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Models for predicting the ratio of particulate pollutant concentrations inside vehicles to roadways

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Abstract

Under closed-window driving conditions, the in-vehicle-to-outside (I/O) concentration ratio for traffic-related particulate pollutants ranges from nearly zero to one, and varies up to five-fold across a fleet of vehicles, thus strongly affecting occupant exposures. Concentrations of five particulate pollutants (particle-bound polycyclic aromatic hydrocarbons, black carbon, ultrafine particle number, and fine and coarse particulate mass) were measured simultaneously while systematically varying key influential parameters (i.e., vehicle type, ventilation, and speed). The I/O ratios for these pollutants were primarily determined by vehicle air exchange rate (AER), AER being mostly a function of ventilation setting (recirculation or outside air), vehicle characteristics (e.g., age, interior volume) and driving speed. Small (±0.15) but measurable differences in I/O ratios between pollutants were observed although ratios were highly correlated. This allowed us to build on previous studies of ultrafine particle number I/O ratios to develop predictive models for other particulate pollutants. These models explained over 60% of measured variation, using ventilation setting, driving speed, and easily-obtained vehicle characteristics as predictors. Our results suggest that I/O ratios for different particulate pollutants need not necessarily be measured individually and that exposure to all particulate pollutants may be reduced significantly by simple ventilation choices.

1.0 Introduction

The in-vehicle microenvironment contributes disproportionally to traffic-related pollutant exposure. For example, Dons et al. (1) measured black carbon exposure for 62 subjects and found that the transport microenvironment was the highest-ranking contributor to the total exposure, with 21% of exposure resulting from less than 7% of daily time spent in transport. Wu et al. (2) found that time spent inside vehicles was most strongly predictive of overall particle-bound polycyclic aromatic hydrocarbon exposure in a cohort of 28 non-smoking women, explaining 48% of total variation in daily exposure and 39% of the variation in individual level exposure.

At the scale of epidemiological studies, however, it is not practical to directly measure pollutant concentrations, so predictive models are needed. Predictive models of on-road

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6. Supporting Information The instrument used are listed and corrections made to improve inter-instrument agreement are discussed (S.1). Vehicles tested and relevant details are listed (Table S.2). Predictive equations for AERs and UFP I/O ratios at the two ventilation settings are presented in Section S.3. Comparison to other studies (S.4) and model residuals (S.5) have also been presented. This material is available free of charge via the Internet at http://pubs.acs.org.

7. Brief Concentration of traffic-related particulate pollutants inside vehicles ranges from 10 to nearly 100% of roadway concentrations, but this fraction is accurately predictable and most strongly determined by ventilation setting.

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concentrations have been attempted with varying degrees of success (3), but exposure estimates also need to take into account differences in penetration rates and the losses that occur inside vehicles for particulate pollutants. Our previous work (4) showed inside-to-outside (I/O) ratios for ultrafine particle number concentrations vary from nearly zero to one between vehicles as well as across operating conditions for a given vehicle, resulting in widely varying personal exposures. Ultrafine particle I/O ratios depended primarily on air exchange rate (AER), which is a function of vehicle characteristics, speed, and ventilation setting. We previously explained 79% of the variability in measured UFP I/O ratios using easily obtainable information such as ventilation settings (i.e., recirculation or outside air selection and fan setting), vehicle age and driving speed (4).

I/O ratios for other particulate pollutants, outside of ultrafine particles, have been seldom quantified in the literature, but Ott et al. (5) measured cigarette-derived fine particle mass decay rates inside vehicles and found that they correlated with AER. They also reported a fine particulate matter I/O ratio of 0.43, averaged across three vehicles and different ventilation settings. Because particle loss rates vary by particle size (6), I/O ratios do as well, but potentially in a complex way, due to non-linearity in loss mechanisms and differences in particle size distributions for different pollutants. The goal of this study was to measure I/O ratios of several particulate species of different sizes (particle-bound polycyclic aromatic hydrocarbons [PB-PAHs], black carbon [BC], ultrafine particles [UFP], and $PM_{2.5}$ and PM_{10} [particulate matter ≤ 2.5 and 10 μm, respectively]); to examine the influence of driving and vehicle characteristics on these I/O ratios; and to develop predictive models for each pollutant's I/O ratios.

2.0 Methods

2.1 Instruments and Inside-to-Outside (I/O) ratio calculation

Simultaneous measurements of inside and outside concentrations were conducted with paired instruments. The I/O ratio was calculated as the ratio of the average inside concentration to the average outside concentration for the duration of the run, typically 20–30 minutes. Instruments used were an Ecochem PAS2000 for PB-PAH, a Micro Aethalometer AE 51 for BC (Aeth Labs), a TSI 3007 condensation nucleus counter for UFP, and a TSI DustTrak 8532 and 8530 for $PM_{2.5}$ and PM_{10}. Details are provided in Table S1 in the Supporting Information (SI). Instrument clock times were regularly synchronized to be within 1 second to the Global Positioning System (GPS) device time (Garmin GPSMAP 76CSC), which also recorded speed. Instruments logged data at different intervals (1–10 seconds), and later all data were averaged over 10 seconds. Data were also aligned with the fastest instrument to account for differences in response times.

