominated by emissions of PM_{2.5}, NO_x and SO_x from Portland cement production, tailpipe emissions, and electricity production. Apart for cases with a dominant fraction of fly ash for binder, impacts are dominated by cement production (up to 90% of the total CO_{2-eq} emissions). Even with 50% of the binder composed of fly ash, the impacts of concrete are still dominated by cement production — 50% is chosen as representing approximately the highest fraction that can be expected over a large scale. The concrete found on the market typically contains 15% fly ash. With an average increase of fly ash content from 15% to 50%, the damage to resources (mostly coal and oil consumption) and greenhouse gas emissions (mostly CO₂) related to concrete manufacturing could be reduced by approximately one third. After cement production, other sources of impacts are the delivery of aggregate (coarse and fine), delivery of cement and delivery of fly ash. For fractions above 50%, delivery of fly ash and slag become dominant. Credits from avoidance of disposal in landfill of fly ash and slag are not important (in general less than 1% — with some exceptions, up to 10% — of the full life-cycle impacts). Impacts from water consumed in the ready-mix plant and impacts from the infrastructure of the ready-mix plant are negligible (the sum of the two is always less than 0.5% of the total impacts). Overall, the impact of concrete is reduced almost linearly with the fly ash content: each percent of Portland cement replaced by fly ash will contribute to a reduction of environmental impacts of concrete by 0.5% for damage to ecosystems quality, 0.7% for damage to resources, 0.8% for damage to human health, and 0.9% for damage to climate.

Admixture

The admixture used to model the damage is the water-reducing Pozzololith® 200 N (by Master Builders) (Bode Gravel, San Francisco, California, personal communication). Interestingly, even if the mass of the admixture is very small (less than one kg per tonne of concrete, i.e., less than 0.1% by weight), its contribution to the final energy demand is not negligible. Indeed, the contribution of admixture to final energy demand is between 0.2% and 10% depending on the fly ash content and assumption of admixture composition. The

contribution of admixture to final energy demand can be expected to be 4% for a concrete using 50% fly ash by volume of binder. Therefore, the fraction of total damage of concrete caused by admixture is 100 times the fraction of admixture in the total mass of concrete. It means that relative to its amount used, admixture has a substantial share of the impacts.

Influence of regionalization on damage to human health

The influence of regionalization on the results of damage to human health related to the impacts of supply chain associated with the case of San Francisco is shown in Figure 10-6. Values of Figure 10-6 are obtained by multiplying the reference flows of Table 10-2 with the impacts per unit process with “with regionalization” indicated in Table 10-5.

One observes that the damage caused by the shipping of coarse and fine aggregate is substantially affected by regionalization. This observation is due mainly to reassessment of boat shipping (barge and transoceanic). Indeed, regionalized damage to human health of boat shipping is one order of magnitude lower than damage to human health using generic characterization factors. Indeed regionalization captures the fact that most of the boat emissions occur in low (or even zero) population density areas. If regionalization is not considered, emissions of boats are weighted the same as emissions from trucks, cement plants or power plants that are situated within populated areas. When train ride is assumed to be 90% in a low and 10% in a high population density area, regionalization reduces the damage caused by train by a factor of two because of lower amount of people exposed to train emissions. No other major process for the production of concrete, in the case of San Francisco, is affected substantially by regionalization. However, if the cement plant was situated in an area with low population density, the damage to human health of the cement production would be reduced by a factor of three. A reduction of damage to human health by a factor of three for cement results in a reduction of damage to human health by one half for concrete based on pure Portland cement. For concrete using a high proportion of fly ash or slag shipped from long distance, the higher the fraction of fly ash or slag, the less substantial the reduction of damage to human health.

