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Intake Fraction of Primary Pollutants: Motor Vehicle Emissions in the South Coast Air Basin

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Abstract

The intake fraction is defined for a specific species and emission source as the ratio of attributable population intake to total emissions. Focusing on California's South Coast Air Basin (SoCAB) as a case study, we combine ambient monitoring data with time-activity patterns to estimate the population intake of carbon monoxide and benzene emitted from motor vehicles during 1996 – 1999. In addition to exposures to ambient concentrations, three microenvironments are considered in which the exposure concentration of motor vehicle emissions is higher than in ambient air: in and near vehicles, inside a building that is near a freeway, and inside a residence with an attached garage. Incorporating data on motor vehicle emissions estimated by the EMFAC2000 model, we estimate that the 15 million people in the

SoCAB inhale 0.003 – 0.009% (34 – 85 per million with a best estimate of 47 per million) of primary, nonreactive compounds emitted into the basin by motor vehicles. This population intake of primary motor vehicle emissions is approximately 50% higher than the average ambient concentration times the average breathing rate, owing to higher concentrations in the three microenvironments and also to the temporal and spatial correlation among breathing rates, concentrations, and population densities. The approach demonstrated here can inform policy decisions requiring a metric of population exposure to airborne pollutants.

Key words: exposure assessment, microenvironment, carbon monoxide, benzene.

1. Introduction

Motor vehicle emissions influence local, regional, and global air quality. In addition to their contributions to photochemical smog and its components, such as ozone and NO_x, motor vehicles also contribute significantly to ambient concentrations of hazardous and US EPA criteria air pollutants. In the United States, on-road motor vehicles account for 48% of benzene emissions and 51% of carbon monoxide (CO) emissions (EPA, 2001b). In California's South Coast Air Basin (SoCAB), on-road motor vehicles contribute 70% and 80%, respectively, of total benzene and CO emissions (CARB, 2000b; SCAQMD, 2000).

Previous investigations have highlighted motor vehicles as an important source of population exposure to benzene and CO (e.g., Duarte-Davidson et al., 2001; Fruin et al., 2001; Gonzalez-Flesca et al., 2000; Law et al., 1997; Macintosh et al., 1995; SCAQMD, 2001). For example, Macintosh et al. (1995) developed a probabilistic, multipathway (inhalation, ingestion, and dermal absorption) benzene exposure and dose model. They applied this model to Arizona and EPA Region 5. For nonsmokers, they reached two main conclusions. First, population exposure to benzene is "predominantly a function of the outdoor source component of indoor air benzene levels rather than indoor source-related exposures." Second, uncertainty in the total dose is mainly due to uncertainty in benzene concentrations rather than to variability in time-activity patterns. Fruin et al. (2001) combined ambient concentration data with time-activity patterns in 14 microenvironments to assess exposure to benzene in California's South Coast Air Basin (SoCAB). They show that the average benzene level at which nonsmoking adults are exposed decreased from 6 ppb in 1989 to 2 ppb in 1997. They attribute this rapid decrease to comparable

changes in ambient concentrations, as well as decreased exposure to environmental tobacco smoke. In a literature review on population exposure to CO from mobile sources, Flachsbart (1999) reported that CO exposures in the US are decreasing owing to reductions in mobile source emissions. He pointed out that because CO is a nonreactive gas, it penetrates building envelopes without loss. If there are no indoor sources, the average indoor concentration will equal the average outdoor concentration (Flachsbart, 1999; Ott et al., 1992).

In this report, we use a recently named exposure metric, the intake fraction (iF), to characterize the emissions-to-intake relationship for the inhalation of primary pollutants from motor vehicles. The iF is the ratio of the total population intake of a pollutant to the total emissions (i.e., the fraction of emissions that are taken in by people). Intake fraction summarizes complex emissions, fate, transport, and exposure relationships in a single number that is easy to use and understand. Because iF is a metric rather than a method, it can be calculated using models, measurements, or both, and it is equally amenable to back-of-the-envelope estimates as to sophisticated analyses. Bennett et al. (2002) and Evans et al. (2002) summarize previous intake fraction research and discuss the motivation for using iF to characterize exposures.

Our investigation characterizes the intake fraction of benzene and CO from motor vehicles in the South Coast Air Basin (Fig. 1) during 1996 – 1999 (inclusive). To our knowledge, no published report has analyzed ambient concentration data to quantify the intake fraction. Two previous investigations have quantified the intake fraction for motor vehicles based on air dispersion modeling. Evans et al. (2002) used a trajectory model, with 448 grid cells of 10,000 km² each, to calculate intake fractions for motor vehicle emissions on 40 highway segments throughout the United States. For primary PM_{2.5}, they report intake fractions of 3-18 per million for urban locations and 1-18 per million for rural locations. Nigge (2001) combined two air dispersion models to calculate intake fractions of nine primary pollutants from point sources in Germany. For short-range transport (within 100 km), he used a Gaussian plume model. For long-range transport (greater than 100 km) he used a trajectory model with 10,000-km² grid cells. Intake fraction results are presented by Nigge for three pollutants: acetaldehyde (3 – 14 per million), PM_{2.5} (8 – 18 per million), and PM₁₀ (3 – 12 per million). These results, which Nigge argues are applicable to motor vehicles, are similar to those of Evans et al. (2002). In contrast with these two studies, we estimate exposures based on ambient monitoring data, and we explicitly include near-source exposures. Our research focuses on an urban area (17,460 km²)

that would occupy less than two grid cells in the trajectory models employed by Evans et al. (2002) and Nigg (2001).

2. Methods

2.1. Intake Fraction

Primary pollutants are those that are emitted directly, rather than being formed by reactions of precursor emissions. For inhalation of a primary pollutant, the intake fraction (*iF*) can be expressed as:

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

Here, T_1 and T_2 are the starting and ending times of the emission (s); P is the number of people in the exposed population; $Q_i(t)$ is the breathing rate for individual i at time t ($\text{m}^3 \text{s}^{-1}$); $C_i(t)$ is the incremental concentration at time t in the breathing zone of individual i that is attributable to a specific source ($\mu\text{g m}^{-3}$); and $E(t)$ is that source's emissions at time t (g s^{-1}). In practice, the integral in the numerator is evaluated until the incremental concentration attributable to the source of interest is negligibly small. For exposures in an urban air basin, the integration time scale need only be much longer than the time scale for pollutant transport through an urban air basin, which is typically less than a day.

Intake fraction is a dimensionless number ranging from zero, which would indicate that no emissions are inhaled, to one, which would indicate that all emissions are inhaled. An *iF* of one per million means one mg of pollution is inhaled for every kg of pollution emitted. Stated differently, an *iF* of one per million means each molecule emitted to the environment has a one per million chance of being inhaled. While this paper focuses on population inhalation of atmospheric emissions, the intake fraction metric can be applied to individuals or subpopulations, and it can be applied to multipathway, multimedia exposure assessments.

