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# Spatial patterns of mobile source particulate matter emissions-to-exposure relationships across the United States

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## Abstract

Assessing the public health benefits from air pollution control measures is assisted by understanding the relationship between mobile source emissions and subsequent fine particulate matter ( $PM_{2,5}$ ) exposure. Since this relationship varies by location, we characterized its magnitude and geographic distribution using the intake fraction (iF) concept. We considered emissions of primary  $PM_{2.5}$  as well as particle precursors  $SO_2$  and  $NO_x$  from each of 3080 counties in the US. We modeled the relationship between these emissions and total US population exposure to  $PM_{25}$ , making use of a source-receptor matrix developed for health risk assessment. For primary PM<sub>2.5</sub>, we found a median iF of 1.2 per million, with a range of 0.12-25. Half of the total exposure was reached by a median distance of  $150 \,\mathrm{km}$  from the county where mobile source emissions originated, though this spatial extent varied across counties from within the county borders to 1800 km away. For secondary ammonium sulfate from SO<sub>2</sub> emissions, the median iF was 0.41 per million (range: 0.050–10), versus 0.068 per million for secondary ammonium nitrate from NO<sub>x</sub> emissions (range: 0.00092-1.3). The median distance to half of the total exposure was greater for secondary PM2.5 (450 km for sulfate, 390 km for nitrate). Regression analyses using exhaustive population predictors explained much of the variation in primary PM<sub>2.5</sub> iF ( $R^2 = 0.83$ ) as well as secondary sulfate and nitrate iF ( $R^2 = 0.74$  and 0.60), with greater near-source contribution for primary than for secondary PM<sub>2.5</sub>. We conclude that long-range dispersion models with coarse geographic resolution are appropriate for risk assessments of secondary  $PM_{2.5}$  or primary  $PM_{2.5}$  emitted from mobile sources in rural areas, but that more resolved dispersion models are warranted for primary  $PM_{2.5}$  in urban areas due to the substantial contribution of near-source populations. © 2006 Elsevier Ltd. All rights reserved.

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# 1. Introduction

Inhaling fine particulate matter  $(PM_{2.5})$  can lead to several adverse health impacts ranging from

reduced lung function to premature mortality (Gauderman et al., 2004; Pope, 2000; Brunekreef and Holgate, 2002; US Environmental Protection Agency, 2004). In recent cost–benefit analyses of the Clean Air Act, the disease burden attributable to  $PM_{2.5}$  exposure dominated health benefits resulting from pollution control in the US (US Environmental Protection Agency, 1999b). Mobile sources such as cars, trucks, trains, ships, and airplanes

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contribute to  $PM_{2.5}$  pollution by direct emissions (primary  $PM_{2.5}$ ) as well as emissions of precursor pollutants like sulfur dioxide, nitrogen oxides, and hydrocarbons which undergo chemical transformations to form secondary  $PM_{2.5}$ . In the US, approximately 30% of primary  $PM_{2.5}$  emissions and 60% of NO<sub>x</sub> emissions can be attributed to mobile sources (US Environmental Protection Agency, 1999c).

Decision-making agencies often follow a risk assessment framework to compare costs and benefits of air pollution control strategies. In this context, risk assessment typically involves identification of the hazardous pollutants, characterization of emissions that might influence exposure to these substances, application of atmospheric dispersion models to determine the concentration impacts of these emissions, estimation of health risks associated with concentration changes, and risk characterization. Thus, it is important to understand the relationship between emissions and exposures across the population in a format that is relevant for risk calculations. This relationship depends on where people live, where emission sources are located, which pollutants are emitted, meteorology, atmospheric chemistry, and myriad other conditions. Atmospheric dispersion models in this context must be applied over a spatial domain large enough to capture most population exposure, but with sufficient resolution to capture spatial gradients and key atmospheric phenomena. A measure that summarizes the emissions-to-exposure relationship can provide significant insight about the importance of the above factors, the necessary model scope and resolution, and the settings in which emission controls would yield greater or lesser health benefits.

The emissions-to-exposure relationship can be characterized by the intake fraction concept, abbreviated iF, simply defined as the fraction of a pollutant (or its precursor) emitted from a source that is inhaled by a specified population during a given time (Bennett et al., 2002). The concept has been in the scientific literature for decades, although with an array of different names, including exposure efficiency (Harrison et al., 1986; Evans et al., 2000, 2002), exposure factor (Smith, 1988), exposure effectiveness (Smith, 1993), inhalation transfer factor (Lai et al., 2000), exposure constant (Guinee and Heijungs, 1993), potential intake (Hertwich et al., 2001), and fate factor (Jolliet and Crettaz, 1997). The impetus behind iF is to find straightforward ways to organize scientific information in a manner that informs risk-based environmental policy, and to allow findings from exposure studies to be extrapolated to other settings.

Other studies have investigated intake fractions from mobile sources, but none have provided the necessary information to understand spatial heterogeneity or to determine the appropriate scope and resolution for a dispersion model in a risk assessment context. A Southern California Air Basin (SoCAB) study combined ambient monitoring data with time-activity patterns to develop local iFs for carbon monoxide and benzene emitted from mobile sources (Marshall et al., 2003). While this study provides useful information for the SoCAB, the appropriate iF values in other settings may differ, and the use of monitoring data in one air basin rather than dispersion modeling makes it difficult to determine whether significant exposures occurred outside of the basin. Building on this work, researchers used three methods to estimate iFs for nonreactive vehicle emissions in US urban areas, including a one-compartment steady-state massbalance model and applied US EPA's Nationalscale Air Toxics Assessment (NATA) for diesel particulate matter (Marshall et al., 2005). Though the three methods provided consistent results, the use of a box model does not capture within or between region iF heterogeneity or more complex meteorology, and the other approaches do not address potential impacts outside of the source region.