Break-even distances

The use of fly ash or slag as partial substitute for Portland cement reduces the impacts of concrete because the production of fly ash or slag is considered to be a waste and thus with no impacts (see Chapter 9 for a discussion about when fly ash is considered a co-product instead of a waste). However, longer shipping distances for fly ash or slag compared to shipping distance of cement can partly undo the benefits provided by the displacement of Portland cement. For the use of fly ash or slag to be more environmentally friendly than Portland cement, shipping distances for fly ash and slag need to be shorter than the distances indicated in Table 10-6. Table 10-6 indicates the break-even distances at which point impacts from shipping are equal to the impacts from producing Portland cement that is 100% made from clinker. These distances are found by dividing the impacts of producing a tonne of Portland cement (Table 10-5) by the impacts of transport (Table 10-5) for different modes. These distances are the ones that can be afforded in addition to the shipping distance of Portland cement to the ready-mix concrete plant. The United States average shipping distances of Portland cement are approximately 220 km by truck, 100 km by train, and 1,100 km by boat (see Table 10-2). Note that if the fly ash or slag is shipped by train or boat, distances driven by truck from the rail yard or port to the ready-mix plant need to be considered in addition to the distance traveled by train or by boat. The credits earned from avoiding disposing of the fly ash or slag in a landfill are not considered in these break-even distances. Indeed, fly ash and slag are often used locally as a substitute for low-grade (i.e., inert) material, and landfill will not necessarily be the only alternative destination.

Table 10-6: Break-even transportation distances (in km) for the shipping of fly ash or slag to be as impacting as the production of Portland cement.

Mode of transportation:	Damage category used as an indicator for the break-even distance					
	Human health (DALY)			Ecosystems quality (PDF·m ² ·y)	Climate change (kg CO ₂ -eq)	Resources (MJ primary non-renewable)
	without regionalization	with regionalization				
		cement plant in a medium pop density area	cement plant in a low pop density area			
Truck, in a high population density area	1,300 (1,200)	560 (460)	210 (190)	590 (490)	6,500 (5,600)	2,300 (1,400)
Truck, in a low population density area	1,300 (1,200)	3,400 (2,800)	1,300 (1,100)	590 (490)	6,500 (5,600)	2,300 (1,400)
Truck, in an area with unknown population density	1,300 (1,200)	1,300 (1,100)	490 (430)	590 (490)	6,500 (5,600)	2,300 (1,400)
Train, diesel	2,800 (2,500)	6,600 (5,400)	2,500 (2,200)	4,400 (3,700)	21,000 (18,000)	8,400 (5,100)
Train, electric	8,400 (7,600)	11,000 (9,100)	4,200 (3,700)	6,200 (5,100)	25,000 (21,000)	9,000 (5,400)
Boat, barge on a river	4,600 (4,100)	35,000 (29,000)	13,000 (12,000)	5,300 (4,400)	27,000 (23,000)	11,000 (6,300)
Boat, transoceanic	11,000 (9,800)	110,000 (90,000)	42,000 (37,000)	24,000 (20,000)	110,000 (91,000)	32,000 (19,000)

Default case indicates the distances when compared to Portland cement modeled with U.S. conditions. Values in brackets indicate the distances when compared to Portland cement modeled with ecoinvent data.

The influences of the regionalization on the inventory as well as impact assessment can be seen in Table 10-6. The influence of the regionalized inventory is reflected in the difference in the processes between United States and Europe for cement production, and between diesel (western United States and Canada) and electric (eastern United States and Europe) trains for transportation. The influence of the regionalized impact assessment is reflected in the difference between a truck driving in an area with low, high or unknown population density area, in a cement plant situated in an area with medium or low population density, and simply between

