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Intake fraction of nonreactive vehicle emissions in US urban areas

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Abstract

Intake fraction, which is the fraction of emissions that are inhaled by people, quantifies the “exposure efficiency” of an emission source. We use three methods to estimate intake fractions for vehicle emissions in US urban areas. First, we use a one-compartment steady-state mass-balance model, incorporating meteorological and demographic data. Second, we use an empirical emissions-to-concentration relationship for vehicle carbon monoxide developed for 15 US urban areas. Third, we analyze model results for benzene and diesel particulate matter from the US Environmental Protection Agency’s National-scale Air Toxics Assessment (NATA). The population-weighted mean intraurban intake fraction for nonreactive gaseous vehicle emissions in US urban areas is estimated to be in the range 7–21 per million, with a best estimate of 14 per million. The intake fraction for diesel particles is 4 per million, based on NATA results. An intake fraction of 4 per million means that 4 mg of pollution are inhaled per kg emitted. Intake fraction values for urban vehicle emissions are usually higher in winter than in summer because of seasonal variability in the atmospheric mixing height. The results presented in this work can be used in health risk assessments, cost–benefit analyses, and other investigations that require a summary of the emission-to-intake relationship.

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1. Introduction

Vehicle emissions are a dominant source of population exposure to several urban air pollutants, including

PM_{2.5}, benzene and other toxic air contaminants, and carbon monoxide. Exposure to motor vehicle emissions causes several acute and chronic health effects (Laden et al., 2000; Pearson et al., 2000; Peters et al., 2004; Pope et al., 2002). Maternal exposure to vehicle emissions is associated with adverse birth outcomes such as low birth weight and premature birth (Wilhelm and Ritz, 2003).

Quantifying the emission-to-inhalation relationship is important for better understanding of air pollution health effects. Intake fraction incorporates in a single number important issues related to population exposure

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to pollution, including the size of the exposed population, the proximity of that population to the emission source, and the persistence of a pollutant in the environment. For a specific source or source class, intake fraction is the ratio of total attributable intake to total emissions (Bennett et al., 2002; Evans et al., 2002). Intake fraction is useful for health risk assessment and for economic and policy analyses.

In this paper, we estimate the intake fraction for nonreactive vehicle emissions in US urban areas. A few previous investigations have addressed this subject. Smith (1993) presented ~20 per million as an order-of-magnitude estimate, and Lai et al. (2000) provided bounding estimates of 0.7–440 per million. Marshall et al. (2003) analyzed ambient monitoring data for carbon monoxide (CO) and benzene, and reported an annual average intake fraction (units: per million) of ~50 (seasonal range: 30 in summer to 80 in winter) for vehicle emissions in California's South Coast Air Basin. Applying trajectory modeling to 40 highway segments in the US, Evans et al. (2002) estimated that intake fractions for primary vehicle PM_{2.5} emissions are 3–18 per million for urban locations and 1–18 per million for rural locations.

This paper advances previous work by being more comprehensive—our analysis covers all urban areas in the US—and by developing and implementing three methods for estimating intake fraction. The three methods incorporate three types of models: a straightforward one-compartment model (Benarie, 1980), an empirical model describing measured CO concentrations in 14 cities (Glen et al., 1996), and the EPA's most sophisticated national-scale exposure model (US EPA, 2002a).

2. Methods

Intake fraction is the fraction of emissions that are taken in by people. For inhalation of a primary pollutant, intake fraction may be expressed as follows:

$$\text{Intake Fraction (iF)} = \frac{\text{Population Intake}}{\text{Total Emissions}} = \frac{\int_{T_1}^{\infty} \left(\sum_{i=1}^P (C_i(t) \cdot Q_i(t)) \right) dt}{\int_{T_1}^{T_2} E(t) dt}. \quad (1)$$

Here, T_1 and T_2 are the starting and ending times of an emissions process; P is the number of people in the exposed population; $Q_i(t)$ is the breathing rate ($\text{m}^3 \text{s}^{-1}$) for individual i at time t ; $C_i(t)$ is the incremental concentration (g m^{-3}) at time t in individual i 's breathing zone that is attributable to the emissions process; and $E(t)$ is the emission rate from the process (g s^{-1}) at time t . In practice, the integral in the numerator is not evaluated to infinite times, but until the attributable

concentration becomes sufficiently small (i.e., until a time significantly later than T_2). Intake fraction is a dimensionless number between zero and one. Typical intake fraction values for urban vehicle emissions are larger than for rural emissions and smaller than for emissions to indoor environments (Lai et al., 2000; Smith, 1993).