Quality assurance procedures included regular flow and zero checks. Side-by-side tests of instrument agreement were conducted at regular intervals including the beginning and end of tests for each vehicle under either ambient conditions or in a moving vehicle with inlets tied together. The results from these tests and any adjustments necessary to improve inter-instrument agreement are discussed in the SI (Section S.1.1, Figures S1 and S2). Briefly, particle concentration data were adjusted for inlet losses and instrument-to-instrument differences in response were typically in the range of 5–8%. Correction was performed by ordinary linear regression, adjusting one instrument to match the other. No corrections were necessary for the two PAH analyzers (PAS 2000). The two Dusttraks (DRX 8532 & 8533) were additionally calibrated against a BAM-1020 (Beta Attenuation Monitor) at a South Coast Air Quality Management District's air quality monitoring site for $PM_{2.5}$ and $PM_{10.}$

2.2 Vehicles and ventilation conditions tested and AER estimation

Six vehicles were selected to reflect different age, mileage, volume and manufacturer; these factors have previously been identified as significant determinants of a vehicle's AER and UFP I/O ratios (4, 7–8). Relevant vehicle characteristics are summarized in the SI (Table S2). Furthermore, vehicles were purposefully selected to provide a range of UFP I/O ratios and AER that are typical of the fleet-wide differences under real driving conditions (see Figure S5 in SI). Measured I/O ratios were well distributed over the range of I/O ratios previously measured in a wider selection of 43 vehicles selected to represent U.S./California fleet distribution (see Figure S6 in SI).

AERs were estimated using the models developed in our previous work (4) that were able to account for 68% of the variation in observed AERs under recirculation conditions and 79% under outside air intake conditions. The predictive equations are given in Section S.3 in the SI (Equations 1 & 2). The AERs at typical arterial and freeway driving speeds covered most of the range expected in the U.S. fleet (See Figure S5 in the SI). For reference, under recirculation (RC) conditions—a low-AER condition under which in-cabin air is re-circulated through the cabin—AERs for an inter-quartile range of fleet age (four- to 11-year-old vehicles) ranged from 2.8 to 4.5 h^{-1} at 35 mi h^{-1} and 5.0 to 7.9 h^{-1} at 65 mi h^{-1}, respectively. Under outside air (OA) setting—a high-AER condition under which outside air is mechanically drawn into the vehicle cabin—the corresponding estimated AERs were 72 and 83 h^{-1} for vehicles travelling at 35 and 65 mi h^{-1}, respectively, at the middle fan setting, roughly an order of magnitude higher than under RC conditions. (Under OA ventilation settings, fan speed is more important than vehicle speed.)

Measurements were conducted in all six vehicles with closed windows under both RC and OA ventilation conditions. The `Max AC' option present in many vehicles re-circulates cabin air. The automatic climate-control option is difficult to generalize vehicle-to-vehicle and was not tested in this study. All vehicles tested were equipped with standard, coarse air filters of low particle collection efficiency. At each ventilation setting, experiments were conducted at medium and high ventilation fan setting, as well as with the fan off. Some tests were also conducted with windows open but I/O ratios for open windows were essentially 1.0 for all but very low (< 15 mi h^{-1}) speeds.

2.3 Test speeds

Tests were conducted while driving on Los Angeles freeways (Interstates 5, 10, 710 and 110) and major arterial roads (Wilshire and Sunset Boulevards and Jefferson and Western Avenues). (Freeway is defined as a `divided highway with full control of access with grade separations at intersections,' and arterial is defined as having `intersections at grade and direct access'). Median driving speeds for the data set modeled were 57 mi h^{-1} (IQR: 52 – 60) for freeway driving and 21 mi h^{-1} (IQR: 17 – 24) for arterial road driving. Average coefficients of variations for freeway and arterial road driving were 63% and 16% respectively. The higher variation on arterial roads was due to traffic signals and stop signs. These variations in speed led to varying AERs during tests, but previous measurements (4, 7–8) have shown that the impact of speed on AER and I/O ratios is nearly linear in the arterial road speed range, and the average of I/O ratios measured at varying speeds matches closely with the I/O ratio measured at the same constant average speed. However, to account for slight non-linearity in AER at higher speeds (7), only freeway runs with standard deviations of speed less than 20% were used. Furthermore, the stable period of a qualifying freeway run needed to exceed six minutes, and the standard deviation of the measured I/O ratio in that period could not exceed 0.05. Of 231 runs analyzed, 64 met these criteria.

2.4 Predictive models

I/O ratios were modeled with a logit transformation (ln [I/O / (1-I/O)]), an appropriate transformation for fractional ratios because absolute changes in such ratios become compressed near zero and 1.0 where good model resolution is desirable. Predictive models were developed using the following candidate independent variables: measured UFP I/O ratio, predicted UFP I/O ratio, predicted AER, ventilation fan strength (notch fraction of maximum number of notches), vehicle cabin volume (ft^3), vehicle age (years), mileage (thousands of mi), speed (mi h^{-1}), along with pair-wise interactions between vehicle speed, volume, age, fan setting and UFP I/O ratios, and higher order terms, similar to procedures described in more detail in Hudda et al. (4).

Multiple measurements of I/O ratio were performed in each vehicle at different speeds and/ or ventilation settings. Consequently, repeated measurements were sometimes correlated. This correlation violates the assumption of completely independent observations in multiple linear regression (MLR) models, and while MLR models fit to correlated data have unbiased regression coefficients, they will have incorrectly tighter standard errors (9). To account for correlated observations, we used Generalized Estimating Equation (GEE) models (10) for continuous outcomes, including an exchangeable correlation structure between vehicles (which assumes that there is a single, uniform correlation between repeated measurements from the same vehicle, after controlling for the predictor variables) and robust sandwich estimates of regression coefficient standard errors (which produces valid standard errors even if the covariance structure is mis-specified).