Two studies in the literature did evaluate spatial patterns in mobile source iFs. The first, (Nigge, 2001), estimated primary  $PM_{2.5}$  iFs in Germany by combining a Gaussian plume model (GPM) and population densities close to the source and a wind trajectory model (WTM) at greater distances. Better near-source model resolution was found to improve iF estimates for densely populated areas and low emission heights, as would occur in traffic congested urban areas. However, this study was limited by the assumption that the contribution to iF from distances greater than 100 km from the source was constant regardless of population density patterns, which may not be appropriate. The second study used CALPUFF, a dispersion model based on Gaussian dispersion theory that models continuous emissions as a series of discrete puffs, to estimate primary and, for the first time, secondary  $PM_{2.5}$  iFs for 40 highway stretches (Evans et al., 2002). This study provided some insight into the appropriate dispersion model scale and heterogeneity of mobile source iFs, however the receptor resolution  $(100 \text{ km} \times 100 \text{ km})$  and limited number of geographic areas did not allow for nationally generalizable results.

In our analysis, we build on the existing literature by estimating both primary and secondary PM<sub>2.5</sub> mobile source iFs for all counties in the contiguous US. These iFs reflect the national, rather than local, public health impacts attributable to mobile source emissions from each county, since emissions from one county can influence ambient concentrations in downwind counties. We applied a source-receptor model developed for air pollution risk assessment to examine the national average and distribution of iF at county-level resolution, which can inform the development and application of more detailed atmospheric dispersion models. Furthermore, we develop regression models to help explain the significant influences on mobile source iFs. These results should provide guidance for future health impact assessment studies with recommendations about the model scope and resolution appropriate for different pollutants in different settings.

#### 2. Methods

We use a dispersion model which covers the 48 contiguous US states and treats all mobile source emissions in each of 3080 US counties (the largest administrative division of most states) as area sources. It is important to note that mobile source emissions impact the county where the emissions originated (termed the source county, j) in addition to downwind counties, and that the national iF incorporates exposure in all counties.

#### 2.1. The intake fraction

A mobile source intake fraction is calculated for each of 3080 counties for primary and secondary  $PM_{2.5}$ . The national intake fraction corresponding to the county where the emissions originated, iF<sub>j</sub>, is the total population  $PM_{2.5}$  exposure divided by the emissions (precursor emissions in the case of secondary  $PM_{2.5}$ ) from the source county. It is calculated according to

$$iF_j = \Sigma_i (P_i \Delta C_{ij}) \cdot BR/Q_i, \tag{1}$$

where  $P_i$  is the population in impacted county *i*,  $\Delta C_{ii}$ (in  $\mu g m^{-3}$ ) is the change in ambient PM<sub>2.5</sub> concentration in impacted county *i*. This change is due to mobile source emissions of PM2.5 or particle precursors,  $Q_j$  (in  $\mu g d^{-1}$ ), originating from source county *j*, and **BR** is the nominal population breathing rate of  $20 \text{ m}^3 \text{ d}^{-1}$ . Eq. (1) is evaluated for all 3080 counties, j, and i ranges from 1 to 3080 for each *j*. County-level population projections for year 2007, estimated from 1990 Census data, were used (Abt Associates et al., 2000), although we tested the sensitivity of our findings to 2000 Census data (US Census Bureau, 2000). The values of  $\Delta C_{ii}$ were estimated by use of a source-receptor (S-R) matrix, described in Section 2.2. Mobile source PM<sub>2.5</sub> and precursor emissions from each county were based upon EPA National Emissions Inventory information (Abt Associates, 2004; US Environmental Protection Agency, 1999a).

Since they are pollutant specific, four mobile source intake fractions were estimated. For primary PM<sub>2.5</sub>, the iF represents the fraction of PM<sub>2.5</sub> emitted that is inhaled by the population and will be denoted as iF(p). Secondary PM<sub>2.5</sub> iFs represent the ratio of the mass of secondarily generated ammonium sulfate or nitrate inhaled by the population to the mass of the emitted SO<sub>2</sub> or NO<sub>x</sub>. Refer to Table 1 for a description of the notation for iF(p) and the three secondary intake fractions, iF(as, SO<sub>2</sub>), iF(an,NO<sub>x</sub>), and iF(an,SO<sub>2</sub>). This last iF is negative indicating that reductions in SO<sub>2</sub> emissions can result in increased NH<sub>4</sub>NO<sub>3</sub> exposures in some settings (Levy et al., 2003; West et al., 1999).

