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Impacts of freeway traffic conditions on in-vehicle exposure to ultrafine particulate matter

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HIGHLIGHTS

► Vehicle ventilation is the dominant influence on in-vehicle UFP.
► Traffic states have a small but significant impact on in-vehicle UFP.
► Traffic impacts vary by vehicle ventilation and can be non-linear or lagged.
► Individual high-emitters eclipse aggregate traffic flow effects on UFP.
► Congestion *per se* does not increase in-vehicle UFP exposure concentrations.

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ABSTRACT

There is evidence of adverse health impacts from human exposure to traffic-related ultrafine particulate matter pollution. As more commuters are spending a significant portion of their daily routine inside vehicles, it is increasingly relevant to study exposure levels to harmful pollutants inside the vehicle microenvironment. This study is one of the first research efforts to combine detailed freeway traffic data (at 20 s intervals) and in-vehicle ultrafine particulate (UFP) exposure data under varying vehicle ventilation conditions. Results show that due to negative correlation between traffic speed and density, traffic states have a small but significant impact on in-vehicle UFP concentrations, highest in high traffic flow-high speed conditions or in high traffic density-low speed conditions. Vehicle cabin barrier effects are the primary determinant of in-vehicle exposure concentrations, providing 15% protection with the windows down, 47% protection with the windows up and the vent open, and 83–90% protection with the windows up and the vent closed (more with the air conditioning on). Unique results from this study include the dominance of ventilation over traffic effects on UFP and the non-linear relationships between traffic variables and UFP concentrations. The results of this research have important implications for exposure modeling and potential exposure mitigation strategies.

1. Introduction

Motor vehicle emissions are a known contributor to negative health outcomes for people with long-term exposures, especially to fine particulate matter (Health Effects Institute, 2010). Traffic congestion, in particular, has been cited as a cause of human health problems (Levy et al., 2010). These concerns raise interest in strategies to mitigate vehicle emissions and human exposure. But the full effects of congestion on motor vehicle emissions and air quality are still not well quantified (Dowling, 2005; Bigazzi, 2011). There is even less research regarding the impacts of congestion on human exposure to traffic-related pollution.

One traffic-related pollutant that has received considerable attention of late is ultrafine particulates (UFP) – particulate matter with diameter <0.1 μm. UFP are a main component, in terms of particle number, of motor vehicle emissions – which are the major source of UFP in urban settings (Morawska et al., 2008). Because of proximity to vehicular emissions sources, UFP concentrations are higher around roadways than in ambient conditions (Shi et al., 1999; Zhu et al., 2002, 2009). UFP exposure concentrations are generally elevated during transportation activities (Kaur et al., 2007; Knibbs et al., 2011), and in-vehicle exposure can be a significant portion of total daily exposure to UFP (Zhu et al., 2007; Fruin et al., 2008).

The negative health impacts of UFP have been shown through toxicology studies (Li et al., 2003; Vinzents et al., 2005; Moller et al., 2008; Grahame and Schlesinger, 2010), but the epidemiological evidence is still scant, due to limited monitoring sites and few long-

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term studies (Knibbs et al., 2011). UFP pose particular health risks because their small size allows for deep deposition in the lungs and passage into the circulatory system. Short-term exposure to traffic-related particulate pollution, such as would be experienced while commuting in traffic, has been shown to have a variety of negative health effects (Peters et al., 2004; Mills et al., 2005; Tornqvist et al., 2007; Knibbs et al., 2011).

The potential health impacts of exposure to UFP during travel suggests a need for mitigation techniques within the transportation microenvironment. In-vehicle UFP exposure is affected by variations in vehicle emissions (which depend on the vehicle fleet, fuels, traffic patterns, and meteorology, among other factors) and variations in pollutant transport from source to receptor (including the effects of dilution, creation, and removal). Emissions of UFP are strongly associated with heavy-duty diesel vehicle activity, though light-duty gasoline vehicles can also generate high UFP emissions under high engine loads. After emission of UFP or its precursors, pollutant transport, secondary formation, and removal depend largely on meteorological and built environment factors. Many aspects of the generation and behavior of traffic-related UFP in urban environments and transportation microenvironments can be found in reviews elsewhere (Morawska et al., 2008; Knibbs et al., 2011).