The *iF* depends on factors such as source type (e.g., indoor versus outdoor, urban versus rural) and pollutant fate and transport (e.g., reaction and removal rates, importance of multi-

media, multi-pathway exposures) (Bennett et al., 2002; Evans et al., 2002; Lai et al., 2000). Two pollutants emitted from the same source with identical fate and transport characteristics will have identical intake fractions. Analogously, two pollutants from the same *type* of source with *similar* fate and transport characteristics will have *similar* intake fractions. The *iF* of a non-reactive pollutant from a given source is expected to evolve more slowly under many circumstances than the rate of emissions from that source. For example, a technology shift such as fuel reformulation may alter emissions without significantly altering *iF*.

Our method for calculating the intake fraction (Eq. 1) requires information on four space- and time-dependent factors: emissions, population size, population breathing rate, and attributable exposure concentration. Each of these parameters is discussed below. If there were no spatial or temporal variability in the attributable exposure concentration, the intake fraction could be computed as the product of the population size, the average breathing rate, and the average exposure concentration attributable to a specific source, divided by the total emission rate for that source. However, a more detailed analysis is required for two reasons. First, publicly available concentration data comes from monitoring stations that record ambient concentrations rather than exposure concentrations. Second, spatial and temporal correlations among population density, breathing rates, and concentrations may alter the actual population intake relative to that determined from combining average values (Hayes and Marshall, 1999).

2.2. Emissions

Emissions data for the SoCAB, shown in Fig. 2, are based on the California Air Resources Board's (CARB) EMFAC database and model (CARB, 2000a). We employed the 2000 version of EMFAC, which combines emission factors and a motor vehicle emission inventory (MVEI7G) to calculate evaporative and exhaust emissions from on-road mobile sources. EMFAC databases include monthly estimates of vehicle-miles traveled and of the age distribution of the vehicle fleet. Exhaust emissions are estimated from dynamometer tests, which are run according to Federal Testing Procedure (FTP) protocols, and from CARB's database of time spent in various operating modes, such as idling, accelerating, and startup. Evaporative emissions include drips, leaks, and "breathing losses" that result from heating and cooling of the gas tank and the engine. Benzene is present in both evaporative and exhaust emissions, because it is a constituent of gasoline and also a product of incomplete combustion. Carbon monoxide is

formed by incomplete combustion and is only present in exhaust emissions. EMFAC directly estimates CO and total organic gas (TOG) emissions; it does not differentiate among the hydrocarbons that make up TOG emissions. We calculate benzene emissions by applying data from recent tunnel studies conducted in northern California, which indicate that benzene comprises 3.3% of the TOG from exhaust emissions and 0.5% of the TOG from evaporative emissions (Kirchstetter et al., 1999a; Kirchstetter et al., 1999b).

2.3. Population Size and Breathing Rate

The SoCAB occupies 17,460 km² and is home to 15 million people (CARB, 2002b), so the average population density is 860 km⁻².

Using an approach based on metabolic activity (Layton, 1993), the population average breathing rate is estimated to be 12.2 m³ d⁻¹. This estimate, which incorporates information about the age distribution of the US (Census, 2001), represents the average breathing rate for men, women, and children. In contrast, risk assessments typically use a higher breathing rate (e.g., 20 or 25 m³ d⁻¹) to provide conservative intake estimates allowing for interindividual variability (EPA, 1997).

Layton (1993) gives breathing rates for five activity levels (sleep, light, moderate, hard, very hard) and the number of hours per day spent in each of those activity levels. As population breathing rates are not available as a function of time, we allocated these data to each hour of the day (Fig. 3) based on our own assumptions about the likelihood that each activity level will occur during each hour. If more detailed information about population breathing rates becomes available in the future, we would be able to refine our calculations.

2.4. Attributable Exposure Concentration

We estimate attributable exposure concentrations from ambient concentrations, the time spent in specific microenvironments (i.e., time-activity patterns), and the exposure concentration associated with these microenvironments. These three parameters are discussed in the following subsections. We consider microenvironments because exposure concentrations can be higher than ambient concentrations when a person is in close proximity to motor vehicle emissions.

2.4.1. *Ambient Concentrations.*

The South Coast Air Quality Management District (SCAQMD) measures and records ambient pollutant concentrations at 34 air quality monitoring stations distributed throughout the SoCAB. During 1996 – 1999, 20 of these stations recorded one-hour average CO concentration every hour. Six stations recorded 24-hour average benzene concentration approximately twice per month. Additional information on the ambient concentration data is given in Table 1.

Monthly average ambient concentrations are shown in Fig. 4.

Our method for population-weighting the ambient concentration data involves two steps. First, we assign an ambient concentration to each census tract by weighting monitoring station data according to the inverse square of the distance between the census block centroid and each monitoring station. We then use year-2000 population data for each census tract to yield population-weighted ambient concentrations.

We tested several methods of accounting for nondetect values. For both CO and benzene, none of the methods changed the mean concentration significantly because (1) the data have a small fraction of nondetects and (2) the detection limit is small relative to the average measured values (Table 1). We decided to assign a concentration of zero to nondetect values. (As a comparison, if we had assigned 50% of the detection limit to nondetect values, the increase in the mean concentration is negligible: 1.0% and 0.2% for benzene and CO, respectively.)

Because hourly ambient concentrations are available for CO but not benzene, we estimate hourly ambient benzene concentrations by applying the characteristic daily profile for ambient CO concentrations in each month and year to the 24-hour average ambient benzene concentration (Fig. 5). We assume that benzene and CO exhibit similar daily profiles. This assumption is expected to be approximately true, since both CO and benzene are emitted by motor vehicles, although differences will exist because CO comes from exhaust emissions while benzene comes from both exhaust and evaporative emissions. Evaporative benzene emissions peak during hot afternoons, while CO emissions peak during “cold start” conditions on cold mornings. More detailed measurements of hourly ambient benzene concentrations would permit refinement of this calculation, but are not expected to change the results markedly.

2.4.2. *Time-Activity Patterns*

Time-activity patterns indicate how much time is spent in various microenvironments. We examined four microenvironments: in a vehicle; in a residence with an attached garage; in a

building near a freeway; and all other indoor and outdoor locations. We used results from the National Human Activity Pattern Survey (NHAPS) (Klepeis et al., 2001) to examine three of the four microenvironments (in vehicle, in a residence with an attached garage, and all other locations). In a separate analysis, we account for exposures in indoor locations immediately downwind of a freeway.

For the first microenvironment, we used data for the NHAPS category “in/near vehicle.” This category includes any outdoor activity that takes place inside or near a transportation vehicle, such as riding in a vehicle, waiting for a bus, train, or automobile, and walking on a sidewalk. For the second microenvironment, we combined an estimate for the Los Angeles-Long Beach Metropolitan Area that ~ 60% of people live in a house with an attached garage (HUD, 2001) with NHAPS data on time spent in a residence. All other time was allocated to the third microenvironment, which includes both outdoor (not in or near a vehicle) and indoor (without an attached garage) locations. Of the 1.30×10^{20} person-hours y^{-1} available to SoCAB residents, 7% is spent in/near vehicles, 41% is spent inside a residence with an attached garage, and the remaining 52% is spent elsewhere. Other microenvironments that have been used in benzene and CO exposure assessments, such as houses with natural gas cook stoves or bars, do not need separate consideration to study exposure only to motor vehicle emissions (Fruin et al., 2001; Macintosh et al., 1995; Ott et al., 1992).