Table	1	
Intake	fraction	notation

	PM <sub>2.5</sub> type	Exposure pollutant	Emitted pollutant	
iF(p)	Primary	PM <sub>2.5</sub>	PM <sub>2.5</sub>	
iF(as,SO <sub>2</sub> )	Secondary	Ammonium sulfate $((NH_4)_2SO_4)$	Sulfur dioxide (SO <sub>2</sub> )	
iF(an,SO <sub>2</sub> )	Secondary	Ammonium nitrate $(NH_4NO_3)$	Sulfur dioxide $(SO_2)$	
$iF(an, NO_x)$	Secondary	Ammonium nitrate (NH <sub>4</sub> NO <sub>3</sub> )	Oxides of nitrogen $(NO_x)$	

# 2.2. The S-R matrix

The S-R matrix is a regression-based derivation of output from the Climatological Regional Dispersion Model (CRDM) which uses assumptions similar to the Industrial Source Complex Short Term model (ISCST3). It was developed by Pechan and Associates for Abt Associates and used in past regulatory impact analyses (US Environmental Protection Agency, 1999d). S-R matrix provides a database of transfer factors that summarize the impact that mobile source PM2,5 and precursor emissions from any one county have on ambient PM<sub>2.5</sub> concentrations in that county as well as all other counties (Abt Associates, 2003). The underlying model, CRDM, incorporates terms for wet and dry deposition of primary and secondary species that constitute PM<sub>2.5</sub> and uses meteorological summaries (annual average mixing heights and joint frequency distributions of wind speed and direction) from 100 upper air meteorological sites throughout North America. Additionally, CRDM uses Turner's sector-average approach, a probabilistic method where relative frequencies of occurrence of combinations of wind and stability conditions at the emissions source are used to calculate the relative frequencies of transport in various sectors (Abt Associates, 2004).

The mass flux of directly emitted PM<sub>2.5</sub> is a function of the material initially emitted and the amount deposited by wet and dry processes during the period of transport from the emission point to the receptor. The mass flux of secondary pollutants, (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> and NH<sub>4</sub>NO<sub>3</sub>, is dependent upon the fraction of the emitted species,  $SO_2$  and  $NO_x$ , that is chemically converted in the atmosphere to the secondary species and the amount of the secondary species that is deposited by wet and dry deposition processes. S-R matrix includes ambient concentrations of ammonia, sulfate, and nitrate by county. The incremental PM<sub>2.5</sub> concentration changes, given  $SO_2$  and  $NO_x$  emission changes, are dictated by some simplifying assumptions. Generally, all sulfate present is assumed to be converted to ammonium sulfate while ammonium nitrate formation is limited by the relative concentrations of nitrate and ammonium remaining after the sulfate neutralization process. Particulate ammonium nitrate is assumed to form only a quarter of the year, given the temperature dependence of the conversion from nitric acid to particulate ammonium nitrate (Abt Associates, 2004).

A set of county-specific calibration factors for PM<sub>2.5</sub> was used to calibrate the S-R matrix model to ambient air quality data. The calibration factors are estimated using the 2001 National Emissions Inventory (NEI) and data from the Federal Reference Method (FRM) and EPA's Speciation Network (ESPN) monitor sites for 2002. Prior to calibration, PM<sub>2.5</sub> concentrations at county centroids were estimated using the S-R matrix as applied to a comprehensive emissions inventory. Then, monitored data from FRM and ESPN sites were spatially interpolated to county centroids using inverse distance weighting to estimate the same baseline PM<sub>2.5</sub> levels. The calibration factors are based on the ratio of the monitor to modeled PM2.5 estimates and range from 0.11 to 3.5, with a median value of 0.90. All iFs presented in this paper reflect calibrated estimates, although we performed a sensitivity analysis of the impact of the calibration factors on iF.

# 2.3. Analysis

One of the primary aims of this study is to understand the spatial extent of mobile source intake fractions. That is, we wish to characterize at what distance from the source county the bulk of the national intake fraction is captured, as this will inform conclusions about appropriate dispersion model scope.  $iF_i$  by definition is a national-scale sum of exposure across receptor counties (Eq. (1)), but we can consider the fraction of  $iF_i$  occurring within various radial distances of the source county. The minimum fraction will occur within the source county itself, as this excludes populations at all other distances. We define  $fTE_i$  to be the fraction of total exposure occurring within the source county borders, as presented in Eq. (2). For simplicity's sake, we consider this to represent a radial distance of 0 km.

$$fTE_j = \Delta C_{jj} P_j / \Sigma_i (P_i \Delta C_{ij}).$$
<sup>(2)</sup>

In counties where  $fTE_j$  is high, eliminating mobile source emissions in the county would primarily influence public health at a local level. (Counties might have high  $fTE_j$  if they are highly urbanized, or if they are surrounded by areas that contain few or no people.) On the other hand, for counties where  $fTE_j$  is low, the benefit of eliminating mobile source emissions in the county would primarily occur in other counties. If source county concentration estimates are off by a factor of 2, this would lead to a greater impact in counties with high  $fTE_j$ . It would be increasingly important to do more resolved modeling in these areas to adequately capture spatial concentration gradients that might occur within counties. Additionally, we compute the distances for which the fraction of total exposure is 10%, 50%, or 90%, thereby characterizing the spatial extent of the iFs.

To explain heterogeneity in  $iF_i$ , we develop simple and multiple linear regression models using population predictors. Simple linear regression models examined the predictive power of the source-county population,  $P_i$ , and included an intercept term to account for the intake fraction that would occur outside of the source county. After a preliminary analysis of the spatial extent of  $iF_i$ , five population predictors were tested in multiple linear regression models (MLR). They were the population within 50 km ( $P_{i,<50}$ ), between 50 and 100 km ( $P_{i,50-100}$ ), between 100 and 200 km ( $P_{j,100-200}$ ), between 200 and 500 km ( $P_{j,200-500}$ ), and outside of 500 km of the source county  $(P_{i,>500})$ . Since the MLR models incorporated exhaustive US population predictors, the intercept was constrained to zero, resulting in an iF of zero if there were no people in any of the population bins.