damage to human health evaluated without and with considering regionalization. Depending on the damage category chosen as an indicator, as well as the location and efficiency of the cement plant, the break-even distance is approximately between 200 km and 7,000 km for truck shipping, between 2,000 km and 30,000 km for train shipping, between 4,000 km and 30,000 km by barge, and between 10,000 km and 100,000 km by transoceanic boat. Thus fly ash should be shipped from a regional coal fired plant (i.e., no farther than a few hundred km) if shipped by truck. However, fly ash can be imported from the Midwest if shipped by train. Fly ash can also be shipped to or from overseas if done with transoceanic boat. The limiting damage indicators alternate between human health and ecosystems quality. Considering the high uncertainties embedded in the evaluation of these two damage categories, damage to resources could also be the limiting damage indicator because, based on Table 10-6 results, the difference with human health is sometimes less than 10%. Climate change is, however, never the limiting indicator. This comes from the fact that Portland cement is CO_{2-eq} intensive because it releases a large amount of CO_{2-eq} during its production (up to one tonne of CO_{2-eq}, including 543 kg CO₂ from decarbonization (Frischknecht 2005), per tonne of pure Portland cement). It is highly unlikely for the different modes of transportation to emit more than this amount of CO₂. Indeed, 5,600 km by truck, 18,000 km by train, 23,000 km by barge or 91,000 km by transoceanic boat (lowest break-even distances based on CO₂ in Table 10-6) are, from a geographic perspective, virtually impossible to achieve. Therefore, the indicator that is the limiting factor when calculating break-even distance, which is often damaging to human health or ecosystems quality, is sensitive to regionalization because its impacts are highly dependent on the local context. The damage to human health, which can vary by a factor of 100 if emissions occur in a high or low population density area (Chapters 2, 3, 4 and 8), is especially sensitive to regionalization. Damage to resources is also sensitive to regionalization through the energy mix for the cement production and the energy efficiency of the shipping (i.e., fuel consumption per tonne of fly ash or slag transported). However, it is less sensitive to regionalization than damage to human health since resource consumption is not expected to vary by more than a factor of two, and definitely less than a factor of ten among regions (compared to a factor among regions of up to 100 for damage to human health). These observations show the importance of regionalization to increase the reliability of the results, which in the present chapter are the shipping distances of fly ash that can be afforded to have fly-ash concrete be more environmentally beneficial than concrete based purely on Portland cement.

Application on a national scale

Results show that when one tonne of cement is displaced by one tonne of fly ash, the production of approximately one tonne of CO_{2-eq} can be avoided. This result is closely correlated to the values of one tonne of CO_{2-eq} per tonne of Portland cement produced in the United States (Table 10-5). On a national scale, 127 million tonnes of cement and 20 million tonnes of fly ash were used in concrete in 2006 (USGS 2008). This corresponds to 260 million m³ of ready-mixed concrete (PCA 2009), or 620 million tonnes of ready-mixed concrete, responsible for the consumption of approximately 1.2 billion GJ of primary non-renewable energy (~ 1% of United States energy consumption) and 150 million tonnes of CO_{2-eq} emissions (~ 2% of United States emissions, which are approximately 7 billion tonnes of CO_{2-eq} (USEPA 2009)). If most of the entire production of fly ash in the United States (~ 70 million tonnes per year) could be used to displace cement (in the United States as well as in neighboring countries or overseas, if transported by boat) instead of the current amount of only 20 million tonnes per year, the savings could increase to 0.3 billion GJ of primary non-renewable energy and 50 million tonnes of

CO_{2-eq}. As a matter of comparison, this would represent a 0.3% reduction of United States energy consumption and a 1% reduction of United States CO_{2-eq} emissions. Hence a wise use of fly ash can significantly reduce United States greenhouse gas emissions.

A similar analysis can be done for damage to human health. Figure 10-2 and Table 10-1 show that benefits from using fly ash in concrete are about 2.3E-4 DALY/t of fly ash. This value is calculated by dividing the reduction in damage to human health from using 50% fly ash concrete instead of 0% fly ash concrete, which is 2.1E-5 DALY/t of concrete (i.e., 6.9E-5 – 4.8E-5 DALY/t of concrete — Figure 10-2) by the fly ash content of 50% fly ash concrete, which is 0.09 t fly ash/t of concrete (Table 10-1). Therefore, if most of the entire production of fly ash in the United States (~ 70 million tonnes per year) could be used to displace cement instead of the current amount of only 20 million tonnes per year, the potential reduction in damage to human health could be approximately 12,000 DALY/y or 0.2% of annual “DALYs” caused by outdoor pollution in the United States (Table 5-7). Using a value of 10 DALY per premature death (Table 4-4), using the full amount of fly ash to displace Portland cement would potentially avoid 1,200 premature deaths annually. The data used in Figure 10-2 were generated considering regionalization. If one would have not use regionalization, the overall calculated benefits would have been a factor of two higher (Figure 10-6). Therefore, if one wants to use life-cycle assessment for policy-decision making — as for example, whether to promote or not reuse of fly ash, or whether to provide a tax credit for fly ash in function of its actual benefit for human health — regionalization may not change the ranking between scenarios (e.g., the ranking does not change in Figure 10-6) but may change the absolute value of the potential benefits that can be expected (e.g., 1,200 instead of 2,400 premature deaths avoided annually). Passed a certain level of absolute benefits, using regionalization may become important to increase the accuracy of life-cycle assessment results. The level is to be discussed, but a difference of about 1,200 premature deaths annually can be considered sufficiently important to justify investing more time in generating the regionalized figures.