Of particular interest in this study are the traffic-related factors that impact in-vehicle UFP exposure. Here we consider the three fundamental traffic parameters of traffic flow (vehicles passing per unit time), traffic speed (distance traveled per unit time), and traffic density (vehicles per unit length of roadway) (May, 1989). Note that here we use the traffic flow theory definition of "traffic density" (other papers have used "density" to indicate what we here describe as "traffic flow").

Other research has shown on-road and roadside UFP concentrations to be positively correlated with traffic flow, though more strongly associated with heavy-duty vehicle flow than light-duty vehicle flow or total vehicle flows (Junker et al., 2000; Fruin et al., 2008; Wang et al., 2008; Knibbs et al., 2009; Aggarwal et al., 2011). Knibbs et al. (2011) points out that often these correlations are based on hourly or daily flows (more often described as traffic "volumes"), but short-term traffic patterns are likely to be important, too. Also, some studies assess UFP correlations with traffic flow using cross-locational comparisons that make traffic effects hard to distinguish from other environmental characteristics. Traffic speed has been shown as both positively and negatively correlated with on-road UFP concentrations (Kittelson et al., 2004; Knibbs et al., 2009; Aggarwal et al., 2011).

Beyond roadway concentrations, in-vehicle exposure is also affected by the vehicle shell. The vehicle shell acts as a barrier, leading to lower UFP concentrations in better-sealed vehicle cabins (Zhu et al., 2007; Knibbs et al., 2010; Xu et al., 2010, 2011). Recirculation of cabin air through a ventilation system increases removal of UFP through surface deposition (though the effect of filtration is small) (Hudda et al., 2011). The penetration of outside UFP into the cabin is expected to increase with vehicle speed because of increased pressure differentials (Xu and Zhu, 2009; Hudda et al., 2011).

Despite many recent advances, the full effect of traffic conditions on UFP exposure for motorists is still far from clear. While traffic speed and flow are both correlated with on-road UFP concentrations, traffic conditions will also affect cabin penetration of roadway UFP and inter-vehicle spacing (proximity to UFP sources). Additionally, traffic variables have strong linear and non-linear inter-relationships (May, 1989) that can be expected to have varying net effects on in-vehicle UFP concentrations.

Little research to date has measured in-vehicle UFP concentrations in varying traffic states under different vehicle ventilation conditions; even fewer studies have combined short-term traffic characteristics with UFP measurements at all. The research presented in this paper illuminates the effects of freeway traffic conditions on in-vehicle UFP exposure. We combine in-vehicle and outside-vehicle UFP measurements with simultaneous traffic data gathered at various states of traffic congestion and with varying vehicle ventilation conditions. The results help identify potential exposure mitigation strategies and gaps in our understanding of in-vehicle UFP exposure. We next describe the experimental methods, followed by results, conclusions, and a discussion of future work.

2. Methodology

This study tests the impacts of various factors on in-vehicle UFP concentrations using statistical analyses of real-world measurements made in a probe vehicle traveling in freeway traffic. The two parts of this methodology section describe the materials used in data collection and the experimental method.

2.1. Data collection equipment

UFP concentrations were measured using two P-Trak ultrafine particle counters (TSI Model 8525). P-Trak instruments are commonly used in personal exposure studies of UFP for transportation modes because of portability (Kaur et al., 2007). Number concentrations at 1 Hz are obtained for particles in the size range 0.02–1 μm, dominated by the ultrafine size range, with a maximum concentration level of 500,000 particles per cubic centimeter (pt cc^{-1}). The P-Trak instruments were factory calibrated by TSI in October2009.[1] The instruments were allowed a "warm-up" period of 10 min before data collection to avoid possible underestimation bias (Wallace et al., 2011). A recent study of UFP monitors showed median precision within 10% for the P-Trak instruments (Wallace et al., 2011). When run side-by-side, the two P-Trak instruments used in this study showed good agreement, with a correlation coefficient of 0.997 over a 5-min interval. The P-Trak particle size range excludes many nucleation mode particles, and so will underrepresent total particle number count, especially near combustion sources such as vehicles (Zhu et al., 2006). Possible implications of this are later discussed.