Three approaches for estimating intake fractions for vehicular emissions in urban air basins are described and applied in the following subsections. In all three cases, the population average breathing rate, Q , is taken as $12.2 \text{ m}^3 \text{ person}^{-1} \text{ d}^{-1}$, based on metabolic activity studies (Layton, 1993). In this paper, we estimate intraurban intake fractions, i.e., those associated with urban residents' inhalation of emissions that occurred in the same urban area. Our approach represents an important and logical step towards a complete treatment, which would also quantify downwind intakes.

2.1. One-compartment model

We use a one-compartment model (Benarie, 1980) to combine meteorological data on wind speed and mixing heights with demographic data on urban population and land area. This model is often assumed to be too simple to offer reasonable estimates of ambient concentrations in urban areas. It does not offer many of the capabilities of more sophisticated models, such as predicting spatial variability in ambient concentrations. However, for conserved or slowly reacting emissions from broadly distributed ground-level sources, the one-compartment model may offer a reasonably accurate estimate of spatially averaged concentrations in an urban area. Marshall et al. (2003) found that for the South Coast Air Basin, the one-compartment model estimated the long-term basin-wide average ambient concentrations of benzene and CO to within a factor of two.

Examples of pollutants that are reasonably modeled as conserved when considering ambient concentrations include benzene, carbon monoxide, and primary PM_{2.5}. Advection is the dominant removal mechanism for conserved pollutants because the time they take to react or deposit is considerably longer than the residence time of air in an urban basin, which may be estimated as $A^{0.5} u^{-1}$. Here, A is urban land area (m^2) and u is wind speed averaged over the mixing height (m s^{-1}). The population-weighted average value for $A^{0.5}$ for US urban areas is 49 km (US DOT, 2003) (the unweighted average for $A^{0.5}$ is 20 km), and the harmonic mean wind speed in the US is 3.4 m s^{-1} (US EPA, 2002b), indicating that the characteristic residence time of air in a US urban area is ~2–4 h. The lifetime of many air pollutants is much greater than 4 h (Atkinson, 1994). For example, of the 130 toxic air contaminants that have a half-life listed in the California Air Resources Board's contaminants

summary database (www.arb.ca.gov/toxics/tac/txctbl2.htm), 81% have a half-life of more than 10 h.

For a square-plan one-compartment model, the intake fraction of nonreactive pollutant emissions is calculated using Eq. (2) (Lai et al., 2000):

$$iF_{\text{compartment}} = \frac{QP}{uH\sqrt{A}}. \quad (2)$$

Here, $iF_{\text{compartment}}$ is the intake fraction (unitless) estimated by means of the one-compartment model, Q is the population average breathing rate ($\text{m}^3 \text{person}^{-1} \text{s}^{-1}$), P is the population, and H is the atmospheric mixing height (m). Eq. (2) derives from a mass balance. The main assumptions in the derivation are that air in an urban area is well-mixed, and that either the system is at steady-state or concentrations are not strongly correlated over time with breathing rates. We assume here that deposition and chemical reactions occur slowly compared to advection, but it is straightforward to extend the approach to incorporate first-order decay processes (Marshall, 2002).

The variables in Eq. (2) can be clustered into three parameter groups. The first parameter group (which we term “linear population density”), $PA^{-0.5}$, is an

attribute of a city’s urban form, i.e., the way in which the urban area is laid out. The second parameter group (“normalized dilution rate”), uH , is an attribute of the meteorology. Normalized dilution rate ($\text{m}^2 \text{s}^{-1}$) is the volumetric airflow rate out of the basin ($\text{m}^3 \text{s}^{-1}$) divided by air basin width (m). The final parameter is the population average breathing rate, Q .