All-subset MLR was used to identify the most important set of predictor variables. From this set, a GEE model was developed. For comparison, MLR models were also fit. Residuals were inspected to assess model assumptions of linearity, normality, and homoskedasticity (equal variance).

3.0 Results and Discussion

3.1 Concentration variability and I/O ratio determination

Reaching stable I/O ratios during varying on-road concentrations required careful consideration of run-wide averages and a stable convergence of I/O ratios (which reflects adequate sampling duration). Figure 1 illustrates the typical relationship between inside and outside concentrations. Here, a 2010 Honda Civic was driven at freeway speeds, 61 mi h^{-1} under RC conditions (left) and 65 mi h^{-1} under OA conditions (right). The fan was set to high to maximize AER. The predicted AER under RC was 8 h^{-1} and under OA was 122 h^{-1}, using previously developed predictive models for AER (4), listed in Section S.3. The PB-PAH roadway concentrations of about 42 ng/m^3 resulted in only 13 ng/m^3 inside the vehicle during RC conditions and concentrations nearly identical to roadway concentrations under OA conditions.

Figure 1 shows that although the instantaneous on-road PB-PAH concentrations are highly variable, these perturbations are dampened in the running cumulative average on-road concentration (middle panel) time series, which was relatively smooth after ten to fifteen minutes. Similarly, the running cumulative average in-vehicle concentration was also stable after a few minutes, and the resulting I/O ratio based on cumulative average concentrations converged to a stable value and stayed stable despite large variations in roadway concentrations. It is interesting to note that the fifteen-fold increase in AER led to a three-fold increase in I/O ratio; the bottom panel in Figure 1 illustrates corresponding I/O ratios for the other particulate pollutants, all similarly impacted by the large differences in AER.

To estimate the uncertainty in the I/O ratios due to measurement error, we assumed 20% accuracy for the UFP particle number concentrations (as stated by the manufacturer) and 10% for the PB-PAH and particle mass concentrations (author's assumption, not stated by the manufacturer). Using mean square error propagation analysis, the maximum uncertainty in I/O ratios was 30%, 14%, 14% for UFP number and PB-PAH and particle mass concentrations, respectively.

3.2 Influence of ventilation setting

Our results verified that AER plays just as critical a role in determining other particulate pollutants' I/O ratios as it does in determining UFP I/O ratios (see previous studies 4, 8, 11). Under open window conditions, even as little as an inch or two at typical driving speeds, AER is sufficiently high for pollutant loss rates to be negligible in comparison to influx rates, resulting in I/O ratios indistinguishable from 1.0 and providing little reduction in exposure to roadway concentrations. Under ventilation conditions of low AER such as RC setting, however, in-vehicle concentrations were much lower than under higher AER ventilation conditions such as OA setting. This directly follows from a low AER reducing the influx of pollutants into the cabin while at the same time increasing particle residence time and possibility of loss to surfaces within the cabin. Ventilation setting, being the foremost determinant of AER, is also the most significant determinant of I/O ratios. The top panel of Figure 2 illustrates the stark difference between the I/O ratios under RC and OA conditions for all the vehicle-speed combinations tested. I/O ratios under RC were mostly 0.1–0.4 while under OA they ranged from 0.3–0.9.

Particulate pollutant I/O ratio rank order differed by ventilation setting, likely driven by particle momentum effects. $PM_{2.5}$ and PM_{10} showed higher losses (i.e., lower I/O ratios) than UFP under OA conditions, but lower losses (higher I/O ratios) under RC conditions (See Figure 2 (b)). Under OA setting, the dominant air influx pathway is the ventilation air intake system, and the higher flow velocities and changes in flow direction likely produced greater impaction losses for larger particles. Although this study did not differentiate between losses in the ventilation system duct work and losses in the coarse filter, these results suggest that the combination of the two loss mechanisms affects $PM_{2.5}$ and PM_{10} more than UFP or PB-PAHs under high-AER, low-residence-time conditions. Cabin filters have been shown to have negligible removal efficiency for UFPs (8). In contrast, under RC conditions, lower AERs and longer residence times appear to make cabin surface losses relatively more important and the higher diffusion losses of the smaller particles likely contributed to lower UPF and PB-PAH I/O ratios compared to $PM_{2.5}$ and PM_{10} ratios.

3.3 Correlation between particulate pollutant I/O ratios

Simultaneously measured I/O ratios for different pollutants showed strong correlation, as shown in Figure 3. Inter-pollutant correlation coefficients (r) exceeded 0.85 and measured UFP I/O ratios alone explained greater than 70% of the variation in I/O ratios of other pollutants ($r^2 = 0.77$ for PB-PAH, 0.77 for $PM_{2.5}$ and 0.74 for PM_{10} using ordinary least square regression). Figure 3 also exhibits the positive correlation between measured I/O ratios and the estimated AER and I/O ratios (lower panel).

The effect of particle size on I/O ratios is potentially more important under RC conditions where I/O ratios themselves are small. For example, the inter-quartile range of difference between UFP, PB-PAH and $PM_{2.5}$ I/O ratios under RC conditions is about 0.15–0.2, so a 0.1 change translates into at least a 50% change in I/O ratio and a 50% change in in-vehicle concentration. So although I/O ratios are similar between pollutants and strongly correlated, it is important to take the effect of particle size into account at low AER conditions.