Three different probe vehicles were used in this study, all gasoline-fueled passenger sedans: a 1999 Pontiac Grand Prix, a 2007 Honda Civic (gas-electric hybrid), and a 2010 Toyota Prius (gas-electric hybrid). The probe vehicles were equipped with a forward-facing digital video camera in the passenger-side front seat recording images through the front windshield. Two Garmin iQue® 3600 GPS (Global Positioning System) receivers were used to collect probe vehicle location and speed data at 1 Hz. A receiver was placed in each of the front and rear windshields. The two GPS data sources were compared and showed good agreement, with a correlation coefficient of 0.998. The final probe vehicle speed and location data were averaged between the two receivers.

Traffic data were obtained from the Portland Oregon Regional Transportation Archive Listing (PORTAL – at www.portal.its.pdx.edu), an archive of transportation data from the Portland metropolitan region. The traffic data were collected by inductive dual-loop vehicle detectors with an average spacing of 1.2 km on the study corridor. Vehicle counts and time-mean speed at 20-s intervals were obtained from PORTAL for all study days. The traffic data were matched to the probe vehicle's temporal and spatial position using the in-vehicle GPS data. Suspect traffic data as identified by

[1] This was within a year of data collection, and so within the recommended re-calibration timeframe.

Table 1
Data collection summary.

	Thurs. June 10, 2010	Tues. August 31, 2010	Thurs. Sept.2, 2010	Tues. Sept.7, 2010	Tues. Oct.12, 2010	Sun. Oct. 17, 2010
Hours	15:00–18:32	14:48–18:02	14:42–17:50	14:27–18:18	15:50–19:18	17:45–20:00
# of Trips	7 SB, 7 NB	7 SB, 7 NB	8 SB, 8 NB	8 SB, 8 NB	9 SB, 9 NB	8 SB, 8 NB
Probe Vehicle	1999 Pontiac Grand Prix	2010 Toyota Prius Hybrid	2010 Toyota Prius Hybrid	2007 Honda Civic Hybrid	2007 Honda Civic Hybrid	2007 Honda Civic Hybrid
Traffic Flow (veh day^{-1})	103,259	99,456	103,905	97,678	97,186	72,205
Temperature[a] (°C)	12	16	27	17	18	12
Wind Speed[a] (km hr^{-1})	1.0	2.3	11.7	1.1	0.8	1.9
Relative Humidity[a] (%)	97	93	37	80	42	57
Hourly Precip[a] (cm)	0.05	0.03	0.00	0.15	0.00	0.00
$PM_{2.5}$[b] (μg m^{-3})	2.6	2.8	3.0	3.6	5.6	7.2
UFP[a] (pt cc^{-1})	25,990	21,547	17,286	21,483	31,145	31,774

[a] Averaged over data collection period.
[b] Averaged over entire day.

PORTAL's data quality flags[2] and validity checks were removed before analysis. After spatial-temporal matching, the probe-based and PORTAL-based speeds had a correlation coefficient of 0.90.

Meteorological data (temperature, pressure, humidity, rainfall, and wind speed) were collected from a permanent weather station 4.8 km east of the study corridor. For reference, daily fine particulate air quality data were obtained from a permanent air quality monitoring station 1.6 km west of the study corridor (24-h average $PM_{2.5}$). Road grade and geometry were obtained from the Oregon Department of Transportation.

2.2. Experimental method

We collected concurrent traffic and air quality data on six non-contiguous days during the summer and fall of 2010. Probe vehicles were driven on a 10 km stretch of OR-217, a freeway in the Portland, Oregon metropolitan area. On each day of data collection, a single probe vehicle was driven continuously on the freeway for a period of approximately 3 h. In total, 94 trips were executed, where a "trip" consists of the probe vehicle traveling the 10 km corridor in a single direction (15.4 h of data in total). The probe vehicle trips were executed in loops, alternating southbound (SB) and northbound (NB) travel directions. Five of the data collection days were on weekdays (Tuesdays and Thursdays), and one was on a Sunday (to capture lighter traffic conditions). On the weekdays, the data collection periods covered varying time spans before, during, and after the afternoon traffic peak period.