10.5. Conclusion and recommendations

The overall impacts of pure Portland cement concrete are dominated by the cement production (50% to 90%, depending on the damage chosen as an indicator). One tonne of pure Portland cement concrete, ready to be delivered to the job site, emits approximately 230 kg CO_{2-eq}. Commercial concrete (with 15% to 25% fly ash) emits approximately 150 to 210 kg CO_{2-eq} per tonne of concrete. The two critical parameters influencing the overall impact per tonne of concrete are (i) the overall amount of binder needed per tonne of concrete, and (ii) the fraction of the binder that is made of fly ash or slag. Indeed, the use of fly ash or slag can significantly reduce the impacts of concrete. However, attention needs to be paid to the shipping of fly ash or slag. If shipped over long distances, the shipping needs to be done by train or boat and the shipping by truck reduced to a minimum (a few hundred km at the most). The limiting factors when evaluating the break-even distance are the damage to human health and ecosystem quality, as well as the damage to resources. Damage to climate is not a limiting factor for the shipping of fly ash or slag. Damage to human health is dominated by emissions of PM_{2.5}, NO_x and SO_x from Portland cement production, tailpipe emissions, and electricity production. Since damage to human health can vary up to a factor of 100 between cases where emissions occur in a low population density area and those that occur in a high population density area, the regionalization of the inventory of emissions (i.e., where PM_{2.5}, NO_x and SO_x are emitted) and impact assessment (i.e., how do the characterization factors for PM_{2.5}, NO_x and SO_x change depending

on where these pollutants are emitted) is important to increase the reliability of the results. Note that even if regionalization modifies significantly the results, it may be important only if the scale of the effects matters. Finally, once a high fraction of fly ash or slag is used, the impact of aggregate shipping starts to be substantial (up to 30% of the impacts for a 50% fly ash concrete) and should be watched carefully. To minimize the environmental impacts associated with concrete, (i) fly ash or slag needs to be used as much as technically feasible, (ii) fly ash or slag needs to be shipped as much as possible by train or boat (and less than a few hundred km if by truck), and (iii) coarse and fine aggregate should be shipped from less than approximately 100 km if by truck or 500 km if by boat. The conclusions about fly ash are valid as long as the fly ash is considered a waste and not a co-product from the coal-fired power plants (see Chapter 9 for a detailed discussion on the issue of waste versus co-product).

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10.8. Supporting information

Table 10-7 shows an example of detailed computation to obtain the climate change score (Figure 10-4) for a generic United States ready-mix concrete plant, containing 15% of fly ash (second bar in Figure 10-4).

Table 10-7: Example of detailed computation to generate the impact score.