In addition to population, windspeed, temperature, precipitation, mixing height and other factors can influence the fate and transport of contaminants. While these factors are clearly significant, they are difficult to include in an interpretable way in iF regression models, since meteorology at the source county may not be representative of downwind meteorology. Given that climate and county size are distributed differently in Eastern versus Western states, a simple dummy variable was tested to determine if it added to the explanatory power of the exhaustive population predictor model. For the purposes of this analysis, the following 11 states were considered Western states: Arizona, California, Colorado, Idaho, Montana, Nevada, New Mexico, Oregon, Utah, Washington, and Wyoming.

#### 3. Results

#### 3.1. Descriptive statistics

Primary PM<sub>2.5</sub> mobile source intake fractions for 3080 US counties varied from 0.12 to 25 per million, with a median of 1.2 and a mean of 1.6 per million (Fig. 1). Alternatively stated, on average 1.6 g of



Fig. 1. Central tendencies and distributions of primary and secondary  $PM_{2.5}$  mobile source intake fractions for US counties. For each iF distribution, the box represents the middle half of the data, called the interquartile range (IQR). On each box, the solid vertical lines represent the 25th, 50th (median), and 75th percentiles, while the dotted line represents the mean. The "whiskers" extending from the boxes include iFs within 150% of the IQR from the lower and upper quartiles and the dots outside the whiskers represent the 5th and 95th percentiles of the distribution. Note that the intake fractions are displayed on a log-scale and that the iF(an,SO<sub>2</sub>) values are negative.

 $PM_{2.5}$  is inhaled by the population for every metric ton emitted by mobile sources generated in a county. The emissions-weighted iF(*p*), which reflects

the average exposure per ton of  $PM_{2.5}$  emitted across the US, is 2.5 per million. The increased magnitude reflects the correlation between mobile



Fig. 2. Maps of national-scale mobile source intake fractions for US counties. The iFs are plotted in units of per million. Shading occurs by 20-percentile iF bin and the circles indicate the top 1-percentile counties: (a) primary  $PM_{2.5}$  iF(*p*); (b) secondary  $PM_{2.5}$  iF(as,SO<sub>2</sub>); (c) secondary  $PM_{2.5}$  iF(an,NO<sub>x</sub>); (d) secondary  $PM_{2.5}$  iF(an,SO<sub>2</sub>).



Fig. 2. (Continued)

source emissions and population density. The highest iF(p) values tend to occur in densely populated counties whose emissions impact densely populated downwind regions (Fig. 2a). The fraction of total exposure that occurs within the source county borders,  $fTE_j$ , ranges considerably from 0.1% to 92%, with median and mean estimates of 11% and 16%, respectively.

The central tendencies of the secondary PM<sub>2.5</sub> iFs are 1–2 orders of magnitude smaller than iF(*p*) (Fig. 1). The median value for iF(as,SO<sub>2</sub>) is 0.41 per million (range: 0.050–10), which is approximately a factor of 6 greater than the median for iF(an,NO<sub>x</sub>) of 0.068 per million (range: 0.00092–1.3). The median iF(an,SO<sub>2</sub>) value is -0.033 per million, with the negative sign indicating that reductions in SO<sub>2</sub> emissions can increase ammonium nitrate exposure. The emissions-weighted iF(as,SO<sub>2</sub>) is 0.66, iF(an, NO<sub>x</sub>) is 0.12, and iF(an,SO<sub>2</sub>) is -0.088 per million. Secondary iFs exhibit as much or more variation in magnitude, but less small-scale geographic variation than the primary iFs (Figs. 2b–d).

By comparing iF(an,SO<sub>2</sub>) to iF(as,SO<sub>2</sub>), we can get a sense of the amount of sulfate exposure reduction offset by nitrate formation. In this way we determined the public health benefits of SO<sub>2</sub> emission controls might be reduced an average of 9% (range: 1–29%) for US counties when the increased nitrate concentrations are incorporated.

#### 3.2. Spatial extent

To determine the spatial extent of the intake fraction, we examined how the cumulative fraction of total exposure increased with distance from the source county from  $fTE_j$  to 100%. All 3080 distributions for iF(p),  $iF(as,SO_2)$ , and  $iF(an,NO_x)$ 

are summarized in Fig. 3. These box plots depict the distance from the source county where 10%, 50%, and 90% of the total exposure for each pollutant is reached. For iF(*p*), half of the total primary PM<sub>2.5</sub> exposure is reached at a median distance of 150 km from the source county, though for 5% of counties, it is met within the county borders while for another 5% of counties it is not met until more than 1000 km from the county where the emissions originated. The median distances where half of the total secondary PM<sub>2.5</sub> exposure is met for iF(as, SO<sub>2</sub>), iF(an,NO<sub>x</sub>), and iF(an,SO<sub>2</sub>) are 450, 390, and 740 km, respectively, signifying that the spatial extent of the secondary iFs is greater than for the primary iFs.

#### 3.3. Regression modeling

Given the significant contribution of sourcecounty populations to iF in many settings, we initially investigated the extent to which the sourcecounty population,  $P_j$ , could explain the variation in iFs (Table 2). For the iF(p) model, the intercept,  $\beta_0$ , of 1.27 per million can be interpreted as the average magnitude of the intake fraction occurring outside of the source county, while the slope times the source-county population,  $\beta_1 P_j$ , represents the magnitude of the national iF<sub>j</sub> occurring inside the source county. In simple linear regression models,  $P_j$ 



Fig. 3. Spatial extent of mobile source intake fractions. The distances where 10%, 50%, and 90% of the total exposure is reached are shown. The boxes indicate the middle half of each distribution, while the dots outside the whiskers represent the 5th and 95th percentile values.