The probe vehicles were driven each day by the same driver, using a median-speed driving approach (approximately equal vehicle passing and overtaking) with free choice of lanes. When queues formed on the roadway, the driver attempted a spacing of 2 m from the leading vehicle. A second passenger rode in the back seat of the vehicle, monitoring the data collection equipment.

The P-Trak instruments were positioned on the back seat of the probe vehicle with inlet tubes connected to the front seat driver-side and passenger-side headrests (to approximate the breathing position of vehicle occupants). For outside-vehicle UFP levels, an inlet tube was also fed outside of the sealed passenger-side front window. Outside-vehicle concentrations were collected on the last three study days only. Because only two UFP monitors were available, when outside-vehicle concentrations were collected, the inside-vehicle P-Trak instrument measured passenger-side concentrations only.

The main experimental factor was vehicle ventilation condition. Trips were executed varyingly with the windows up or down, the air vents open or closed (recirculating cabin air), and the air conditioning (A/C) on or off. The A/C "on" was only tested with windows up and vents closed. The "windows down" condition was conducted with three of the four windows open. The fan in the vehicle's ventilation system was set to medium. We sought a wide range of traffic conditions and allowed other factors of secondary interest to vary by date (meteorology, background concentrations, starting time, and probe vehicle).

The six data collection days are summarized in Table 1, with average UFP values shown for in-vehicle passenger-side measurements with the windows down.

The study corridor, OR-217, is a freeway located about 8 km west of the Portland, Oregon central business district. The speed limit is 55 miles hour^{-1} (89 kph) and the freeway has 2–3 lanes in each of the NB and SB directions. This freeway had average annual daily traffic of approximately 100,000 in 2010, with weekday (non-holiday) two-way daily traffic flows ranging from 95,000 to 107,000 vehicles per day during the months when data were collected. Weekend two-way daily traffic flows ranged from 59,000 to 92,000 vehicles per day during these months. The road grades on the corridor range from 0.2% to 6.2% (positive or negative depending on the direction of travel). These grades were calculated as the average slope between crest and sag vertical curves, with average spacing of 0.7 km.

From the measured traffic speeds, v, in kph and traffic flow, q, in vehicles per hour per lane (veh hr^{-1} ln^{-1}), traffic density, k, in vehicles per lane-kilometer (veh (ln-km)$^{-1}$) is calculated as $k = q/v$ (May, 1989). Freeway Level of Service (LOS) is calculated based on traffic density thresholds from the Highway Capacity Manual (Transportation Research Board, 2000). LOS is a widely-used indicator of traffic congestion level, ranging from free-flow conditions (LOS A) to heavy congestion (LOS F). Table 2 shows the number of 20-s aggregated observations broken down by LOS and ventilation conditions.

The joined data from the sources described above were validated using reasonableness checks. Most of the analysis was carried out at 20 s aggregation, matching the resolution of the traffic data. At this aggregation, around 2800 data points were available for analysis (depending on the variables of interest, because of missing

Table 2
Number of 20-s observations by freeway LOS and probe vehicle ventilation condition.

Ventilation conditions	Level of Service (LOS)						Total
	A	B	C	D	E	F	
Windows down	2	50	160	333	222	566	1333
Windows up, Vent open, A/C off	23	81	120	158	130	193	705
Windows up, Vent closed, A/C off	14	59	116	115	47	110	461
Windows up, Vent closed, A/C on	1	2	23	69	46	153	294
Total	40	192	419	675	445	1022	2793

[2] See http://portal2.its.pdx.edu/dataquality/

data). The next section presents the results of the data analysis and a discussion of findings.