Linear population density (people m^{-1}) values are calculated from year-2002 population and land area data for the 379 urban areas in the US with more than 50,000 people (US DOT, 2003). The results, shown in Fig. 1, account for 63% of the US population. Harmonic mean normalized dilution rates ($\text{m}^2 \text{s}^{-1}$) are calculated from twice-daily derived values of wind speeds and mixing heights for the 75 meteorological stations in the US EPA’s Support Center for Regulatory Air Models (SCRAM) database (US EPA, 2000, 2002b). Wind speeds in this database are the average speed over the mixing height (US EPA, 2002b). The mean and median values of the meteorological stations’ harmonic mean normalized dilution rate, uH (units: $\text{m}^2 \text{s}^{-1}$), are 610 and 480, respectively.

Using Eq. (2), we combine each of the 379 linear population densities with each of the 75 annual

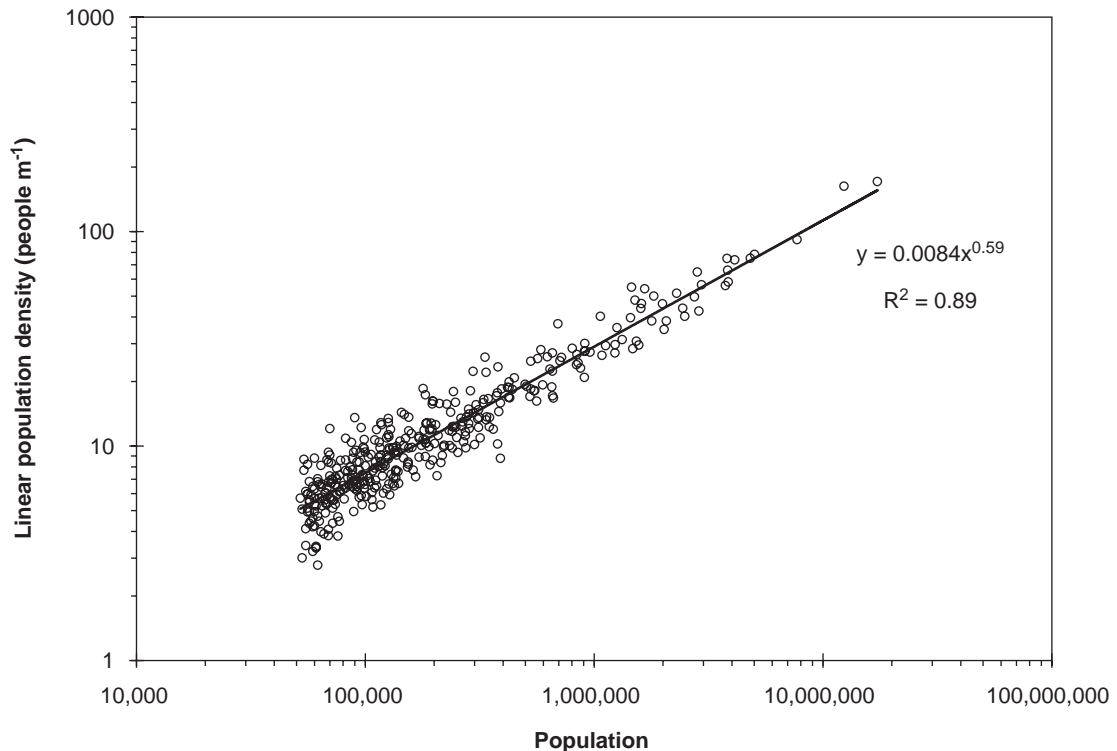


Fig. 1. Relationship between population and linear population density for the 379 urban areas in the US with more than 50,000 people (US DOT, 2003). Linear population density is the population divided by the square root of the land area. Urban areas with large populations tend to have large linear population densities, suggesting that increasing the urban population size increases the intake fraction.

harmonic mean normalized dilution rates, yielding 28,425 estimates of intake fraction. This method of combining the two datasets implicitly assumes that the SCRAM data are representative of meteorological conditions throughout the US and that the DOT data are representative of urban areas throughout the US. Visual inspection of a US map showing the locations of the 75 meteorological stations and the 379 cities did not reveal a systematic location bias.