Life cycle stage	Composition (Table 10-1) (t/t of concrete)	Reference flows (composition multiplied by reference flow in Table 10-2) (per t of concrete)	Impact score of the unit process (Table 10-5) (kg CO _{2-eq})	Damage score (multiplication of the reference flows column and the unit process impact score column) (kg CO _{2-eq} per t of concrete)
Coarse aggregate - production	0.430	0.43 t	4.30 per t	1.9
Coarse aggregate - delivery	0.430	0.43 t × 100 km = 43 tkm by boat	0.04 per tkm	1.7
		0.43 t × 100 km = 43 tkm by truck	0.16 per tkm	6.9
Sechelt aggregate - production	-	-	-	-
Sechelt aggregate - delivery	-	-	-	-
Sand (generic) - production	0.320	0.32 t	2.40 per t	0.77
Sand (generic) - delivery	0.320	0.32 t × 100 km = 32 tkm by boat	0.04 per tkm	1.3
		0.32 t × 100 km = 32 tkm by truck	0.16 per tkm	5.1
Sechelt sand - production	-	-	-	-
Sechelt sand - delivery	-	-	-	-
Angel Island sand	-	-	-	-
Cement - production	0.153	0.153 t	1,063 per t	163
Cement - delivery	0.153	0.153 t × 1139 km = 174 tkm by boat	0.01 per tkm	1.7
		0.153 t × 101 km = 15.5 tkm by train	0.05 per tkm	0.78
		0.153 t × 218 km = 33.4 tkm by truck	0.16 per tkm	5.3
Fly ash/slag - landfill (credit)	0.027	-0.027 t (Inert material landfill, infrastructure)	4.60 per t	-0.12
		-0.027 t (Inert material landfill, operation)	2.60 per t	-0.070
		0.027 t × -50 km = 1.35 tkm by truck	0.16 per tkm	-0.22
Fly ash - delivery	0.027	0.027 t × 1000 km = 27 tkm by train	0.05 per tkm	1.4
		0.027 t × 100 km = 2.7 tkm by truck	0.16 per tkm	0.43
Slag - delivery	-	-	-	-
Water	0.070	0.070 m ³	0.36 per m ³	0.025
Admixture	0.00054	0.54 kg (production)	2.10 per kg	1.1
		0.00054 t × 50 km = 0.027 tkm by truck	0.16 per tkm	0.0043
Ready-mix plant, infrastructure		2.1E-8 plant	3.1E+6 per plant	0.065
Ready-mix plant, operation		1.85 kWh	0.73 per kWh	1.4
		1t for other processes	1.60 per t for other processes	1.6
Net impact				193

A similar approach is used to generate the results for all other damage scores (“bars” in the different figures) presented in this chapter.

11. Conclusions

Life-cycle assessments, and especially carbon footprint assessments, are increasingly used for decision and policy-making in industry and government. However, when it comes to impacts other than carbon footprint, some are skeptical about the accuracy and robustness of life-cycle assessment results. A typical example of skepticism relates to the impact categories that are sensitive to regionalization, especially if those impact categories are calculated using generic scenarios and parameters (i.e., national, continental or global average inventory and characterization factors). One of the categories that is sensitive to regionalization is damage to human health. Regionalization can be important for cases that have significant and extended supply chains that may cross regions. To address the skepticism, the current practice in life-cycle assessment stresses the importance of being “geographically differentiated” or “regionalized.”

In this dissertation I addressed the issue of regionalization in life-cycle assessment with a particular emphasis on impact assessment. I explored how life-cycle assessment results can potentially be influenced when the location of emissions is taken into account, with an emphasis on damage to human health.

Through the development of the IMPACT North America and UFPM models, and the review of other models evaluating particulate matter, I studied questions related to regionalization of life-cycle assessment. I then drew conclusions about regionalized life-cycle assessment practice and the level of accuracy that can be achieved when addressing human health in life-cycle assessment.

11.1. *Summary of this dissertation*

By combining and extending the results of previous studies and models addressing regionalization in impact assessment, a spatially resolved model for North America, called IMPACT North America, is developed. Geographically differentiated intake fractions and characterization factors are evaluated for the North American continent for human toxicity.

I show that particulate matter contributes to more than 90% of the damage to human health from outdoor emissions. Therefore, more importance is placed on modeling fate, exposure and effect of particulate matter. I review previous work done on particulate matter and provide several modeling approaches. I explore the concept of uptake fraction and use it to re-evaluate intake fractions (through the development of the UFPM model) and effect factors for particulate matter. I generate intake fractions, effect factors and characterization factors for particulate matter for different conditions, with an emphasis on population density patterns at the location of emission.

These factors can be used in damage-oriented impact assessment methods to North America. I identify critical parameters influencing intake fractions and characterization factors, such as population density and agricultural production intensity. The importance of modeling an “urban box” and a “remote area” is also presented and I suggest incorporating these archetypes in life-cycle inventory databases.