2.4.3. *Microenvironment Concentrations.*

The estimated increase in concentration relative to the ambient concentration is discussed below for each of the four microenvironments. Attributable exposure concentrations are calculated as follows:

$$C_{\mu} = \phi C_{amb} + (\gamma_{\mu} - 1) C_{amb} \quad (2)$$

Here, C_m is the exposure concentration ($\mu\text{g m}^{-3}$) attributable to motor vehicle emissions in microenvironment m ; C_{amb} is the ambient concentration ($\mu\text{g m}^{-3}$); f is the fraction of ambient concentrations attributable to motor vehicles; and g_m is the ratio of concentration in microenvironment m (owing to ambient sources) to the ambient concentration. For the SoCAB, f is 70% for benzene and 80% for CO (CARB, 2000b; SCAQMD, 2000). Note that f incorporates

two factors: the fraction of ambient concentrations attributable to local emissions and the fraction of local emissions attributable to motor vehicles. As there are no population centers immediately upwind of the South Coast, all ambient concentrations are assumed attributable to local emissions. Values for g_m are given below and summarized in Table 2. For this study g_m is always greater than or equal to one. For situations in which g_m is less than one (e.g., to account for indoor concentrations of particulate matter being less than ambient concentrations), the term $(g_m I)C_{amb}$ in Eq. (2) would need to be replaced with $(g_m I)F C_{amb}$.

2.4.3.1. *In and Near Vehicles*

The published literature contains many data sets of in- and near-vehicle concentration measurements for carbon monoxide and benzene. Our review of twenty-five reports and journal articles on concentrations of motor vehicle pollutants inside motor vehicles indicates a high degree of variability. In-vehicle concentrations depend on many factors, including meteorological conditions, traffic density and speed, and emission rates from neighboring cars (Alm et al., 1999; Chan et al., 1991a; Chan et al., 1991b; Conceicao et al., 1997; EPA, 1998; EPA, 2001a; Fernandez-Bremauntz and Ashmore, 1995a; Fernandez-Bremauntz and Ashmore, 1995b; Flachsbart, 1995; Flachsbart, 1999a; Flachsbart, 1999b; Jo and Park, 1998; Jo and Park 1999; Johnson, 1995; Koushki et al., 1992; Lawryk et al., 1995; Macintosh et al., 1995; McCurdy, 1995; Park et al., 1998; Rhodes et al., 1998; Wallace, 1990; Wallace, 1991; Wallace, 1996; Weinhold, 2001; Weisel et al., 1992). Several of these studies report both in-vehicle and ambient concentrations. Across many cities and over several years of data with differing levels of ambient air pollution, typical in-vehicle CO and benzene concentrations are roughly four times greater than ambient concentrations (Flachsbart, 1995; Flachsbart, 1999b; Rhodes et al., 1998; Wallace, 1996), leading us to adopt $g_m = 4$ for the in- and near-vehicle microenvironment.

2.4.3.2. *Concentrations in Residences with an Attached Garage*

In an enclosed garage, evaporative emissions lead to higher concentrations of benzene but not CO. In a residence with an attached enclosed garage, these evaporative emissions can migrate into the household via airflow coupling between the garage and living space (CMHC, 2001; Wallace, 1990). To our knowledge, no experimental study has investigated long-term elevations in population exposure to motor vehicle emissions due to attached garages. By analyzing the limited data available, we estimate that residences with an attached garage have vehicle-associated benzene concentrations that are ~ 20% higher than the ambient counterparts

(Fruin et al., 2001; Macintosh et al., 1995; Thomas et al., 1993). On the other hand, we estimate that motor vehicles cause no significant enhancement of CO concentrations in houses (with or without an attached garage) above the local ambient concentration (Flachsbart, 1999; Ott et al., 1992). During the several hours people spend at home each day there may be sustained in-garage evaporative emissions of benzene but not CO. Thus, in residences with attached garages $g_m = 1.2$ and 1.0 for benzene and carbon monoxide, respectively.

2.4.3.3. *Indoor Concentrations near Freeways*

We analyze time spent indoors near freeways separately because this microenvironment is not included in the NHAPS data. Our approach combines three pieces of data: the distance downwind of a freeway in which the observed concentration is significantly elevated because of local emissions; the fraction of the population present within that distance; and the concentration in this microenvironment.

Using a tracer-gas approach, Drivas and Shair (1974) found that concentrations of pollutants emitted from a roadway were elevated over a distance less than 100 m downwind. This result agrees broadly with the Gaussian plume dispersion equation for a line source (Nazaroff and Alvarez-Cohen, 2001), which indicates that the impact distance is typically less than 300 m. Both of these analyses assumed that the wind is perpendicular to the freeway. Since all other wind directions will result in lower values for this characteristic distance, ~ 200 m represents a reasonable upper bound for the average characteristic distance. This distance is consistent with an epidemiological study by Wilhelm and Ritz (2003) that used 229 m (750 feet) buffers around subject homes to assign distance-weighted traffic density values.

Combining this 200 m characteristic distance with the length of freeways in the SoCAB (316 km (Bhat, 2001)) yields 660 km² of “near-freeway” land, or 4% of the total area of the SoCAB. For this portion of the analysis, we assume that the population density is uniform throughout the basin, and therefore ~ 4% of the people in the SoCAB are in buildings near freeways at any given time. Although there are major roads in the SoCAB that are not freeways, we have not accounted for them explicitly in this analysis because their impact on concentrations is reflected in the ambient concentration data. That is, we assume monitoring station data adequately capture typical outdoor concentrations except for locations immediately downwind of freeways.

We estimate that average CO and benzene concentrations within 200 m downwind of a freeway are twice the ambient concentration not near a freeway ($g_m = 2$). This estimate is based on data showing in-vehicle concentrations as four times ambient concentrations and incorporates a Gaussian-plume approach to account for the rapid decrease in concentration immediately downwind owing to atmospheric dispersion.

2.4.3.4. Concentrations in Other Locations.

In all locations other than the three microenvironments above, the attributable exposure concentration is assumed to equal the attributable ambient concentration. Both benzene and CO are relatively nonreactive gases, and outdoor concentrations readily penetrate into indoor environments without loss (Flachsbart, 1999; Ott et al., 1992). Indoor environments may have additional sources of benzene or CO, such as gas stoves or cigarette smoke, but the existence of these sources does not alter exposure to motor vehicle emissions.

3. Results

3.1. Intake Fraction Within the SoCAB

Fig. 6 summarizes the attributable exposure concentrations that we have estimated by combining ambient concentration measurements, time-activity patterns, and relative increases in exposure concentrations associated with microenvironments. Emissions are relatively constant throughout the year (Fig. 2). However, ambient and exposure concentrations of CO and benzene (Figs. 4 and 6) are about twice as high in winter as in summer. The varying concentration-to-emissions ratio generates a similar seasonal pattern in the intake fraction. As is shown in Fig. 7, the intake fractions for motor vehicle emissions of CO and benzene are about two times higher in winter than in summer. This variability is a consequence of varying seasonal meteorological patterns. Atmospheric transport and dispersion are slower on average during the winter because of the weaker incident solar radiation. Poorer pollutant transport means that the same emissions of primary pollutants will lead to higher attributable concentrations and a higher intake fraction.