3.4 Black Carbon concentration inside vehicles

It was not possible to measure BC I/O ratios in this study as accurately as for the other pollutants. This was because the generation of Micro Aethalometer used was sensitive to mechanical vibrations if used at high time resolution, especially in motion when mechanical shocks are frequent. BC was measured at 60 seconds (as opposed to 1–10 seconds for other pollutants) which reduced but did not eliminate the incidence rate of concentration spikes due to vibration. These spikes usually consisted of a readily-identifiable positive and negative pair of extreme values. Figure S3 (a) in the SI shows a typical time-series plot before censoring two vibration spikes. Figure S3 (b) shows the same time-series after censoring and re-aligning the concentration axes with PB-PAH concentrations. Only runs with greater than 95% data completeness after removing vibration spikes were considered for analysis. However, due to frequently low BC concentrations (e.g., below 1 $\mu g/m^3$), the signal-to-noise ratio was often too low to produce adequate inter-instrument agreement for accurate I/O ratios. However, enough measurements were available to compare BC I/O ratios to other pollutants on a semi-quantitative basis. Typical time-series that qualified for such comparison under both OA and RC ventilation settings are shown in Figure S4 in SI (test vehicle was Ford Focus).

Because on-road PB-PAH size distributions are the most similar in size to BC size distributions and somewhat larger than UFP size distributions (12), we would expect BC I/O ratios to be similar to both PB-PAH and UFP I/O ratios, and perhaps closer to PB-PAH. Figure 4 shows instantaneous BC I/O ratios measured during a run compared to run-wide average UFP and PB-PAH I/O ratios. For RC conditions, there was no distinguishable difference in I/O ratios between these pollutants, but for OA conditions, BC appeared to behave more like PB-PAH than UFP (i.e., somewhat lower I/O ratios). Therefore, under OA conditions, I/O ratios for BC are best approximated by PB-PAH's.

3.5 Predictive models for I/O ratios

Because predicted UFP I/O ratios from our previous models (4) were used as inputs in models presented in this section, it was important to evaluate the agreement of these previous predictive models for the vehicles used in this study, since the number of study vehicles was smaller and any significant deviation of true versus predicted UFP I/O ratios for a particular vehicle might introduce bias to the models. Agreement between new measurements and model predictions appeared acceptable. Deming regression gave the equation: Predicted logit(UFP I/O) = 0.75 × Measured logit(UFP I/O) + 0.097. (Deming regression was used since the predicted and measured UFP I/O ratios have independent errors). The average residual (measured minus predicted) was only −0.04, with the inter-quartile range being −0.12 to 0.07 (see Figure S6 in the SI).

I/O ratios for PB-PAH, $PM_{2.5}$ and PM_{10} were modeled using driving speed, interior volume, vehicle age and predicted UFP I/O, the UFP I/O ratio itself being a linear combination of ventilation setting (RC or OA), fan strength, age and speed (see Equations 3 and 4 in SI Section S.3). The resulting MLR and GEE models explained 74% of the variation in measured I/O ratios of PB-PAH, 62% of $PM_{2.5}$ under RC, 62% of $PM_{2.5}$ under OA, 66% of PM_{10} under RC and 64% of PM_{10} under OA. The GEE model based predictive equations are as follows:

Equation 1: PAH I/O Ratio under RC

$$logit\ (I/O_{PAH}) = -2.56 + (0.20 \times Age) + (0.020 \times Speed) + (0.44 \times \text{Fan Setting})$$

Equation 2: PAH I/O Ratio under OA

$$logit\ (I/O_{PAH}) = -0.18 + (0.13 \times Age) + (0.020 \times Speed) + (0.44 \times \text{Fan Setting})$$

Equation 3: $PM_{2.5}$ I/O Ratio under RC

$$logit\ (I/O_{PM2.5}) = -2.05 + (0.23 \times Age) + (0.020 \times Speed) - (0.41 \times \text{Fan Setting})$$

Equation 4: $PM_{2.5}$ I/O Ratio under OA

$$logit\ (I/O_{PM2.5}) = 0.54 + (0.0056 \times Age) + (0.0079 \times Speed) + (0.17 \times \text{Fan Setting}) - \left(0.085 \times [Vol - 97] + 0.010 \times [Vol - 97]^2\right)$$

Equation 5: PM_{10} I/O Ratio under RC

$$logit\ (I/O_{PM10}) = -2.13 + (0.21 \times Age) + (0.025 \times Speed) - (0.57 \times \text{Fan Setting})$$

Equation 6: PM_{10} I/O Ratio under OA

$$logit\ (I/O_{PM10}) = 0.025 + (0.0087 \times Age) + (0.012 \times Speed) + (0.27 \times \text{Fan Setting}) - \left(0.089 \times [Vol - 97] + 0.0092 \times [Vol - 97]^2\right)$$

Model predicted I/O ratio values have been plotted in Figure 5 against measured I/O ratios to illustrate the goodness of model fit. Coefficients, confidence intervals and relevant statistics for both MLR and GEE models are presented in Table 1. Summary statistics for model residuals (signed and absolute differences and percentage differences) are presented in Section S.5 in the SI.