2.2. Empirical model

The empirical model developed by Glen et al. (1996) estimates ambient concentrations of CO, which is a good tracer for nonreactive vehicle emissions. As a statistical model based on measured concentrations, this approach offers a good complement to the two other methods presented here. Unlike the other approaches, the empirical model focuses explicitly on vehicle emissions, incorporating US EPA's MOBILE5 emission factors (www.epa.gov/otaq/m5.htm). The model offers good predictions of observed data, based on only a few empirically determined parameters.

For the years 1984–1991, Glen et al. (1996) compared monthly average ambient concentrations of CO in 15 US cities with meteorological data and MOBILE5 emission factors. They report the following empirical relationship:

$$C_{i,n} = k_n E_{i,n} \exp \left\{ -\frac{H_{i,n}}{h^*} - \frac{u_{i,n}}{u^*} \right\}. \quad (3)$$

Here, $C_{i,n}$ is the modeled ambient CO mole fraction (ppm) in month i for city n ; k_n is an empirically determined constant (ppm mile g^{-1}) for city n ; $E_{i,n}$ is the average CO emission factor (g mile^{-1}) in month i for city n ; $H_{i,n}$ and $u_{i,n}$ are the average mixing height (m) and wind speed (m s^{-1}), respectively, in month i for city n ; and h^* and u^* are empirically determined constants with units of length (m) and speed (m s^{-1}), respectively, used to make dimensionless the argument in the exponential. They report one value for h^* (1626 m) and for u^* (9.55 m s^{-1}) and the following information for each city: k_n , mean summer and winter wind speed and mixing height, and modeled and measured CO concentration time series.

For each of the 15 cities analyzed by Glen et al., we calculate the winter and summer intake fraction using Eq. (4), which is derived from Eqs. (1) and (3).

$$iF_{\text{empirical}} = \left(\frac{QP}{V} \phi k_n \exp \left\{ -\frac{H_{i,n}}{h^*} - \frac{u_{i,n}}{u^*} \right\} \right) \left(\frac{0.00125 \text{ g}}{\text{m}^3 \text{ ppm}} \right). \quad (4)$$

Here, V is the daily vehicle miles travelled (VMT) in an urban area (mile d^{-1}), ϕ is the fraction of ambient concentrations attributable to motor vehicle emissions

(unitless), and 0.00125 converts the CO mole fraction (ppm) to CO concentration (g m^{-3}). For the years 1984–1991, ϕ is ~ 0.7 (US EPA, 2003). Population and VMT data are taken from US DOT (2003).

2.3. National-scale Air Toxics Assessment

The National-scale Air Toxics Assessment (NATA) estimates year-1996 population inhalation of atmospheric emissions in the US (US EPA, 2002a). To our knowledge, NATA is the most comprehensive national-scale exposure model available. Two main steps within NATA are important here. First, the ASPEN Gaussian plume dispersion model uses meteorological data and the year-1996 National Toxics Inventory to estimate ambient concentrations in all US census tracts. Next, a probabilistic exposure model combines (1) ASPEN-estimated ambient concentrations, (2) time-activity information for 30 hypothetical individuals from each of 10 cohorts (5 age groups, two genders), and (3) estimates of differences between ambient and micro-environment exposure concentrations. The results are summarized as the population average incremental exposure concentration attributable to four source categories (point, area, on-road mobile, and off-road mobile) in two county types (urban and rural). We calculate intake fractions for urban on-road mobile sources based on two conserved pollutants in NATA: benzene and diesel particulate matter.

Intake fraction is calculated from the NATA values using Eq. (5)

$$iF_{\text{NATA}} = \frac{CQP}{E}. \quad (5)$$

Here, C is the mean urban attributable exposure concentration (g m^{-3}), and E is the emission rate from on-road mobile sources (g h^{-1}). Consistent with the EPAs caveat that NATA results are more meaningful when aggregated rather than presented for individual counties, we present here the mean intake fraction among US urban counties. Because the NATA exposure concentrations are the mean values across census tracts, they are approximately population-weighted values. (Census tracts are sized to contain ~ 4000 people each (US Census, 2004).)