3. Results

This section first presents an overview of the UFP data, then discusses the relationships between study variables and the measured UFP concentrations inside and outside of the probe vehicle. At 20-s aggregation (means), the range of observed UFP concentrations inside the vehicle is wide: from 993 pt cc^{-1} to 435,250 pt cc^{-1}. The passenger-side and driver-side UFP concentrations show good agreement when measured concurrently, with a correlation coefficient of 0.996. The in-vehicle and outside-vehicle UFP concentrations are less correlated, as expected, with a correlation coefficient of 0.575. The mean and median passenger-side in-vehicle concentrations are 25,871 pt cc^{-1} and 17,628 pt cc^{-1}, respectively, with the windows down, and 11,176 pt cc^{-1} and 8661 pt cc^{-1}, respectively, with the windows up.

3.1. Extreme-concentration episodes

There were five observed extreme-concentration episodes with sustained concentrations over 100,000 pt cc^{-1} for duration of more than 1 min (and even reaching the detection limit of 500,000 pt cc^{-1} in the second-by-second data). By consulting the video data, an analysis of these periods reveals an individual suspected high-emitting vehicle closely ahead of the probe vehicle during each of these episodes. Suspected high-emitting vehicles were subjectively identified as those with visible emissions (smoke) from the tailpipe, those whose presence correlated with observed foul odors during data collection, and any other heavy-duty vehicles. Three of the suspected high-emitting vehicles are heavy trucks, one is a large passenger pickup truck, and one is a sedan.

The temporal and spatial correlation of the presence of one of these vehicles with high exposure concentrations makes their emissions a plausible explanation for the extreme-concentration episodes. A similar effect of large particulate exposures being attributable to leading diesel and heavy-duty vehicles was found in previous research (Fruin et al., 2004). Measurement of the contribution of individual vehicles to total roadway UFP concentrations is left to future research efforts. In order to look at more generalized traffic relationships with UFP concentrations, time periods with these suspected high-emitting vehicles present are excluded from the initial analysis but later included in the regression analysis in Section 3.5. The 5 episodes were each 2–7 min in length, resulting in 80 time periods at 20-s aggregation (2.85% of the total) identified as having suspected high emitting vehicles.

3.2. Traffic congestion and UFP concentrations

Initial comparison of measured UFP concentrations to the traffic variables reveals no clear relationship. Neither in-vehicle nor outside-vehicle UFP concentrations correlate well with traffic flow, density, or speed (as measured by PORTAL or the probe vehicle): all have correlation coefficients between −0.07 and 0.07.

Fig. 1 shows boxplots of outside-vehicle UFP concentrations segmented by traffic LOS, with suspected high-emitting vehicle episodes excluded. The boxplots show the range, upper/lower quartiles, and median observed values, with statistical outliers as circles. Fig. 1 also includes the number of 20-s aggregation intervals included in the plot for each LOS (as "N") – note that outside-vehicle concentration data were not collected during all time periods.

As can be seen in Fig. 1, outside-vehicle concentrations do not notably trend up or down with LOS. Using a non-parametric

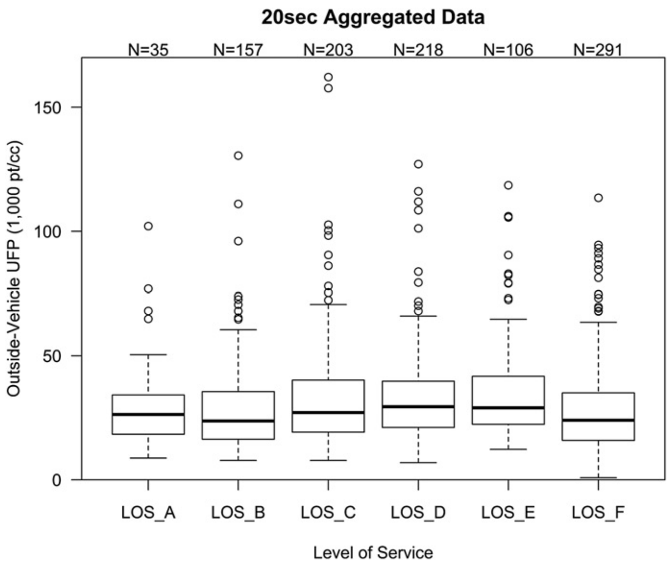

Fig. 1. Comparisons of traffic LOS and outside-vehicle UFP concentrations (suspected high emitting vehicle episodes excluded).