Figure 6 shows the predicted I/O ratios plotted against two influential predictive variables, age and speed. Under both conditions fan setting was relatively less important and strongly influential only for UFP under OA setting (4). Volume was found to significantly impact I/O ratios for PM under OA settings; however, volume variation was limited in this study to the typical range of sedans (89 –104 ft^3), but not SUVs. Therefore, the model predictions for OA condition PM I/O ratios should be used with caution for larger vehicles. (Caution is also warranted for vehicle age exceeding 12 years or speeds over 75 mi h^{-1}, the upper limits of the vehicles or conditions tested.) It should also be pointed out that the I/O ratios decreased with increasing volume, in agreement with OA AER models reported in earlier work (4). This effect can be seen in the rightmost plot of Figure 6 for the two $PM_{2.5}$ surfaces for vehicle volumes 90 and 105 ft^3.

Ventilation mode, vehicle volume and age are easily and accurately attainable via questionnaire (vehicle volume by knowing vehicle model). If questionnaire data are used to determine ventilation choice, it is important to validate this key variable. Fan setting and speed are, however, more difficult to accurately assess. For the range of data shown in Figure 6, a switch in fan from midway (0.5) to lowest or highest fan setting is expected to change I/O ratios for PB-PAH and PM on the order of ±0.05. For a median aged vehicle (7 yr, 100 ft^3 at medium fan), if speed is measured, a ±10% uncertainty in speed will also lead to ±0.05 uncertainty in I/O ratios for all pollutants except UFP under RC setting at higher speeds where the uncertainty would be ±0.1.

3.6 Expected I/O ratios in vehicle fleet

A wide range of I/O ratios results when distributions of influential factors (i.e., age, interior volume, driving speed during rush hour and ventilation fan setting) are combined to simulate the distribution of I/O ratios in the current U.S. fleet of vehicles. Expected distributions are plotted in Figure 7 for a sedan-type fleet, similar in its age distribution to the U.S. fleet while travelling during rush hour (see section S.2 in SI for further details on distribution of age, speed and other factors). The median I/O ratio under OA is several-fold higher than under RC (4-fold for UFP, 2.5-fold for PB-PAH and 2.0-fold for $PM_{2.5}$). Thus, a switch in ventilation setting from OA to RC will more effectively reduce exposure to UFP or PB-PAH compared to $PM_{2.5}$ or PM_{10}.

The inter-quartile range of I/O ratios (i.e., the range in I/O ratio resulting from variation in vehicle characteristics and driving speeds) also differed by pollutant, its value ranging from 0.1 to 0.3. For UFP and PB-PAH this inter-quartile range was less than that due to ventilation setting change (although for $PM_{2.5}$ it was comparable). Thus, the inter-quartile range of I/O variation due to vehicle and speed factors was either comparable to or less than the difference caused by ventilation setting alone. Overall, this simulation indicates that exposure to traffic related pollutants may vary several-fold even for the same roadway environment and drive time, owing to differences in ventilation settings, driving speed and vehicle characteristics. For example, the highest quartile I/O ratios of the fleet under OA conditions will, on average, experience in-vehicle UFP concentrations that are 5.5 times that of in-vehicle concentrations for the lowest quartile I/O ratios of the fleet under RC. This factor would be about 3.7 for PB-PAH and about 3.0 for fine and coarse particulate matter.

4.0 Implications

This work demonstrated that the factors that increase the air exchange rate (AER) of a vehicle also increase I/O ratios across all particulate pollutants. Thus, ventilation setting, the most important factor in determining AER, is critical to estimating in-vehicle exposures to any type of particulate pollution. Often in studies comparing exposures across travel modes (13) or studies quantifying exposure contributions of different microenvironments (1), ventilation setting of the vehicle is not taken into consideration. This lack of ventilation setting information severely limits the generalizability of the results of such studies.

The models developed in this study are useful for allowing estimation of I/O ratios for PB-PAH, $PM_{2.5}$ and $PM_{10.}$ (BC I/O ratios appeared to be similar to those for PB-PAH.) Overall, our results show that under identical roadway concentrations, a two-to five-fold difference in exposure inside vehicles can be expected due to differences in ventilation setting, speed, and vehicle characteristics such as age. Limited measurement data from other independent studies is available to compare our models but $PM_{2.5}$ model predicted I/O ratios were compared to Ott et al. (5) measurements and results agree, on average, 0.47 versus 0.55, respectively, and linear regression gives an r^2 of 0.79 and a slope within 10% of 1.0 (see Section S.4 in SI).

It is not feasible in large cohort studies to conduct in-vehicle measurements for all subjects, and in smaller panel studies it is difficult to measure in-vehicle exposures for multiple pollutants. Therefore, our models that can predict I/O ratios for multiple pollutants based on information that can be gathered through questionnaires can aid exposure assessment. However, if questionnaires or other self-reports are used to determine ventilation setting, these results should be validated in a sub-sample using independent measures such as CO_2, which will vary strongly with ventilation setting (7) and knowledge of the quartile of fan setting used or at least low, medium, high/full categories is suggested. Studies that investigate the effect of ambient weather conditions and people's ventilation and window opening choices can also be useful references (14). Most of the other variables in our predictive models are likely to be reliable via self reporting, including vehicle age, mileage, and make and model (to ascertain interior volume). However, travel activity information, unless gathered via GPS, will likely contribute additional uncertainty. To reduce this travel activity uncertainty, self reports of typical commute duration, time of day and speed, for example, can be validated with publicly available roadway speed datasets (like California Department of Transportation's Performance Management System), and estimates of total driving can be compared to odometer and mileage accrual estimates.