We estimate annual average intake fractions for SoCAB motor vehicle emissions to be 46 per million for CO and 48 per million for benzene. These estimates indicate that ~ 50 grams of primary, nonreactive motor vehicle pollutants are inhaled for every million grams of pollutants emitted. The intake fraction for benzene is slightly higher than for CO due to the slightly

increased exposures from attached garages, but this difference is small compared to the seasonal variability for both benzene and CO. These iF values aggregate over all motor vehicles. The iF for emissions from specific vehicles are variably distributed about this mean, depending on factors such as the meteorology and the time and location of emissions. Using 48 months of data, with a single intake fraction calculated for each month, we calculate standard deviations of 15 per million for CO and 20 per million for benzene. These standard deviations indicate variability in the monthly mean iF from the annual-mean value.

Note that we have used a population breathing rate of $12.2 \text{ m}^3 \text{ day}^{-1}$ rather than the adult breathing rate of $19 - 20 \text{ m}^3 \text{ day}^{-1}$ used in most previous intake fraction research (e.g., Lai et al., 2000; Nigge, 2001; Evans et al., 2002). If we were to use a breathing rate of $20 \text{ m}^3 \text{ day}^{-1}$, our results would increase to 76 per million and 79 per million for CO and benzene, respectively.

As a comparison with our main intake fraction estimate of ~ 50 per million, we performed a second analysis using the average attributable ambient concentration as a surrogate for the attributable exposure concentration. For this simplified analysis, we ignored spatial and temporal variability and computed the intake directly as the product of the monthly average ambient concentration, the fraction of emissions attributable to motor vehicles, the population size, and the monthly breathing rate per person, divided by the estimated pollutant emission rates from motor vehicles. The input data and results for this calculation are shown in Table 3. The intake fractions estimated by this approach are ~ 30 per million for CO and benzene, about a third less than obtained by the more detailed analysis.

3.2. Intakes in Downwind Air Basins

Exposures are not confined to the air basin in which emissions occurred. We used a one-box model to estimate exposures occurring outside the SoCAB that are attributable to motor vehicle emissions within that air basin (Marshall et al., 2002). We modeled both a conserved pollutant and a hypothetical decaying pollutant with a lifetime of 80 hours, and we considered both downwind regional and national exposures. Regional exposures are evaluated based on population intake in the two air basins to the east of SoCAB (the Salton Sea Air Basin and the Mojave Desert Air Basin). Combining the regional and nationwide intakes, we estimate an additional intake fraction increment of $0.08 - 0.2$ per million for a reactive pollutant and $0.2 - 0.7$ per million for a nonreactive pollutant. These results are $70 - 600$ times less than the

estimates of within-basin intake. Consequently, we conclude that for the case being studied, regional and national intake increments of primary and reactive pollutants are significantly less than within-basin intakes of urban emissions. (For comparison, the one-box model was also used to predict the intake fraction for within-basin exposures. The results are in the range 10 – 80 per million, which bracket the value of ~ 50 per million obtained by the more detailed assessment.)

3.3. *Uncertainty in the Estimates*

Errors in our results may arise from errors in our inputs and from errors in the method employed. We address the former issue in this section in terms of the four main inputs (concentrations, emissions, breathing rates, and population). The latter issue is explored in the discussion.

With the exception of monitoring data, uncertainty bounds have not been reported for most of the data used here. During the years considered (1996 – 1999), audits of monitors throughout California yielded an average percent difference between the calibration sample and the monitor's measurement of 0.5% and -11% for CO and benzene, respectively (CARB, 2001b; CARB, 2002a). These audits indicate that CO monitors have a high degree of accuracy while benzene monitors tend to underestimate the true concentration.

A comparison between EMFAC and a fuel-based emission inventory (Singer and Harley, 2000) suggests EMFAC may underestimate emissions by ~ 20%. In contrast, recent updates to EMFAC suggest the emissions may be overestimated by ~ 30% (CARB, 2001a; CARB, 2002c). Among the four main inputs, the emissions inventory is the most uncertain.

Confidence intervals were not provided for population and breathing rate data. We estimate uncertainty in the Census population data is ~ 3% or better, and uncertainty in the breathing rate data is ~ 8% or better.

Based on these uncertainty ranges for the four main inputs we arrive at the following determinations for motor vehicle emissions in the SoCAB. The intake fraction for CO is likely to be in the range of 34 – 73 per million. The intake fraction for benzene is likely to be in the range of 36 – 85 per million. These ranges represent bounding estimates (i.e., they assume errors in our inputs are aligned to yield maximum error in our outputs), which are likely to overestimate uncertainty in our results. The CO results are somewhat more certain than the benzene results because of greater accuracy in the ambient concentration data. Combining the results and

uncertainties for CO and benzene, we conclude that the annual average intake fraction for nonreactive primary pollutants from motor vehicles in the South Coast Air Basin is likely to be in the range 34 – 85 per million, with a best estimate of 47 per million.

4. Discussion

Our findings are consistent with limited prior research. Previous studies have used methods other than the one presented in this paper to characterize the emissions-to-concentration or emissions-to-intake ratio. Based on previous studies, one would expect the intake fraction for an outdoor release in an urban area to be on the order of 1 – 100 per million. For example, using Gaussian plume equations, Lai et al. (2000) calculated an iF of 4 – 230 per million for outdoor sources, depending on the meteorology, population density, and urban area. Smith (1993) reported 20 per million as an order-of-magnitude estimate for outdoor ground-level emission sources in urban settings. Evans et al. (2002) and Nigge (2001) modeled both urban and rural emissions in the US and Germany, respectively, and reported intake fractions of 1 – 18 per million for motor vehicles. Evans et al. (2000) used a Gaussian plume model to calculate an intake fraction of 6 – 22 per million for ambient emissions from dry cleaners in the US. Schauer et al. (1996) reported a value of $0.4 (\mu\text{g m}^{-3}) \text{ per } (\text{t d}^{-1})^{-1}$ for the ratio of attributable ambient concentration to emissions for elemental carbon from diesel exhaust in downtown Los Angeles. Applying an inhalation rate of $12.2 \text{ m}^3 \text{ day}^{-1}$ and a population of 7 million for the $\sim 1600 \text{ km}^2$ downtown region yields an iF of about 34 per million for the local impacts of this urban emission source. A study of Taipei City, Taiwan, presented modeled and measured ambient CO concentrations of 1.1 ppm, a population of 2.6 million people, and CO emissions – over 99% of which are from motor vehicles – of 400,000 tonnes y^{-1} (Chen et al., 2002). Using a breathing rate of $12.2 \text{ m}^3 \text{ day}^{-1}$, their results indicate an iF of 39 per million. Consistency between previous findings and the results presented here substantiates the accuracy of our results and reinforces the validity and potential utility of the intake fraction concept.