3. Results and discussion

3.1. Intake fraction values

Intake fraction values vary among urban areas. The first two methods we use (the one-compartment model and the empirical model) provide information about this variability. Table 1 presents population-weighted and

Table 1

Estimated annual average intake fraction (per million) for urban vehicle emissions using the three methods employed in this work

	One-compartment model	Empirical model, summer	Empirical model, winter	NATA, benzene	NATA, diesel PM
Range of values	0.1–280	5.7–31	7.7–54	—	—
<i>Population-weighted</i>					
Mean	21	10	15	7.0	4.4
Median	12	9.3	13	—	—
Inter-quartile range ^a	5.1–25	8.4–11	11–16	—	—
10%-trimmed range ^b	2.4–50	7.6–15	11–29	—	—
<i>Unweighted</i>					
Mean	5.3	12	19	—	—
Median	3.0	9.3	13	—	—
Inter-quartile range ^a	1.8–5.6	8.2–13	11–24	—	—
10%-trimmed range ^b	1.1–11	6.2–19	11–33	—	—

^aThe inter-quartile range is the range of values excluding the top 25% and bottom 25% of the distribution.

^bThe 10%-trimmed range is the range of values excluding the top 10% and bottom 10% of the distribution.

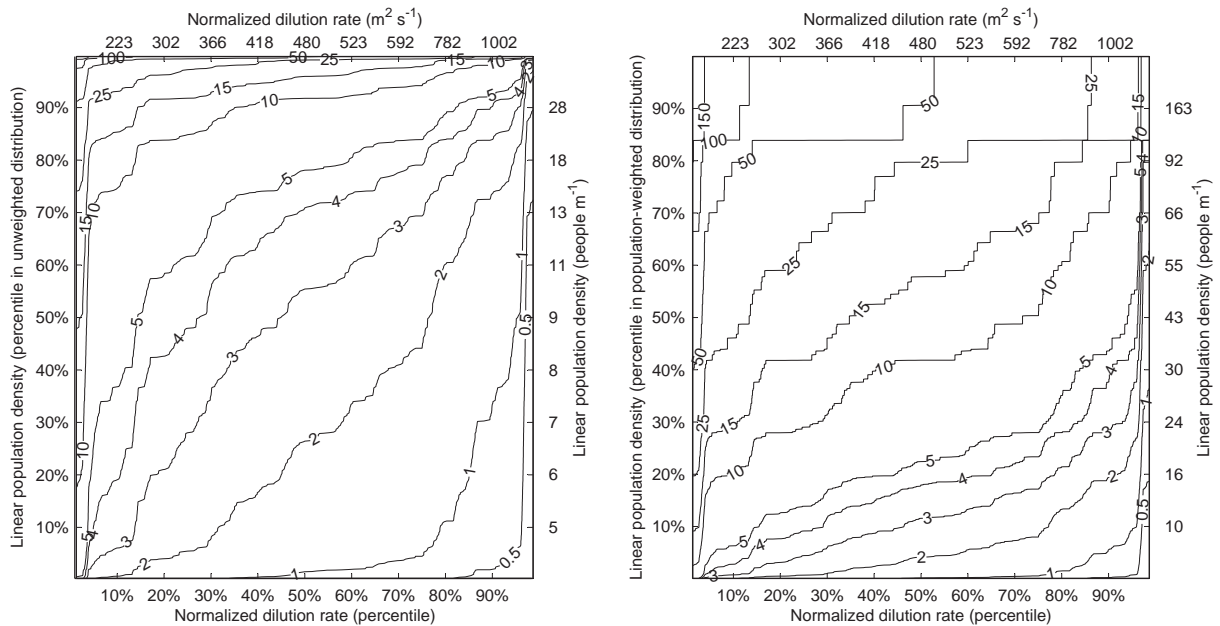


Fig. 2. Isopleths of intake fraction values (per million) for vehicle emissions in US urban areas based on the one-compartment model. In the left figure (the unweighted plot), the linear population density percentile values on the y-axis represent the distribution among urban areas in the US DOT (2003) database. In the right figure (the population-weighted plot), the linear population density percentile values on the y-axis represent the distribution among people in urban areas.

unweighted intake fraction results. The unweighted mean, for example, is the mean value of the intake fraction among urban areas (i.e., giving equal weighting to each urban area). These values are applicable when considering each US urban area as a distinct unit. The population-weighted mean weights the intake fraction value for each urban area based on urban population (i.e., giving equal weighting to each person). These

values are applicable for population-weighted measures including total US urban environments.