Table 2 Mobile source intake fraction simple linear regression models for  $iF_j = \beta_0 + \beta_1 P_j$ 

Dependent variable	$R^2$		Independent variables	Parameter estimate	Std. error	<i>t</i> -value
$iF_j(p)$	0.43	$\frac{\beta_0}{\beta_1}$	(Intercept) Source-county population	$1.27 \times 10^{-6}$ $3.42 \times 10^{-12}$	$2.31 \times 10^{-8}$ $7.10 \times 10^{-14}$	55** 48**
iF <sub>j</sub> (as,SO <sub>2</sub> )	0.49	$\beta_0$ $\beta_1$	(Intercept) Source-county population	$3.64 \times 10^{-7}$ $7.34 \times 10^{-13}$	$4.75 \times 10^{-9}$ $1.46 \times 10^{-14}$	76** 51**
$iF_j(an, NO_x)$	0.07	$\beta_0$ $\beta_1$	(Intercept) Source-county population	$7.73 \times 10^{-8}$ $5.29 \times 10^{-14}$	$1.33 \times 10^{-9}$ $4.07 \times 10^{-15}$	58** 13**
iF <sub>j</sub> (an,SO <sub>2</sub> )	0.46	$\beta_0 \\ \beta_1$	(Intercept) Source-county population	$-3.17 \times 10^{-8} \\ -1.56 \times 10^{-13}$	$9.89 \times 10^{-10}$ $3.03 \times 10^{-15}$	$-32^{**}$ $-52^{**}$

\*\*p<0.0001.

Table 3 Exhaustive population predictor multiple linear regression models for  $iF_j = \beta_1 P_{j,<50} + \beta_2 P_{j,50-100} + \beta_3 P_{j,100-200} + \beta_4 P_{j,200-500} + \beta_5 P_{j,>500}$ 

Dependent variable	Adjusted $R^2$		Independent variables <sup>a</sup>	Parameter estimate	Std. error	<i>t</i> -value
$iF_j(p)$	0.83	$\beta_1$	$P_{i < 50}$	$1.07 \times 10^{-12}$	$1.94 \times 10^{-14}$	55**
		$\beta_2$	$P_{i,50-100}$	$1.60 \times 10^{-13}$	$1.42 \times 10^{-14}$	11**
		$\beta_3$	$P_{i100-200}$	$2.73 \times 10^{-14}$	$6.12 \times 10^{-15}$	4.4**
		$\beta_4$	$P_{i200-500}$	$1.16 \times 10^{-14}$	$1.32 \times 10^{-15}$	8.8**
		β5	$P_{i > 500}$	$2.12 \times 10^{-15}$	$1.12 \times 10^{-16}$	19**
iF <sub>j</sub> (as,SO <sub>2</sub> )	0.74	$\beta_1$	$P_{i < 50}$	$1.31 \times 10^{-13}$	$5.76 \times 10^{-15}$	23**
		$\beta_2$	$P_{i,50-100}$	$3.11 \times 10^{-14}$	$4.12 \times 10^{-15}$	7.4**
		$\beta_3$	$P_{i100-200}$	$6.92 \times 10^{-15}$	$1.81 \times 10^{-15}$	3.8**
		$\beta_4$	$P_{i200-500}$	$4.04 \times 10^{-15}$	$3.91 \times 10^{-16}$	10.3**
		$\beta_5$	$P_{i > 500}$	$8.35 \times 10^{-16}$	$3.32 \times 10^{-17}$	25.1**
$iF_j(an, NO_x)$	0.60	$\beta_1$	$P_{i \le 50}$	$1.56 \times 10^{-14}$	$1.43 \times 10^{-15}$	11**
		$\beta_2$	$P_{i,50-100}$	$4.89 \times 10^{-15}$	$1.04 \times 10^{-15}$	4.7**
		$\beta_3$	$P_{i100-200}$	$6.44 \times 10^{-16}$	$4.50 \times 10^{-16}$	1.4
		$\beta_4$	$P_{i200-500}$	$-1.69 \times 10^{-16}$	$9.70 \times 10^{-17}$	$-1.7^{*}$
		$\beta_5$	$P_{i > 500}$	$2.75 \times 10^{-16}$	$8.25 \times 10^{-18}$	33**
iF <sub>j</sub> (an,SO <sub>2</sub> )	0.44	$\beta_1$	$P_{i \leq 50}$	$-2.15 \times 10^{-14}$	$1.31 \times 10^{-15}$	$-16^{**}$
		$\beta_2$	$P_{i,50-100}$	$-3.84 \times 10^{-15}$	$9.55 \times 10^{-16}$	$-4.0^{**}$
		$\beta_3$	$P_{i100-200}$	$-1.45 \times 10^{-15}$	$4.12 \times 10^{-16}$	$-3.5^{*}$
		$\beta_4$	$P_{i,200-500}$	$-6.84 \times 10^{-16}$	$8.91 \times 10^{-17}$	-7.7**
		$\beta_5$	$P_{j,>500}$	$-4.09 \times 10^{-17}$	$7.57\times10^{-18}$	$-5.4^{**}$

<sup>a</sup>All population terms are calculated by summing country populations that fall within the specified distance in km between country centroids.

\*p < 0.005.

\*\**p*<0.0001.

explained over 43% of the variability in iF(p),  $iF(as,SO_2)$ , and  $iF(an,SO_2)$ , but only 7% for  $iF(an,NO_x)$ .