Similarly, the close agreement between the intake fractions for benzene and CO also substantiates the value of the intake fraction metric. Carbon monoxide and benzene from motor

¹ Owing to a typographical error, the units are given in the publication as $(\mu\text{g m}^{-3}) \text{ per } (\text{kg d}^{-1})$ (Schauer 2001).

vehicle emissions are expected to have similar intake fractions because they have similar fate and transport characteristics in the atmosphere. The dominant removal mechanism from the air basin for nonreactive gases is advection, and nonreactive gases penetrate building envelopes without impedance or removal. We characterize CO and benzene as relatively nonreactive because their characteristic lifetimes in urban atmospheres (~ 900 h for CO (CARB, 1999) and ~ 500 h for benzene (EPA, 1993)) are significantly greater than the typical residence time of air in the air basin (~ 10 hours).

Within a specific air basin, the intake fraction for emissions of a primary pollutant from any broadly distributed ground-level outdoor urban source should be similar to the intake fraction for CO and benzene from motor vehicles if its characteristic lifetime is significantly greater than 10 daylight hours. Pollutants emitted from a distributed source with a lifetime on the order of 10 hours or less will be associated with a smaller intake fraction because a significant fraction of the emissions will degrade before people inhale them. For emissions with a relatively short lifetime (less than ~ 1 hour), a significant fraction of the total intake will occur during near-source exposures, such as in vehicles. For such pollutants, it would be difficult to deduce the average concentration to which people are exposed from measurements taken at a small number of ambient monitoring stations.

Further work is needed to determine the applicability of the SoCAB results to other locations. Differences in the intake fraction could arise because of differences in meteorology, such as the wind speed, rate of dispersion, and mixing height, or because of differences in demographics, such as size of the urban area and population density. Further, the intake fraction depends on proximity between people and vehicles, which is related in a complex manner to transportation infrastructure and to social patterns that influence time-activity patterns.

Our study suggests that, for benzene and carbon monoxide from motor vehicles, the direct use of ambient concentrations in an urban air basin as surrogates for exposure concentrations results in $\sim 50\%$ error in assessing population intake. Furthermore, consistency between the one-box model and monitoring data suggests that in some circumstances the one-box model may be used to estimate intake for motor vehicle emissions in an urban area. Additional studies of other sources and other urban air basins are necessary to confirm these inferences.

Comparing the parameters for which we were able to quantify uncertainty, most (~ 70%) of the uncertainty in our results is attributable to uncertainty in the emissions inventory. Substantial effort has already gone into refining extant emissions inventories. Therefore, we do not expect uncertainty in these inventories, and the resulting uncertainty in the iF as determined by the method used in this paper, to improve significantly in the near future.

The uncertainty estimates presented in this paper do not include potential methodological errors. For example, we use census population density data to weight ambient concentration measurements. These data account for where people live, but not where they travel during the day (i.e., downtown to shop or work). Data are not currently available from open sources to estimate population densities within an urban air basin as a function of time. As another example, although monitoring stations offer the most comprehensive ambient concentration data available, these data may misrepresent exposures. Our approach would over-estimate exposures if monitoring station locations were, on average, closer to roadways than people are to roadways. In addition, a limited number of monitoring stations might not suffice to accurately assess the population-weighted average ambient concentration, either because there are not enough monitoring stations or because they are not well situated throughout the air basin. Our method employs average values for parameters such as the percent of ambient concentrations attributable to motor vehicle emissions and the percent of TOG emissions that are benzene. If more detailed information on these parameters becomes available in the future we would be able to refine our calculation.

Any exposure metric will have strengths and weaknesses, depending on the situation for which it is being used. As used here, the intake fraction incorporates, but does not convey, information about inter-individual variability. It is most applicable to evaluating health effects for pollutants with a linear dose-response relationship. In situations where the distribution or time dependence of intakes is important, such as in evaluating acute health effects, iF may have lesser utility. Because intake fraction stresses overall population burden, assessments in support of regulations and permit decisions that focus on the maximum risk to an individual will not be likely to use iF . On the other hand, analyses that assess broad environmental policy issues may be greatly facilitated by the use of intake fractions. For example, iF may be useful in exploring matters of environmental justice that relate to the distribution of the air pollution exposure burden among specific subpopulations.

While this article focuses on primary pollutants, iF could also be quantified for secondary pollutants, which are formed in the atmosphere rather than emitted directly (Evans et al., 2002). For example, in considering ozone, an analyst might apply Eq. (1) by tracking ozone concentrations in the numerator and precursor emissions (e.g., NO_x) in the denominator. In considering PM, an analyst might apply Eq. (1) by incorporating primary PM, secondary PM, or both in the numerator, and emissions of primary PM or emissions of precursor emissions (e.g., NO_x can lead to formation of ammonium nitrate PM) in the denominator. Alternatively, one might evaluate an *incremental* intake fraction as the change in iF arising from a small change in the emissions of a precursor species. (Because there are multiple ways to calculate iF for secondary pollutants, analysts should specify their method precisely and readers must be aware of potential methodological differences when comparing studies.)

One of the merits of the intake fraction approach is that results from one investigation may be applicable to other situations involving similar pollutant and source types. This generalizability offers the potential for substantial efficiency gains in understanding exposures. By analogy, emission factor handbooks are frequently used because of the efficiency of determining an emission factor based on the source and pollutant of interest. Similarly, a compendium of intake fractions, based on source and pollutant type, could offer great utility for exposure assessments.

5. Conclusion

Intake fraction (iF) summarizes the emission-to-intake relationship in a concise and easy to understand manner: iF is the fraction of the emissions of a pollutant taken in by people. For motor vehicle emissions of primary, nonreactive pollutants in the South Coast Air Basin of southern California, we calculate an annual average iF of 47 per million. The results for CO and benzene are similar and consistent with previous intake fraction studies. The monthly-average intake fraction is approximately two times higher in winter than in summer.

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Figure Captions

Fig. 1. Map of California's air basins.

Fig. 2. Motor vehicle emissions of carbon monoxide and benzene in California's South Coast Air Basin during 1996 – 1999.

Fig. 3. Cumulative hourly breathing rate for people in South Coast Air Basin by time of day and activity intensity (based on Layton, 1993).

Fig. 4. Ambient concentration attributable to motor vehicles in the SoCAB during 1996 – 1999. Attributable ambient concentrations show a “U-shaped” profile owing to the predominant meteorology. Summer meteorological conditions tend to disperse primary pollutants more efficiently than winter conditions.

Fig. 5. Normalized diurnal CO concentration profile in the SoCAB during 1996 - 1999. Normalized concentration is the concentration in each hour divided by the average concentration for that day. Concentrations are highest during the morning commute, when emissions are high and dispersion is weak.

Fig. 6. Exposure concentration attributable to motor vehicles in the SoCAB during 1996 - 1999. Attributable exposure concentration shows the same seasonal pattern as attributable ambient concentrations in Fig. 4.