Fig. 2 presents isopleths of one-compartment-model-derived intake fraction values as a function of linear population density ($PA^{-0.5}$) and normalized dilution rate (uH). At a given pair of percentiles, intake fractions are larger for the population-weighted values (right plot) than for the unweighted values (left plot). Fig. 3 is a

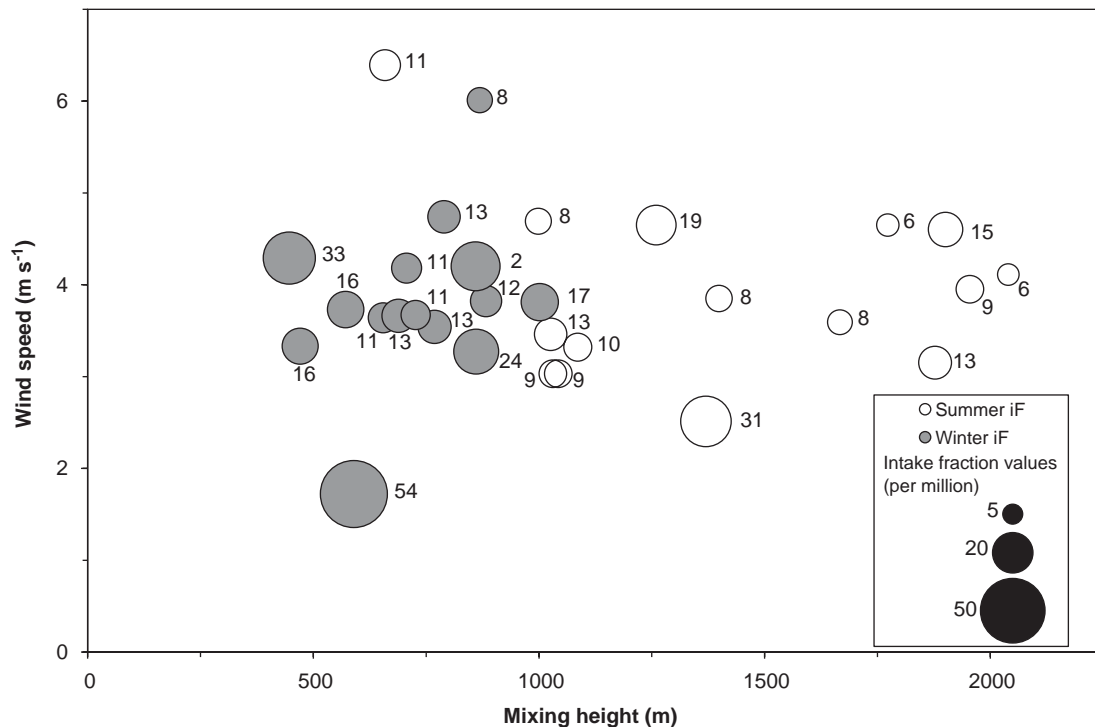


Fig. 3. Summer and winter intake fraction (iF) values for vehicle emissions in the 15 urban areas in the empirical model. The area of each circle is proportional to the intake fraction value.

bubble plot of results from the empirical model, with the icon size proportional to the intake fraction. Each of the 15 cities in the empirical model is represented by two icons (summer and winter).

3.2. Comparisons among the three methods

Intake fraction values calculated by the three methods employed in this paper are consistent with each other. The range of intake fraction values is broader for the one-compartment model than for the empirical approach. One reason for this difference is that the one-compartment approach considers significantly more urban areas than the empirical approach (379 versus 15 urban areas). In addition, as applied here, the one-compartment model estimates values for all combinations of linear population density ($PA^{-0.5}$) and normalized dilution rate (uH), rather than incorporating only the one, true set of meteorological conditions found in any urban area. Hence, it is more appropriate to consider the one-compartment model results presented in this paper in terms of central tendencies (e.g., median and inter-quartile range) rather than for extreme values (e.g., maximum and minimum).