On-road concentration data, directly measured or predicted, are necessary to accurately translate our I/O models into estimates of exposure. Many urban areas now have some amount of on-road measurements, though meteorological and roadway representativeness needs to be carefully considered in each case. Current predictive models of on-road concentrations such as Fruin et al. (3) or Li et al. (15) were able to explain 60–90% of variance in UFP, BC, $PM_{2.5}$ and PB-PAH concentrations on arterial roads and freeways in Los Angeles. The variables used are readily obtainable in most urban areas, but they have not been tested for generalization across different urban areas. This is an important research question to explore.

Accounting for variations in in-vehicle exposure to different particulate pollutants can also help address the multi-pollutant correlation challenge in air pollution epidemiology in two ways. First, there are remarkably different pollutant concentrations and pollutant mixtures on arterial roads and freeways, in part due to large differences in truck traffic (3, 15). Therefore, accounting for the various mixes of people's arterial and freeway drive time will tend to decrease the inter-pollutant correlations of estimated exposures. Second, our I/O models predict somewhat different changes in I/O with vehicle and driving characteristics for different pollutants, thus also contributing somewhat to reduced correlation in exposures.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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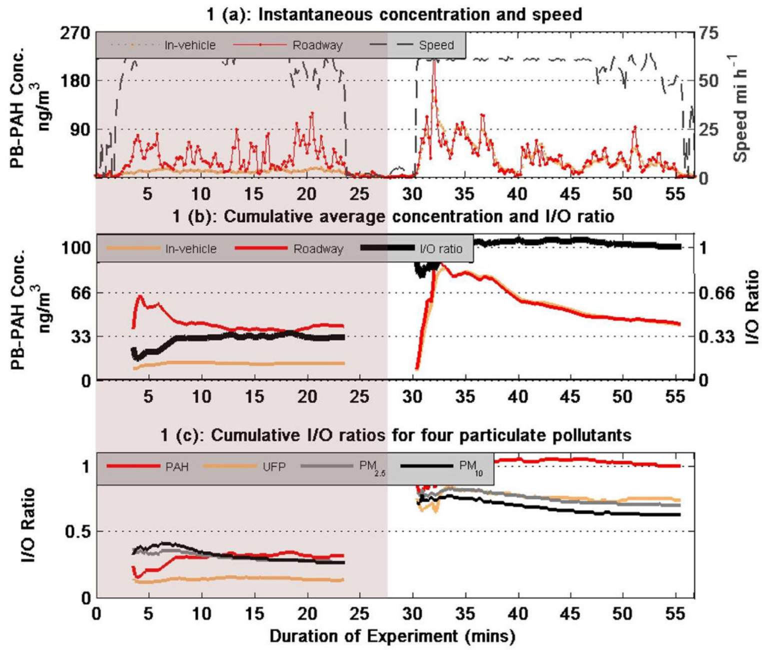

Figure 1.

Time-series for roadway and in-vehicle pollutant concentrations and I/O ratios under the two ventilation conditions tested. The portion of run under recirculation setting has been shaded.

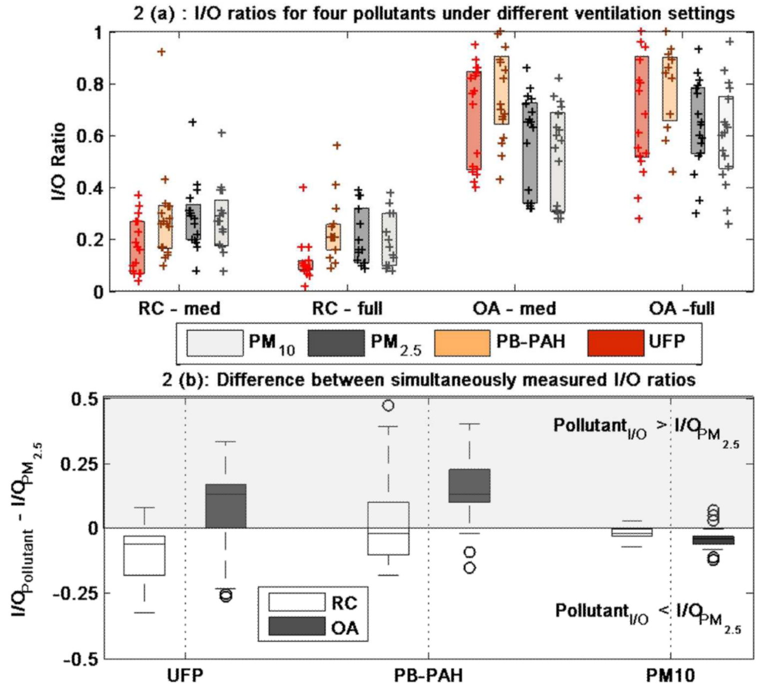

Figure 2.

I/O ratios for four pollutants measured under different ventilation settings and under medium (med) and full fan setting.

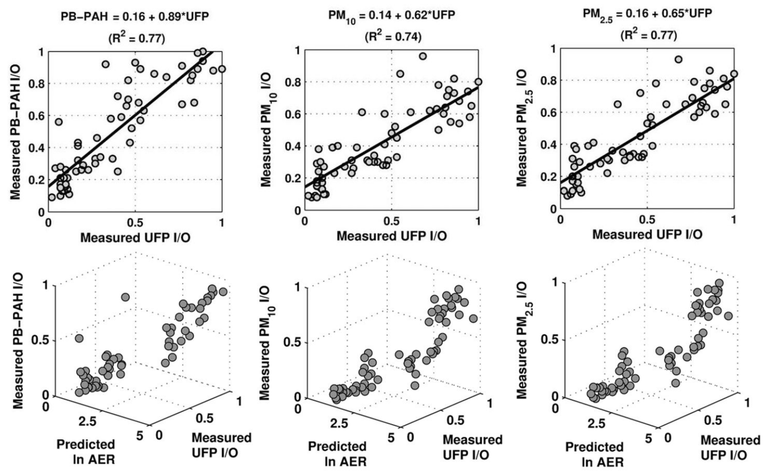

Figure 3.
Correlation among simultaneously measured I/O ratios for four pollutants and trend with AER.