We next considered exhaustive population predictor ( $P_{j,<50}$ ,  $P_{j,50-100}$ ,  $P_{j,100-200}$ ,  $P_{j,200-500}$ , and  $P_{j,>500}$ ) multiple linear regression models for iF(p), iF(as,SO<sub>2</sub>), iF(an,NO<sub>x</sub>), and iF(an,SO<sub>2</sub>) (Table 3). Except for the  $P_{j,100-200}$  term for iF(an,NO<sub>x</sub>), all population predictor estimates are significant at the 0.05 level. The iF(p) model tells us that for each additional person located within 50 km of the source county, iF(p) would increase by 1.07 per trillion if all other population terms were to remain unchanged. The partial slope terms decrease in magnitude with distance from the county where mobile source emissions originate, so, for iF(p), a person located within 50 km of the source county would experience about 7 times the exposure of a person located between 50 and 100 km away. The population based MLR models explained between 44% and 83% of the variability in all iFs as indicated by the  $R^2$  values. A dummy indicator was added to the MLR model to help account for the differences in climate, county size, and population density that might be expected in eastern versus western US counties. Interaction terms including the West dummy indicator for iF(*p*) were significant for  $P_{<50}$  and  $P_{50-100}$  and the adjusted  $R^2$  for this model was 0.89. Adding this indicator to the secondary iF MLR models increased the adjusted  $R^2$  by 0.02, 0.11, and 0.22 for iF(an,NO<sub>x</sub>), iF(as,SO<sub>2</sub>), and iF(an,SO<sub>2</sub>), respectively, but did not fully capture regional patterns associated with secondary PM<sub>2.5</sub> chemistry.

We undertook limited sensitivity analyses, focusing on the population estimates and calibration factors. We present results that use 2007 countylevel population projections based on 1990 Census data; recalculating all iFs using 2000 Census data did not significantly change the descriptive statistics, spatial extent, or regression models. For example, the median iF(p) was the same to 2 significant figures. Furthermore, the results presented in this study make use of calibration factors in S–R matrix that calibrate model output with ambient monitoring data. Although the median value for iF(p)shifted upwards by 17% when the calibration factors were removed, the core results remained the same.

# 4. Discussion

Our use of S–R matrix allowed us to explore the magnitude, geographic distribution, and spatial extent of primary and secondary  $PM_{2.5}$  mobile source intake fractions across the US There is considerable heterogeneity in these iFs, with values spanning several orders of magnitude across US counties. However, much of this heterogeneity can be explained using population predictors, particularly those close to the source county.

One of the objectives of this paper was to examine the spatial extent of mobile source intake fractions, to inform future modeling efforts. We found that half of the total exposure for primary  $PM_{2.5}$  from mobile sources occurs by a median distance of 150 km from the source county, and at least twice as far away for secondary  $PM_{2.5}$ . These distances are in marked contrast to other studies that found a drop-off in ultrafine particle counts within approximately 100 m of a Los Angeles freeway (Zhu et al., 2002) and associations with traffic related air pollution at schools located within 400 m of motorways in the Netherlands (Janssen et al., 2001). While

these findings appear to contradict our results, our focus is on total population exposure, which would be expected to exhibit a greater spatial extent than individual exposure. In other words, even if concentrations were an order of magnitude lower beyond 400 m, if 100 times more people lived beyond 400 m, then iF would not be dominated by local exposures. The geographic resolution of S-R matrix, at county-level, also limited our ability to fully examine the spatial extent of the iF. Another national-level study using а model with  $100 \text{ km} \times 100 \text{ km}$  resolution found half of the total exposure to be between 100 and 350 km (Evans et al., 2002), but others have not explored this question with more spatially resolved models.

To help interpret our findings, it is useful to compare our results to other mobile source iF studies in the literature. For estimates in the US, we can directly compare our results to the counties modeled in previous studies, and we can approximately compare our results with non-US studies by matching on population densities. The results of these comparisons are summarized in Table 4. Despite using different dispersion models, our national iF(p) are comparable to those reported by Evans et al. (2002), while our iF(as,SO<sub>2</sub>) and  $iF(an, NO_x)$  values are 4–14 times higher. This is consistent with another study's power plant iF comparison to Evans et al. (2002), which also used CALPUFF and found that after controlling for population, the primary PM iF estimates were similar, but the secondary iF estimates remained greater (Zhou et al., 2003). This may indicate underestimation of secondary iFs by Evans et al. (2002). For all iF types, we found greater urbanrural differences than did Evans et al., potentially related to their relatively coarse geographic resolution and small number of modeled locations. Our iF(p) estimates are approximately a factor of 3–6 lower than those by Marshall et al. (2003, 2005), likely due in part to their consideration of timeactivity patterns and smaller-scale geographic resolution, as well as their modeling of non-reactive gases rather than particulate matter. Similar iF(p)results to Marshall, 2003 were reported in a study in Finland that combined source apportionment techniques with personal exposures in microenvironments (Jantunen et al., 2004). After categorizing US county population densities into the same bins as Nigge (2001) and separating the impacted populations into within and outside of 100 km from the source, we calculated iF(p) that were 3-8 times

Table 4 Comparison of mean mobile source intake fractions<sup>a</sup> from selected studies to the present study

Study	Evans et al. (2002)	Marshall et al. (2003)	Marshall et al. (2005)	Nigge (2001)
Scale of iF Number of sites modeled	National 20 rural, 19 urban	Local 4	Local 1105	Local and national NA
Model	CALPUFF	Monitoring and time activity	NATA	GPM for < 100 km; WTM otherwise
iF( <i>p</i> )				
Previous estimates	9.0. 9.4	77 <sup>°</sup>	7.2 <sup>c</sup>	8-19
Present study <sup>b</sup>	1.8, 9.8	13	2.6	1–7
iF(as,SO <sub>2</sub> )				
Previous estimates	0.14, 0.12	NA	NA	NA
Present study <sup>b</sup>	0.52, 1.7	—	—	—
$iF(an, NO_x)$				
Previous estimates	0.024, 0.024	NA	NA	NA
Present study <sup>b</sup>	0.10, 0.23	—	—	—

<sup>a</sup>All iFs are presented in units of per million.