I evaluated the influence of using a regionalization scheme based on archetypes (i.e., focusing on the characteristics of the location of emission) instead of on geography (i.e., focusing on where in the world the emission occurs) and found that an archetype based regionalization is less data intensive and can provide the same accuracy as a geographic regionalization.

Following the introduction and evaluation of regionalized life-cycle assessment, I apply regionalization to the case of fly ash concrete. I identify critical parameters influencing the environmental trade-offs between fly ash concrete and Portland cement concrete. Within this context, I also analyze the influence of modeling choices within the life-cycle inventory, revealing that the use of fly ash in concrete may not always be environmentally beneficial if reusing fly ash contributes to increase competitiveness of coal-based electricity.

Through the development of the new models, along with the different applications, various questions related to spatial life-cycle assessment can be studied, and several conclusions can be drawn about what is important to consider when performing regionalization in life-cycle assessment.

I observed that when looking at specific pollutants, when looking at only foreground emissions, at processes where impacts are dominated by foreground emissions or by a specific background process in the supply chain, regionalization of human health can be important. Indeed the characterization factors for specific pollutants can vary by several orders of magnitude depending on emission location. However, if the processes studied or scenarios compared have their impact score on human health dominated by background emissions that are either correlated (e.g., production of diesel for car or coach) or distributed over an extensive supply chain (and therefore with distributed), regionalization will not be important anymore. Indeed, correlated processes will not change ranking among scenarios and distributed emissions will not be sensitive to regionalization.

I also observed that for some specific pollutants, differentiation between urban and rural emissions can make a significant difference in the characterization factors (up to several orders of magnitudes). However, for primary PM, urban versus rural emissions have only limited influence on the characterization factors (one order of magnitude) and for secondary PM, differentiating between urban and rural emissions does not change significantly the characterization factors (~10%). Since processes are often dominated by impacts of PM, regionalization schemes should address PM as a matter of priority. Though differentiating urban and rural emissions is still of importance, it is also important to differentiate urban or rural emissions from remote emissions (this will influence characterization factors of primary PM but especially also of secondary PM by more than a order of magnitude). Because processes situated in remote locations may have significantly lower emission control devices than processes occurring in populated areas, using generic characterization factors would over emphasize contributions from parts of the supply chain occurring in remote area compared to the parts occurring in populated areas (this could partly explain the surprising results obtained for the car and the truck where background impacts seems more important than foreground impacts).

11.2. Uncertainty, variability and future research needs

Modeling the fate, exposure and effect of pollutants is subject to uncertainty and variability. The models and methods developed in this dissertation are complex and data intensive. However, the human health damage score (in DALY/y) from fine particulate matter evaluated with fate, exposure and effect modeling in this dissertation falls within the human health damage (in DALY/y) observed with epidemiological studies. In addition, both life-cycle assessment modeling and epidemiological studies suggest that fine particulate matter dominates overall damage to human health from outdoor emissions. These results are of great interest and suggest that fate, exposure and effect modeling approaches used in damage-oriented life-cycle assessment manage to quantify correctly the overall absolute damage to human health from

outdoor emissions. This suggests that damage-oriented life-cycle assessment is mature enough to be used for strategic decision-making in industry as well as in policy.

Several sources for uncertainty and variation can be identified, indicating where future research is needed.

The IMPACT North America model developed in Chapter 2 is developed for North American conditions. It would be of interest to expand this model to other regions of the world, and especially for regions with fate and exposure conditions significantly different than found in North America. This would also remove potential uncertainties caused by edge effects (i.e., bias in the results from emissions originating in location near the edge of the model).

The results for intake fraction and effect factors generated in Chapter 4 are for global conditions. Further research is needed to identify whether results for specific continents would differ significantly from global results (e.g., intake fractions for North America might be significantly lower than global intake fractions), and if so, generate the results for specific continents.

Intake fractions can vary by several orders of magnitude depending on the location where the emission occur. Therefore, for scenarios or processes that have uncorrelated emissions (i.e., emissions that are not coming from the same process), life-cycle assessment results for damage to human health using generic characterization factors might not be meaningful because the actual, regionalized result can differ by several orders of magnitude from the results obtained using generic characterization factors.