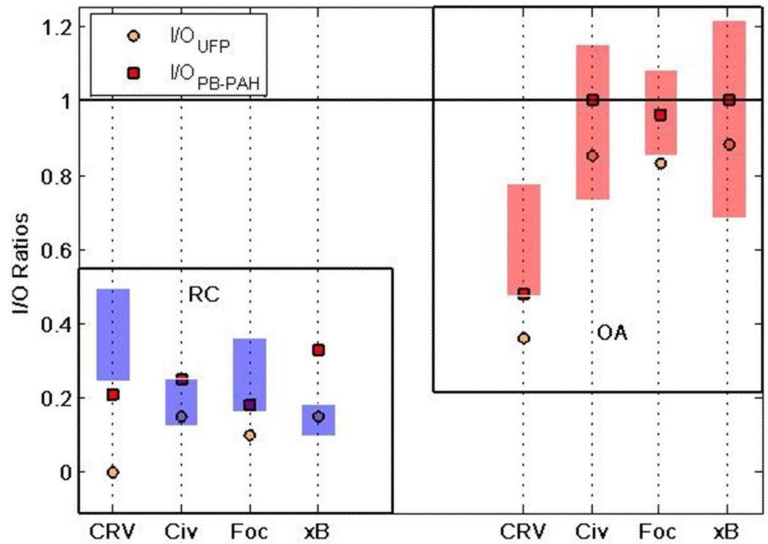

Figure 4.

BC I/O ratio (solid blocks) compared to UFP and PB-PAH I/O ratios under RC setting (left) and OA setting (right).

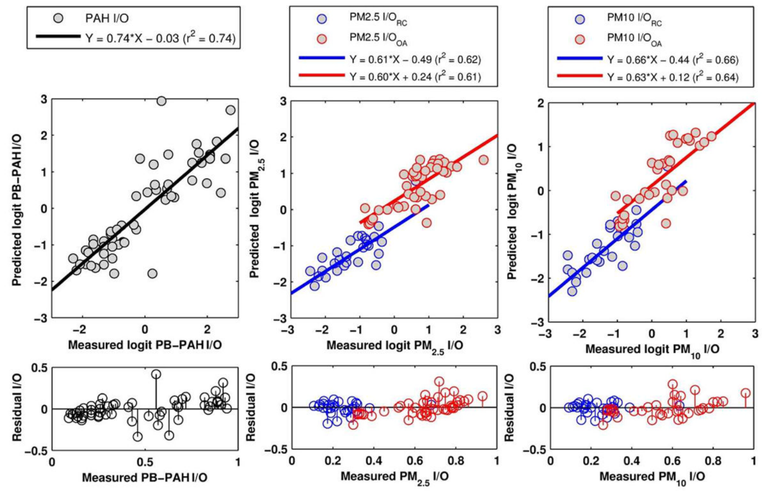

Figure 5.

Model predictions vs. measured logit I/O ratios and illustration of agreement (linear fits) between predictions and modeled variable (logit I/O ratio). Model residuals have been plotted against measured I/O ratios.

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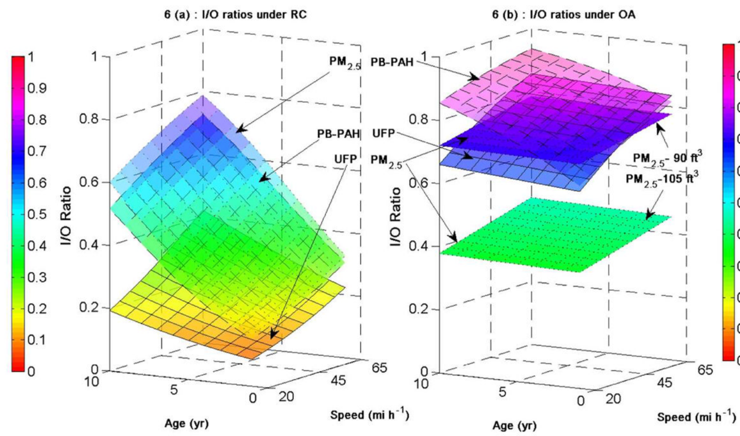

Figure 6.
Model predictions plotted against predictive variables (age and speed) underRC setting (left) and OA setting (right).