Unweighted intake fraction values for the empirical model are larger than for the one-compartment model

mainly because of differences in urban population size. The mean urban population is 5.5 times larger for the 15 cities in the empirical model than for the 379 urban areas in the one-compartment model (1.2 million versus 220,000 people). The best-fit relationship in Fig. 1 indicates that linear population density is proportional to $P^{0.59}$. Based on this relationship, the population difference of 1.2 million versus 220,000 would yield a factor of 2.7 difference in the linear population density values, and thus, in the mean unweighted intake fraction. Consistent with this expectation, the difference in unweighted mean intake fraction between the empirical model (15 per million, summer and winter combined) and the one-compartment model (5.3 per million) is a factor of 2.8.

For comparison, we also calculated the NATA-derived mean urban intake fraction value for a reactive vehicle emission, 1,3-butadiene (characteristic lifetime ≈ 6 h (US EPA, 1993)). The result, 3.1 per million, is less than the NATA-derived intake fraction for benzene (7.0 per million) because chemical reactions remove a portion of the 1,3-butadiene from ambient air.

Our results support the idea that a one-compartment model can yield reasonably accurate results for investigations of typical intake fraction values. NATA accounts for several factors that the one-compartment model does not, such as concentration differences in

microenvironments and spatial heterogeneities in emissions and ambient concentrations. The more sophisticated approach employed by NATA allows it to address questions the one-compartment model cannot. Nevertheless, the two approaches yield similar results for the primary research question considered in this work.

3.3. Intake fraction in urban areas not studied in this work

One method for estimating the intake fraction in a specific urban area would be to scale our results up or down based on the linear population density. For example, based on the one-compartment model results, when considering an urban area with a linear population density that is two times greater than the US DOT (2003) median value of $9.5 \text{ people m}^{-1}$, the intake fraction would be estimated as ~ 6 per million (i.e., two times greater than the unweighted one-compartment model median intake fraction value of 3.0 per million). For the South Coast Air Basin (linear population density $\approx 120 \text{ people m}^{-1}$), this approach suggests a value of 38 per million, which is close to the published value of 48 per million (Marshall et al., 2003). If the linear population density is not known, the intake fraction for an urban area can be approximated from the urban population (P) using the following relationship: intake fraction $\approx 0.0025 P^{0.59}$, where intake fraction is in units of per million. This relationship combines Eq. (2), the empirical relationship in Fig. 1, a breathing rate of $12.2 \text{ m}^3 \text{ d}^{-1} \text{ person}^{-1}$, and a normalized dilution rate of $480 \text{ m}^2 \text{ s}^{-1}$.

3.4. Uncertainty

We discuss here uncertainty in the input parameters and in the methods used. For the one-compartment and empirical models, method uncertainty is expected to be larger than input uncertainty. While rigorous uncertainty bounds are not known for the input parameters used in these two approaches, most of the input data (e.g., population, land area, wind speed) have relatively tight confidence intervals. (An exception is the vehicle CO emission factor, for which the uncertainty is a factor of ~ 2 (Singer and Harley, 1996).) These two methods do not account for differences between ambient and exposure concentrations, such as those occurring while traveling in a vehicle, nor do they incorporate emissions' spatial and temporal variability. To the extent that such factors increase the estimated inhalation intake rate, these two methods may underestimate the true intake fraction value. The NATA approach accounts for these two factors, which reduces method uncertainty, but in doing so, it increases both the number of input parameters and the input uncertainty. Most of the

uncertainty information that NATA provides is qualitative. One exception is uncertainty in modeled ambient concentrations: comparisons between modeled and measured concentrations indicate that the model under-predicts ambient concentrations by $\sim 40\%$ for conserved gases and by a factor of ~ 5 for particles (US EPA, 2002a). Because intake fraction incorporates the ratio of concentrations to emissions, differences between measured and modeled concentrations may or may not lead to errors in the NATA-derived intake fraction estimates presented in this work. If these differences were attributable to the air dispersion model, then the NATA-derived intake fraction values presented in this work would be too low by $\sim 40\%$ for conserved gases and by a factor of ~ 5 for particles. In contrast, if these differences were attributable to errors in the emission inventory, then they would not indicate errors in the NATA-derived intake fraction values. The EPA considers the latter case to be more likely than the former (US EPA, 2002a).