Wilcoxon signed-rank test to compare each LOS in Fig. 1 with its neighbors, only the LOS E versus LOS F comparison is statistically significantly different at $p = 0.01$. Observe that here the difference is *lower* concentrations at the heavier congestion level – and that the difference in medians is small compared to the range of concentrations observed. Thus, on-road UFP concentrations are not correlated with traffic LOS. The traffic-UFP relationship is explored in more detail using regression analysis below in Section 3.5.

3.3. Vehicle ventilation and UFP concentrations

We next examine the effects of varying vehicle ventilation conditions. Fig. 2 illustrates the observed effects of ventilation conditions on in-vehicle UFP concentrations. In Fig. 2, data from 4 sample trips with varying ventilation are shown: in-vehicle UFP, outside-vehicle UFP, and probe vehicle speed (as the shading of the circles, with darker shading being faster) at 20 s aggregations. On the top left, the trip with the most air exchange (windows down) had the most agreement between in-vehicle and outside-vehicle concentrations. On the top right, we see that rolling up the windows (but leaving the vent open) reduced the in-vehicle concentration compared to the outside-vehicle concentrations, but that the two still generally moved together. The bottom two panels in Fig. 2 show that with the windows up and the vent closed, in-vehicle UFP concentrations are nearly unresponsive to outside-vehicle concentrations. Furthermore, when the A/C is "on" the in-vehicle UFP concentrations are slightly lower.

We next combine the traffic and UFP data with ventilation conditions. Fig. 3 shows natural log-transformed in-vehicle UFP concentrations versus probe vehicle speed, segmented by ventilation condition, at 20-s aggregations (again excluding suspected high-emitting vehicle episodes). The windows-up condition has slightly lower in-vehicle concentrations than windows-down, which are further lowered when the vents are closed and the A/C is on. These effects are consistent across the range of observed speeds, and the in-vehicle concentrations do not trend notably with speed.

The vehicle ventilation condition affects the UFP concentration variability, in addition to the mean values. Aggregating to longer intervals, Fig. 4 shows boxplots of UFP peaking at 1-min