<sup>b</sup>The present comparisons only reflect the counties included in previous studies or equivalent population density areas, and not national average values. Where local iFs were reported, we also compare just the local portion of the national iF.

<sup>c</sup>Marshall et al. reported iFs using breathing rate of  $12.2 \text{ m}^3 \text{ d}^{-1}$ . The above values reflect a BR of  $20 \text{ m}^3 \text{ d}^{-1}$  to be comparable to the present study.

lower than those found in Germany. However, the long-range component of the Nigge iF(p) was assumed to be a constant for all population density ratios, while we found this component of the iF to vary by a factor of 10.

Another study examined power plant iFs using S–R matrix for 507 power plants across the US (Wilson, 2003). This provides a unique comparison point for our mobile source estimates, since the same iF methodology and underlying S–R model were employed. By comparing the iFs for the counties where power plants were located, we can gain insight about the difference in iF for a ground-level source and an elevated stack. As expected, we found the mobile source iFs to be greater than the corresponding power plant iFs, although the magnitude of the difference varied across sites (Fig. 4).

In addition to the magnitude, we can compare the spatial extent for power plants versus mobile sources. Wilson, 2003, found that half of the total exposure for iF(p), iF(as,SO<sub>2</sub>), and iF(an,NO<sub>x</sub>) was reached by a median distance of approximately 990 km for all three PM<sub>2.5</sub> components. For mobile sources, we found the median distance for the corresponding counties for three iF values to range between 150 and 640 km, signifying population exposures closer to the source as well as greater

differences between primary and secondary fine particles. Thus, better model resolution may not offer any additional utility in estimating power plant iFs, but might be necessary for mobile sources, especially in dense urban areas where much of the total exposure is captured close to the source county.

There are several limitations to this analysis, many of them to do with S-R matrix, the reducedform model that was the basis of the iF calculations. Firstly, S-R matrix had geographic and temporal resolution limitations. Geographically, S-R matrix resolution was limited to county-level. Counties vary in size across the US, with eastern counties tending to be smaller, thus having better model resolution. As concentration impacts are assumed to be spread equally over a county, some areas within the county will be underestimated while others will be overestimated, leading to exposure misclassification. If population distribution and concentration impacts are highly correlated, this model will underestimate the population exposure. Furthermore, the spatial extent of the iF may have been overestimated due to coarse geographic model resolution. In some urban areas where much of the total exposure occurs within the county, finer model resolution is necessary. However, S-R matrix served the purposes of this analysis by providing insight



Fig. 4. Ratio of mobile source to power plant intake fractions. For each distribution, horizontal lines on the box indicate the 25th, 50th (median), and 75th percentiles, while the dotted line indicates the mean. The dots outside the whiskers show the 5th and 95th percentile values.

about geographic areas where more resolved modeling might be needed, in the spirit of iterative risk assessment.

Temporally, S-R matrix was limited to annual average concentration impacts. Mobile source impacts vary diurnally and seasonally, neither of which is captured in the model. Previously mentioned local mobile source studies that presented higher iF results than this one made use of methods that better capture spatial and temporal resolution, or time-activity patterns. Our focus on only ambient concentrations may have underestimated total exposure from mobile sources. For example, a recent study reported that the total mass of bus exhaust inhaled by students commuting on a diesel bus was comparable in magnitude to the total mass of bus exhaust inhaled by everyone else in the SoCAB (Marshall and Behrentz, 2005), emphasizing the importance of microenvironments to mobile source exposures. However, it could be argued that ambient exposure at a representative site within a county is most meaningful for risk assessment at the present time. The two largest PM2.5 mortality epidemiological cohort studies commonly used in risk assessment applications rely upon annual average ambient concentrations from central site monitors (Pope et al., 2002; Dockery et al., 1993). In a risk assessment application, the iF multiplied by the emissions reduction and normalized by the

breathing rate can be combined directly with a mortality concentration-response function (assuming linearity) to estimate public health benefits of air pollution control.

In addition, the treatment of secondary sulfate and nitrate chemistry in S-R matrix was somewhat simplified. However, previous studies have shown that S-R matrix yields similar secondary PM2 5 intake fraction estimates as more complex models such as CALPUFF (Levy et al., 2003) or REMSAD (Abt Associates et al., 2000). Moreover, S-R matrix has been used by EPA in past regulatory impact analyses (US Environmental Protection Agency, 1999d), indicating that interpretation of its outputs could be useful. Another limitation of S-R matrix was the lack of adequate treatment of secondary organic aerosols (SOA). In more polluted areas of the US, organic carbon (OC), a mix of primarily and secondarily generated organic compounds, can contribute 10-40% of the PM2.5 mass (Seinfeld and Pandis, 1998). Although S-R matrix allowed us to estimate secondary ammonium sulfate and nitrate formation, it did not allow us to adequately estimate SOA formation from volatile organic compound reactions. Although it may be a non-negligible contributor to mobile source PM<sub>2.5</sub>, we would not expect the relative values of iF(p),  $iF(as,SO_2)$ ,  $iF(an, NO_x)$ ,  $iF(an, SO_2)$  to change with the inclusion of SOA impacts. Future studies should address SOA intake fractions, as well as other potentially important contributors to public health benefits from mobile source emission controls, such as ozone from NOx and VOCs.