Comparing results among the three methods provides information about the overall method uncertainty. Combining results from the three methods with equal weight, the mean population-weighted annual average intake fraction for conserved gaseous vehicle emissions is estimated to be ~ 14 per million. The mean values from the three methods (excluding diesel PM because it is not a nonreactive gas) are within $\sim 50\%$ of this average. Thus, we estimate method uncertainty to be roughly 50%. Further investigations, applying additional methods to urban areas throughout the US, are necessary to confirm the intake fraction results and the uncertainty estimates presented in this work.

Total intake for urban emissions is the sum of the intakes within and downwind of the urban area (Marshall, 2005). This work only quantifies intraurban intake. Previous work indicates that for urban vehicle emissions, the downwind intake may either be small relative to, or be comparable to, intraurban intake. Not enough work has yet been done to arrive at firm conclusions. Evans et al. (2002) found that 50% of the total inhalation of urban highway emissions occurs within 100 km of the source. Greco et al. (2004) found that 41% of people in the US live in counties where a majority of the total inhalation intake of mobile source emissions occurs within their county borders. Investigating vehicular emissions into California's South Coast air basin, Marshall et al. (2003) found that the downwind increment of intake was a few orders of magnitude smaller than the intake within the air basin.

3.5. Seasonal variability

We investigate seasonal variability for the 15 cities in Glen et al. (1996) and for six cities from the SCRAM

database: Waycross, Georgia; Denver, Colorado; Atlantic City, New Jersey; Oakland, California; Peoria, Illinois; and Tucson, Arizona. These six cities were chosen to span a range of climates throughout the US. Following the approach by Glen et al., we group the SCRAM data into four summer months (May–August) and four winter months (November–February).

The analysis reveals that intake fraction values are, on average, higher in winter than in summer. The median intake fraction value calculated using the empirical model is ~40% larger in winter than in summer. (One city of the fifteen in the empirical model does not follow this trend: the calculated intake fraction for Buffalo, New York, is essentially the same in summer as in winter.) For five of the six SCRAM stations, the average normalized dilution rate is greater in summer than in winter, causing the calculated intake fraction to be 50–200% larger in winter than in summer. (At the sixth station, Peoria, Illinois, the normalized dilution rate is 80% larger in winter than in summer.) These estimates are consistent with the finding of Marshall et al. (2003) that the vehicle intake fraction in the South Coast Air Basin is ~2 times larger in winter than in summer.

For these 21 cities, seasonal variability in the calculated intake fraction is more attributable to changes in mixing height than to changes in wind speed (see Fig. 3). On average, mixing heights are 90% higher in summer than in winter. This seasonal trend, mixing heights being higher in summer than in winter, occurs in all 21 cities. In contrast, wind speeds for the 21 cities change by an average of 20% between summer and winter. For 10 of the 21 cities, wind speed is larger in summer than in winter; for the remaining 11 cities, the reverse is true.

4. Conclusion

We have used three independent methods to characterize intake fraction for nonreactive vehicle emissions in US urban areas. These three methods incorporate empirical results and models with different levels of sophistication. Intake fraction varies among locations, based on factors such as meteorology, linear population density, and the spatial distribution of emissions. Population-weighted annual-average mean intake fractions for nonreactive gaseous vehicle emissions in US urban areas are estimated to be ~14 per million, with an uncertainty of approximately 50%. Intraurban removal mechanisms, such as chemical reactions (as for 1,3-butadiene) and physical removal as air migrates from outdoors to indoors (as for diesel PM), reduce the intake fraction. Seasonal average intake fractions are usually higher in winter than in summer, owing primarily to changes in atmospheric mixing height.

Intake fraction is a useful metric for health risk assessments, cost–benefit analyses, and other investigations that require a summary of the emission-to-intake relationship. Earlier work (Bennett et al., 2002; Evans et al., 2002; Lai et al., 2000; Marshall et al., 2003) highlighted the merits of compiling intake fraction values and methods for various sources and pollutants. This paper contributes to that goal.

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