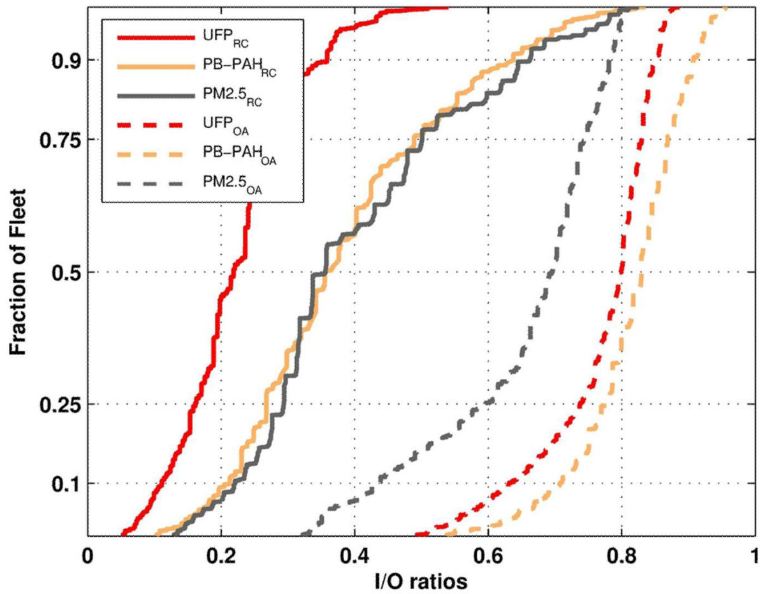

Figure 7.

Predicted distribution of I/O ratios of traffic related pollutants in U.S. type sedan fleet during rush hour diving.

Table 1

Coefficients, p-values and confidence intervals for logit (I/O) predictive models for PB-PAH, $PM_{2.5}$ and PM_{10}

	Pollutant	Variable	Coefficient	Std. Error	t-value / Wald	p-value / Pr>(IWI)	95% CI	
							lower limit	upper limit
GEE	PB-PAH	Intercept	0.06	0.08	0.72	4.71E-01	−0.10	0.21
		Predicted Logit UFP I/O	0.81	0.08	10.50	0	0.66	0.96
		Age (years)	0.12	0.01	11.28	0	0.10	0.14
	PM2.5 under RC	Intercept	−4.96	0.50	−9.89	0	−5.94	−3.98
		Predicted Logit UFP I/O	−0.93	0.17	−5.42	5.92E-08	−1.27	−0.59
		Age (years)	0.31	0.020	15.88	0	0.27	0.35
		Speed (miles/hour)	0.043	0.0065	6.61	3.76E-11	0.030	0.056
	PM2.5 under OA	Intercept	−0.63	0.32	1.99	0.047	0.009	1.25
		Predicted Logit UFP I/O	0.32	0.14	2.35	0.019	0.054	0.58
		[Vol-97] (cubic feet)	−0.085	0.013	−6.78	1.24E-11	−0.11	−0.061
		$[\text{Vol-97}]^2$ (cubic $\text{feet})^2$	−0.010	0.0052	−1.98	0.048	−0.020	−9.4E-05
	PM10 under RC	Intercept	−5.57	0.45	−12.25	0	−6.46	−4.68
		Predicted Logit UFP I/O	−1.06	0.18	−5.88	4.15E-09	−1.42	−0.71
		Age (years)	0.32	0.02	18.21	0	0.28	0.35
		Speed (miles/hour)	0.051	0.0056	9.01	0	0.040	0.062
	PM10 under OA	Intercept	0.17	0.44	0.38	0.71	−0.70	1.03
		Predicted Logit UFP I/O	0.50	0.17	2.92	3.5E-03	0.16	0.83
		[Vol-97] (cubic feet)	−0.089	0.0084	−10.62	0	−0.11	−0.073
		$[\text{Vol-97}]^2$ (cubic $\text{feet})^2$	−0.009	0.0067	−1.37	0.17	−0.022	0.0039
MLR	PB-PAH	Intercept	0.06	0.12	0.45	0.66	0.0032	0.11
		Predicted Logit UFP I/O	0.81	0.07	11.54	3.35E-16	0.047	1.57
		Age (years)	0.12	0.04	2.89	5.60E-03	0.0069	0.23
	PM2.5 under RC	Intercept	−4.75	1.88	−2.53	0.018	−0.27	−9.23
		Predicted Logit UFP I/O	−0.85	0.68	−1.24	0.23	−0.05	−1.65
		Age (years)	0.31	0.074	4.15	3.34E-04	0.018	0.60
		Speed (miles/hour)	0.042	0.0165	2.55	0.018	0.0024	0.082
	PM2.5 under OA	Intercept	0.59	0.22	2.73	9.3E-03	1.15	0.03
		Predicted Logit UFP I/O	0.38	0.15	2.60	0.013	0.022	0.74
		[Vol-97] (cubic feet)	−0.084	0.013	−6.49	8.6E-08	−4.8E-03	−0.16
		$[\text{Vol-97}]^2$ (cubic $\text{feet})^2$	−0.011	0.0044	−2.48	0.017	−0.021	−6.2E-04
	PM10 under RC	Intercept	−5.56	1.82	−3.05	5.31E-03	−0.32	−10.80

	Pollutant	Variable	Coefficient	Std. Error	t-value / Wald	p-value / Pr>(IWI)	95% Cl	
							lower limit	upper limit
		Predicted Logit UFP I/O	−1.06	0.66	−1.61	0.12	−0.060	−2.06
		Age (years)	0.32	0.07	4.42	1.68E-04	0.018	0.62
		Speed (miles/hour)	0.051	0.0160	3.17	3.97E-03	2.9E-03	0.099
	PM10 under OA	Intercept	0.17	0.36	0.46	0.65	0.32	0.01
		Predicted Logit UFP I/O	0.51	0.18	2.79	9.2E-03	0.029	0.98
		[Vol-97] (cubic feet)	−0.089	0.016	−5.38	8.1E-06	−5.1E-03	−0.17
		$[\text{Vol-97}]^2$ (cubic feet)^2	−0.0094	0.0065	−1.44	0.16	−0.018	−5.4E04