More general concerns could be raised regarding the reliance on atmospheric dispersion models to estimate iF values, as opposed to relying on monitoring data and other empirical evidence. However, it would be difficult to extract from monitoring data the marginal contribution of a single source category in a single geographic location, and impossible in downwind areas where the absolute impact is smaller. Other approaches would also be unable to address critical questions regarding spatial extent and resolution. The primary form of validation and uncertainty characterization therefore comes from such features as calibrating model outputs to ambient concentration measures, as well as by verifying that iF estimates are similar with different dispersion models (Levy et al., 2003; Abt Associates et al., 2000).

Beyond S–R matrix, there are some additional limitations in interpreting our regression models. First, although the  $R^2$  values are quite high, especially for iF(p), it is possible that this is driven by the skewed distributions of iF since significant outliers may remain. Fig. 5 compares the iF(p)

predicted by the MLR model to the iF(p) calculated from Eq. (1). Most of the 3080 regression model outputs fall within a factor of 2 of the actual values. though some of the high iF(p) tend to be underestimated and some of the low iF(p) tend to be overestimated by the MLR model. Some coastal settings, for which a radial population parameter does not capture the distribution of exposed individuals downwind of the source county, have greater errors. Furthermore, an examination of the Pearson correlation coefficients revealed significant correlation between the exhaustive population predictors. Although this might inflate or deflate the standard errors, the parameter estimates themselves should be unaffected. Still, we developed simple linear regression models for each of the iFs versus each of the exhaustive population predictors and found results consistent with the MLR models.

Finally, the application of our estimates or regression models outside of the US should be undertaken with caution, as meteorology, population patterns, and myriad other factors impacting the intake fraction might be different. The uncertainty associated with our regression models has been understated by the  $R^2$  values reported in Table 3, as these values only represent the extent to which the chosen population predictors in the MLR



Fig. 5. Multiple linear regression model based on exhaustive population predictors versus Eq. (1) for primary  $PM_{2.5}$  intake fractions, iF<sub>j</sub>(p), in units of per million. The dashed 1:1 line represents perfect agreement between the MLR model and Eq. (1), while the dotted lines represent a factor of 2 from a perfect agreement. There are 3080 points plotted, most of them below 5 per million.

model explain the variation in  $iF_{j}$ , not agreement of our data with monitoring data or other forms of validation.

In spite of these limitations, our findings provide some important guidance for both public policy and future modeling efforts. For example, we can estimate the relative public health benefits of a 1 ton reduction in primary  $PM_{2.5}$ ,  $SO_2$ , and  $NO_x$  emissions by comparing emissions-weighted iF values for power plants and mobile sources (appropriate if such a reduction were distributed across the US in proportion to current emissions). Assuming a linear doseresponse function for PM<sub>2.5</sub> mortality where all particles have equal toxicity, we would expect the public health benefits of a 1 ton reduction of primary PM<sub>2.5</sub> emissions from mobile sources to be 2.7 times greater than from power plants. The corresponding ratios are 2.2 for  $SO_2$  emissions and 2.4 for  $NO_x$ emissions. However, it is important to remember that these ratios vary spatially across the US and any regional pollution control strategies need to take that into account. For example, although the 507 power plants emitted four times more SO<sub>2</sub> than the mobile sources in S-R matrix, there are some settings where the health benefit per unit emissions is more than four times greater for mobile sources, indicating that mobile sources should not be dismissed as an important contributor in some settings.

One of the primary goals of this analysis was to determine which types of atmospheric dispersion models to use in a risk assessment context. Our analysis indicates that national-scale dispersion models with county-level geographic resolution, such as S-R matrix, are appropriate for secondary  $PM_{2.5}$  or primary PM<sub>2.5</sub> emitted in rural areas, but that the substantial contribution of near-source populations to primary PM in urban areas warrant more resolved dispersion models to better inform risk-based regulatory decisions. In dense urban areas, in particular, near-source models with better resolution may be necessary to adequately capture the variation in mobile source iF that can occur within the county, as well as to yield an accurate estimate of the average iF for that county. In high-density settings, monitoring may provide a useful supplement to near-source models, if a significant portion of the iF were found to occur in close proximity to the source.

#### 5. Conclusions

This study has provided comprehensive estimates of primary and secondary  $PM_{2.5}$  mobile source

intake fractions across the US at county-level resolution. Mean primary PM2.5 iFs (on the order of 1 per million) were 1-2 orders of magnitude larger and exerted their impact closer to the county where mobile sources emissions originated than secondary PM<sub>2.5</sub> iFs. Since a good deal of the national primary PM2.5 iF exposure occurred close to the source county in dense urban areas, nearsource models with finer resolution may be necessary to better capture the variation in exposure at better geographic resolution. Multiple linear regression models using exhaustive population predictors explained a substantial amount of variation in national primary and secondary PM<sub>2.5</sub> iFs. Compared to power plants, the mobile source iFs tended to be larger and exhibit their impacts closer to where the emissions originated due to lower stack heights and co-location of populations with emission sources. The use of a national-scale county-resolution model may be inappropriate for mobile source primary PM<sub>2.5</sub> iF in dense urban areas, but sufficient for secondary PM<sub>2.5</sub> iF and for power plant iFs.

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