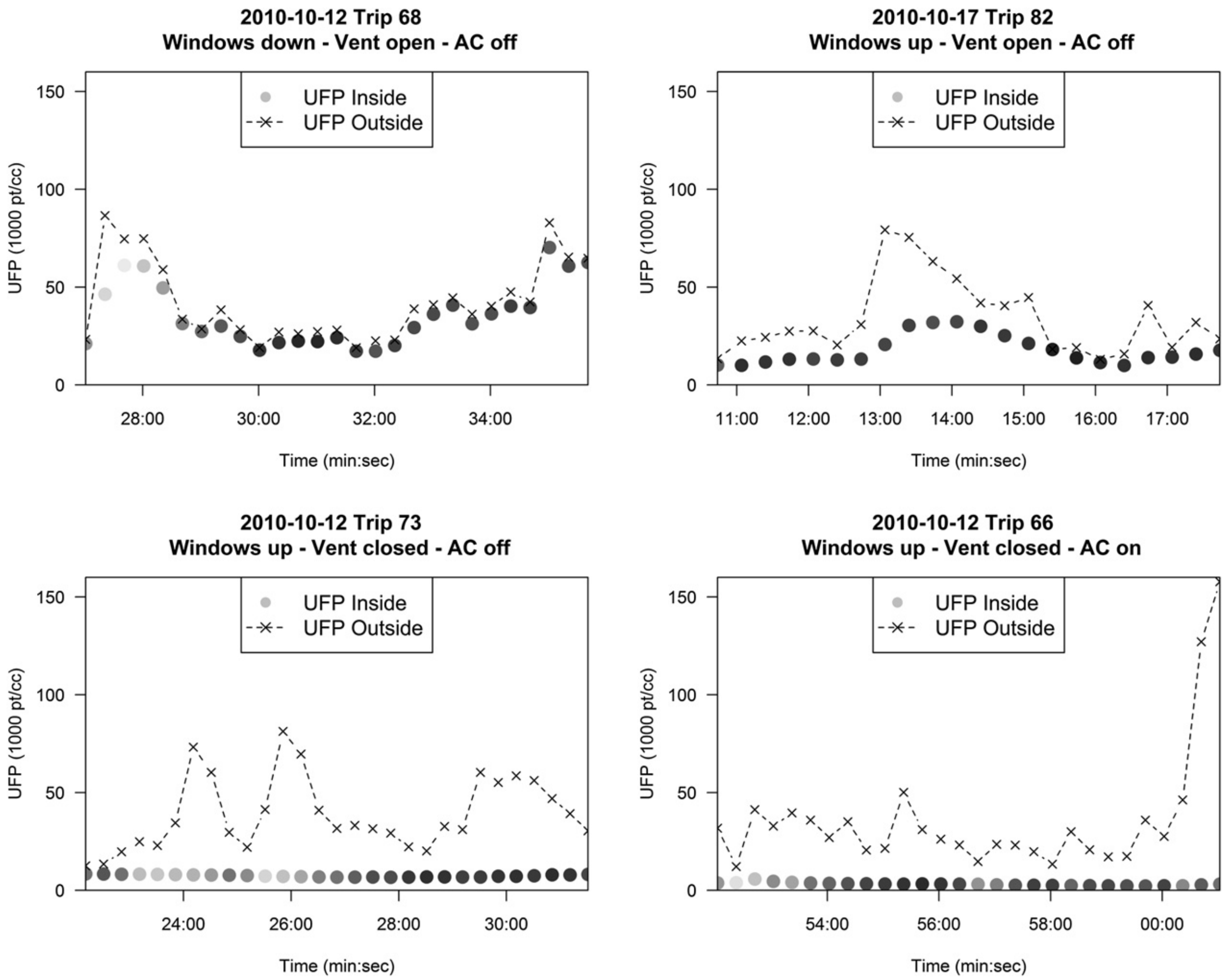

Fig. 2. UFP concentrations from sample trips for different ventilation conditions.

aggregations (calculated as the 90th percentile concentration divided by the mean concentration for the time interval). The figure is grouped with the first three boxplots showing in-vehicle UFP peaking for different vehicle ventilation conditions and the fourth boxplot showing outside-vehicle UFP peaking. The outside-vehicle UFP peaking is the highest, and similar to the in-vehicle UFP peaking with the windows down. The in-vehicle UFP peaking with the windows up is much lower, and lower still when the vents are closed. Again using a non-parametric Wilcoxon signed-rank test to compare the peaking distributions, all conditions are statistically significantly different at $p = 0.01$. Fig. 4 shows that rolling up the windows and closing the vents has a damping effect on the UFP concentrations, in addition to the mean-reducing effect shown in Fig. 3. This damping effect with the windows up is consistent with previous research on cabin penetration of UFP (Zhu et al., 2007).

3.4. In/out-vehicle concentration comparisons

We next compare the in-vehicle to outside-vehicle UFP concentrations for different ventilation conditions. As stated in Section 2.2, outside-vehicle concentrations were measured on the last three study days only (all using with the 2007 Honda Civic Hybrid). Regressing untransformed in-vehicle UFP concentrations on outside-vehicle concentrations, segmented by ventilation type and constrained to the origin, produces slope coefficients of 0.851, 0.531, 0.172, and 0.103 for Windows down, Windows up-Vent open, Windows up-Vent open-A/C off, and Windows up-Vent open-A/C on conditions, respectively (all significant at $p = 0.01$). This indicates that in-vehicle concentrations increase at about 85% of the increase in outside-vehicle concentrations with the windows down. With the windows up, in-vehicle concentrations increase at 53% of the increase in outside-vehicle concentrations with the vents open and at 10–17% with the vents closed, depending on A/C conditions.[3]