Average intake fractions should be based on emission-weighted average intake fractions. However, as emissions data are often unavailable, I suggest using population distribution as a proxy for emissions distribution. However, one needs to keep in mind that using population-weighted intake fraction as a proxy for emission-weighted intake fraction introduces a potential source of error. Thus, further research is needed in order to identify what averaging methods are most appropriate and to have better data on emission distributions.

Because particulate matter dominates among the damages to human health from outdoor emissions, further research is needed to better address (i) the different types of particulate matter and especially the influence of their particle size distribution and composition (e.g., the effect factors for road dust might be different the effect factor of tailpipe emissions), (ii) fate and exposure modeling such as environmental conditions (mixing height, wind speed, etc.) but also the averaging methods, (iii) exposure scenarios around the world (that can vary by more than an order of magnitude between different urban areas and by three orders of magnitude between remote and urban areas), (iv) exposure from indoor emissions, (v) the influence of buildings on the fate, exposure or effect factors for indoor exposure to PM from outdoor origin, (vi) effect factors, how they are evaluated and whether they depend on local conditions, (vii) how the two separate processes that elicit a health response when particles deposit in the lungs (i.e., either because they are foreign objects or because of the chemicals sorbed on their surface) interconnect, and finally (viii) secondary PM from organics, which has not been treated in the present dissertation.

I showed that when indirect emissions have a significant contribution to the overall impact score (which is often the case), the need for regionalization in life-cycle impact assessment is reduced.

This dissertation focuses on outdoor emissions. However, as shown in Chapters 2 and 4, indoor emissions can have intake fractions several orders of magnitude higher than outdoor emissions – and could potentially have higher damage to human health than outdoor emissions.

Therefore more research is needed to evaluate both emission inventory and exposure scenarios for indoor emissions.

More research is needed to understand how many archetypes are needed and how to make the best use of them in life-cycle inventory databases and life-cycle impact assessment methods.

In this dissertation, I focus on damage to human health. Future research is needed to address regionalization for other damage categories, such as ecosystems quality or water use.

In order to generate usable, meaningful and trustful characterization factors that can be used for policy-based studies, it would be important to give more attention to the quality of the physico-chemical parameters of each substance. A better consistency as well as explicit uncertainty parameters would be needed in physico-chemical parameters databases.

Future research should aim at better implementing regionalization schemes in both life-cycle inventories and life-cycle impact assessment methods.

Finally, even if regionalized life-cycle impact assessment methods are available, applying them requires a matching level of information from the life-cycle inventories. However, life-cycle inventory databases often do not contain the matching level of information — indeed, most life-cycle inventory databases only account for the media of emission (e.g., air) without giving any indication about the environment (e.g., urban or rural). For example, if an impact assessment method can consider important to differentiate between urban and rural emissions and provides characterization factors for both environments, but that the life-cycle inventory database only contains emission in the air, not specifying whether it is in urban or in rural environment, one will not be able to match correctly the characterization factors to the inventory results. Therefore, more research is needed to better address regionalization in life-cycle inventory. I strongly advise life-cycle inventory developers to consider the information needs for regionalized life-cycle impact assessment when developing regionalization schemes in inventory databases.

11.3. Summary of the outcomes

This research provides spatially resolved intake fractions and characterization factors to address human health impacts for North America. In North America, intake fractions can vary by up to several orders of magnitude, mainly depending on population density close to the location of emission.

I show that particulate matter dominates damage to human health from outdoor emissions. This finding points out the need to (i) devote more detailed attention to particulate matter when modeling the fate, exposure and effect of pollutants, and (ii) address particulate matter as a priority in regionalization schemes adopted in life-cycle assessment, such as archetypes selection in life-cycle inventory database.

This work demonstrates that, when addressing the issue of regionalization in inventory and impact assessment, priority should be given to the development of archetype-based regionalization. Spatialization (i.e., evaluating the exact location) should only be used in occasional cases when results from key processes need to be explored specifically.

THE END