Fig. 7. Intake fraction for motor vehicles in the South Coast Air Basin during 1996 – 1999. Seasonal variability in intake fraction follows from seasonal variability in ambient concentrations (Fig. 4). Note the consistency between the intake fractions for benzene and CO.

Table 1. Summary of ambient pollutant monitoring data

	Carbon monoxide	Benzene
Number of data points	623,534	518
Percent non-detects	5%	6%
Precision	0.1 ppm	0.1 ppb
Detection limit	0.1 ppm	0.2 - 0.5 ppb
Average value	1.20 ppm	1.29 ppb

Table 2. Values for γ_{μ} , the ratio of microenvironmental concentrations to ambient concentrations, used in Eq. (2).

Microenvironment	Carbon monoxide	Benzene
In-vehicle and near-vehicle	4.0	4.0
Residences with an attached garage	1.0	1.2
Indoor location near freeways	2.0	2.0
All other locations	1.0	1.0

Table 3. Simplified intake fraction analysis

	Carbon monoxide	Benzene
Mole fraction (ppm)	1.20	0.00129
Concentration ($\mu\text{g m}^{-3}$)	1410	4.22
Fraction of ambient concentrations attributable to motor vehicles	80%	70%
Breathing rate ($\text{m}^3 \text{d}^{-1}$)	12.2	12.2
Population	1.5×10^7	1.5×10^7
Intake attributable to motor vehicles (g month^{-1})	6.3×10^6	1.6×10^4
Emissions from motor vehicles (g month^{-1})	2.0×10^{11}	5.0×10^8
Estimated intake fraction (per million)	32	33

Fig. 1



Fig. 2

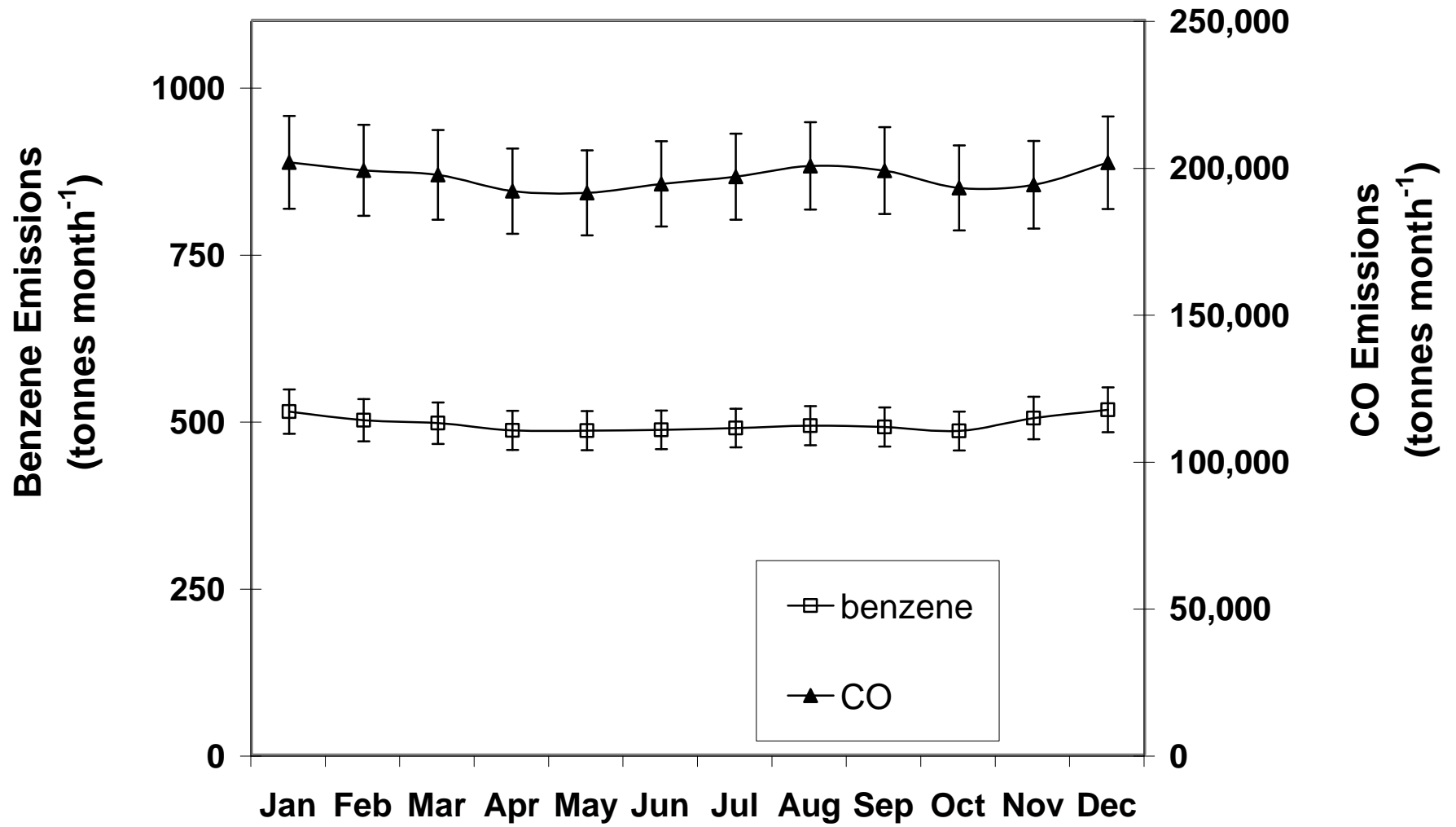


Fig. 3

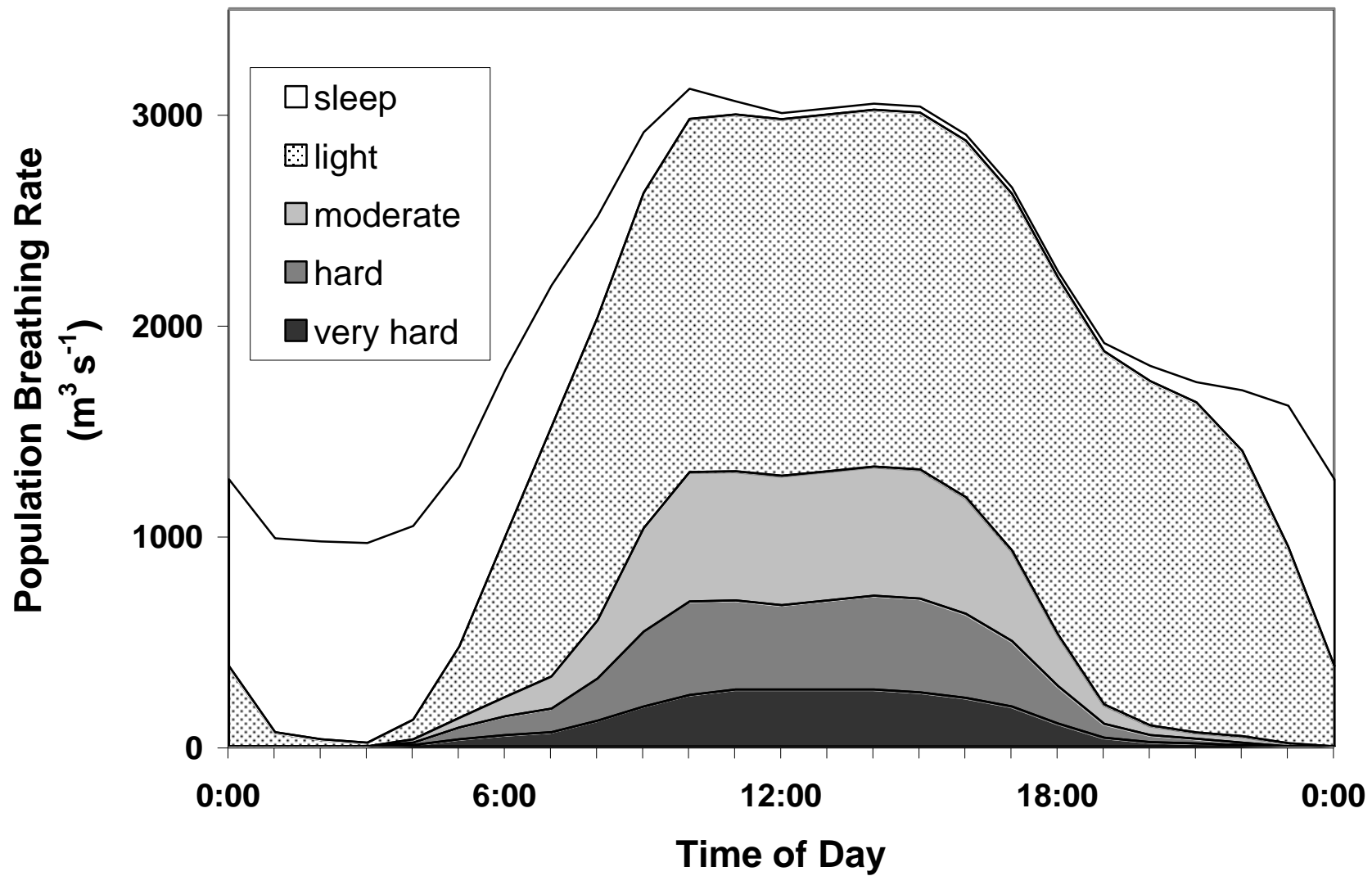


Fig. 4

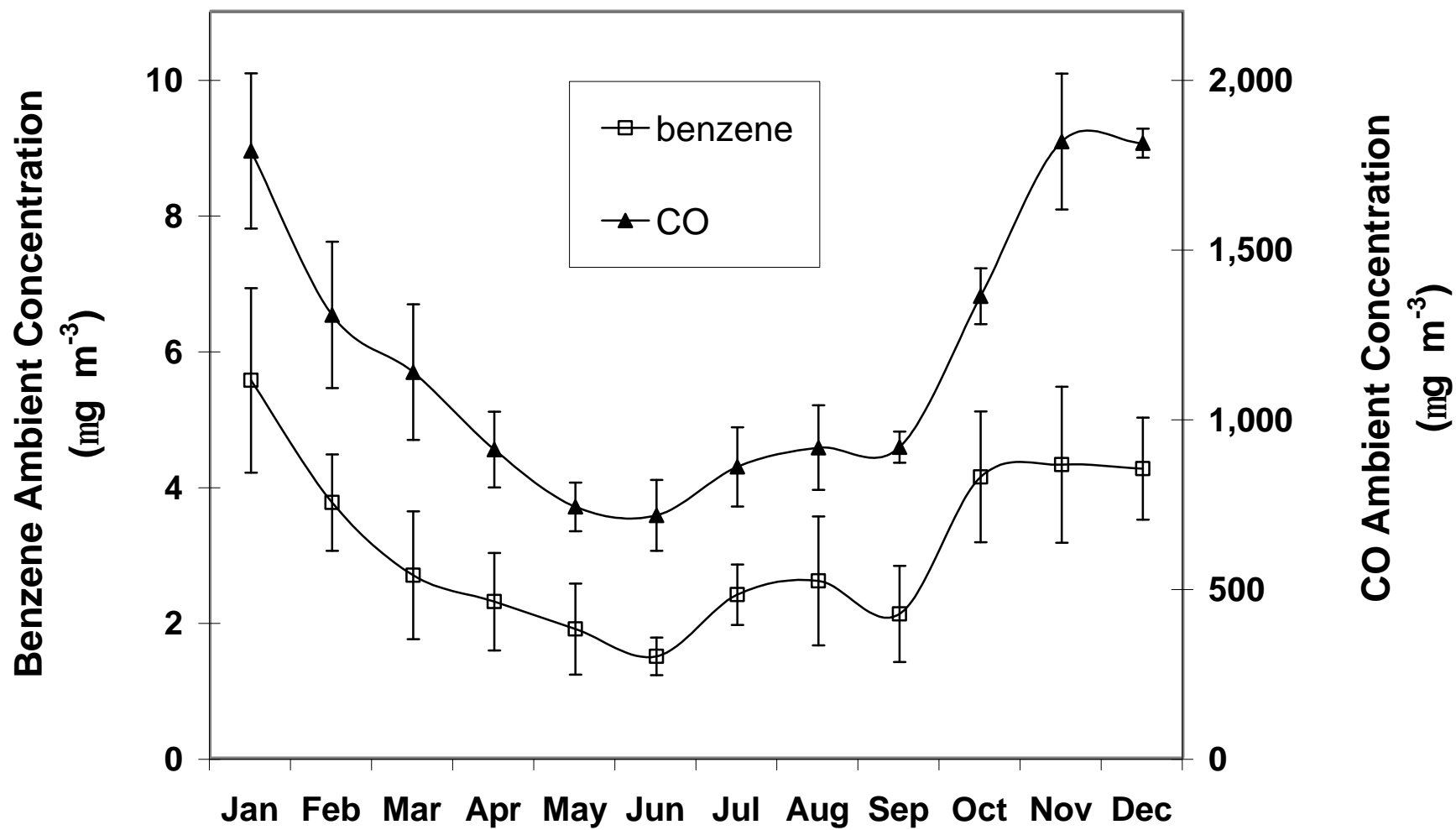


Fig. 5

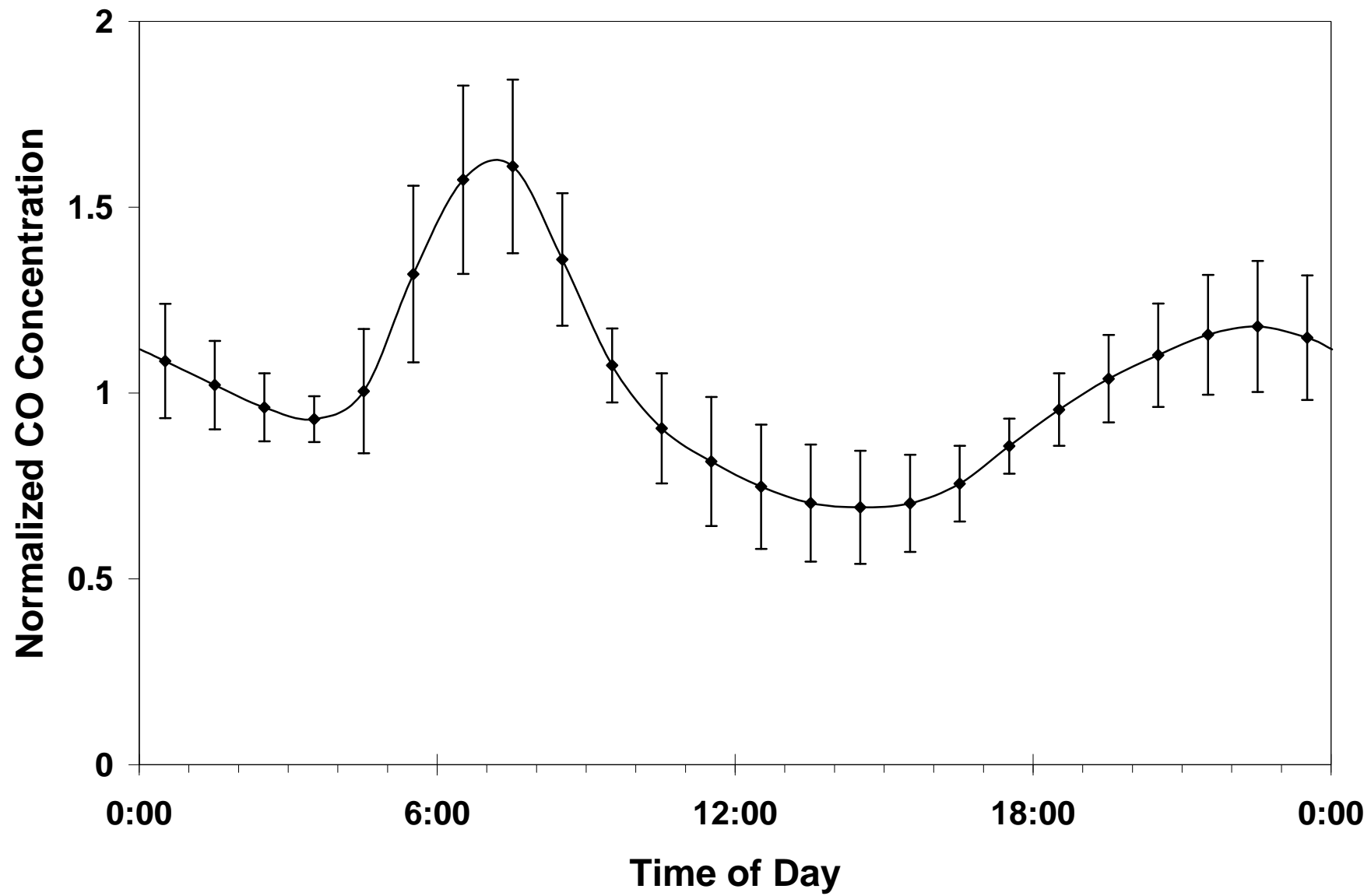


Fig. 6

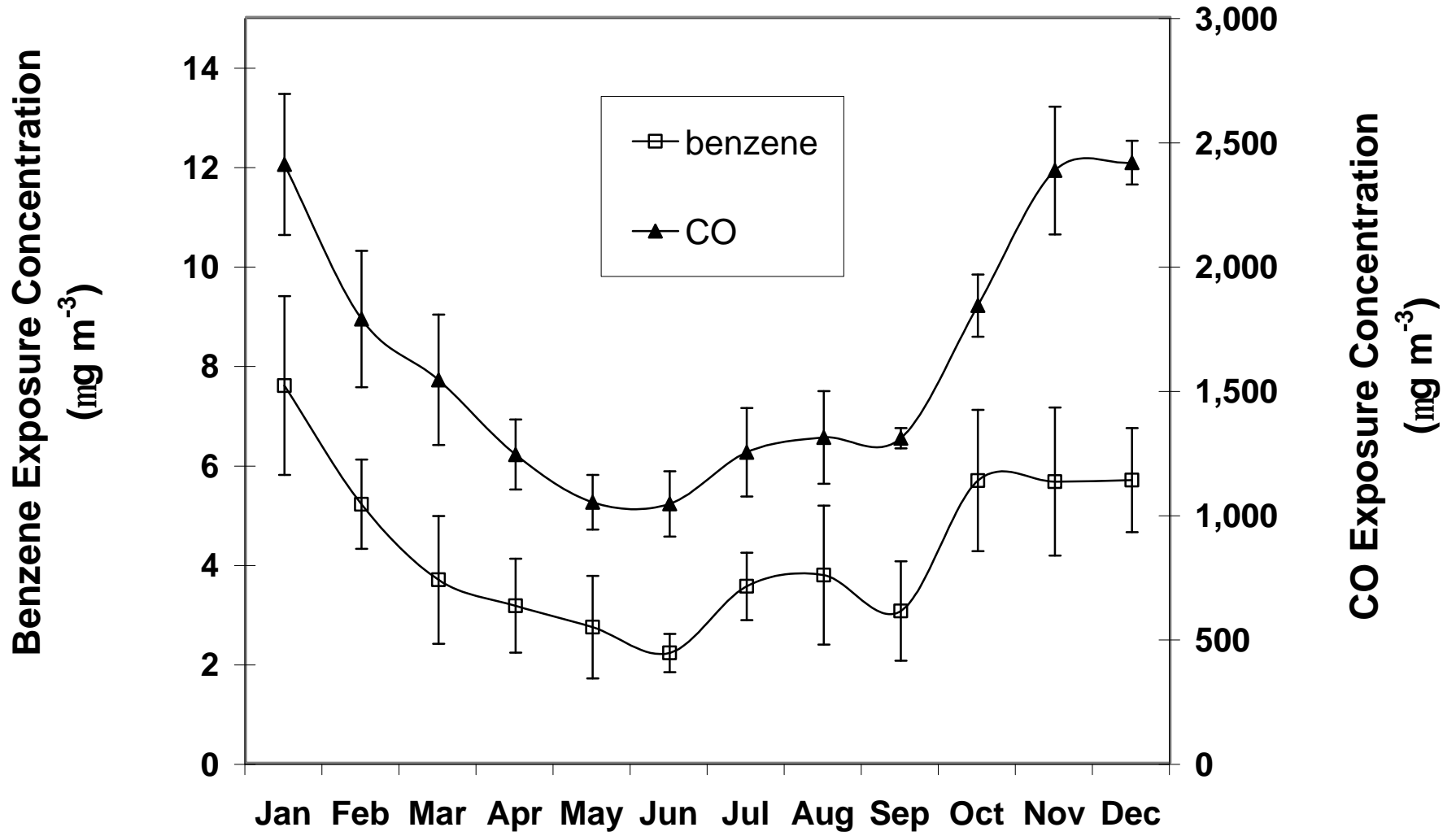


Fig. 7