These results agree with previous empirical research on in/out concentration ratios (Zhu et al., 2007; Xu and Zhu, 2009; Knibbs et al., 2010; Hudda et al., 2011) – though the reduction can vary greatly with a number of factors, especially the vehicle. Hudda et al. (2011) found in/out ratios from around 0.4 to 0.8 with the windows up, the fan on, and the vent open – and from less than 0.1 to around 0.3 with the vent closed. Their results using only a 2009 Honda Civic agree even more closely with our results, with Vent open and Vent closed in/out ratios of about 0.6 and 0.1, respectively. Zhu et al. (2007) found a similarly wide range of around 0.3–0.7 for Windows up-Vent open conditions, and from less than 0.1 to almost 0.6 for

[3] Note that the effect of the vehicle shell observed here could be underestimated if the P-Traks are disproportionately under-reporting more recently emitted UFP (Zhu et al., 2006). However, much of the unobserved nucleation mode particles would be the result of secondary formation from volatile gas emissions (Morawska et al., 2008), so we do not know that the unobserved particles would be lag-biased in this way.

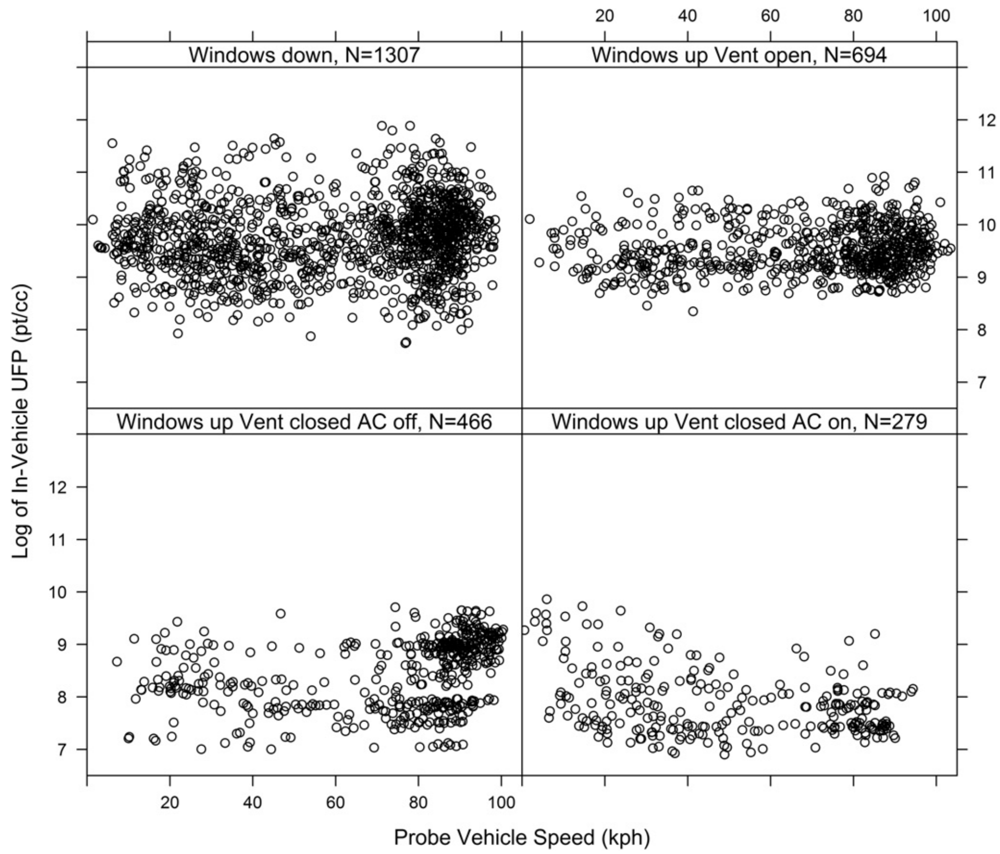

Fig. 3. UFP concentrations versus speed, by ventilation conditions.

Windows up-Vent closed conditions. We found no other studies with which to compare our results for the Windows-down condition.

We also perform an analysis of the lagged correlations between in-vehicle and out-vehicle UFP concentrations using the second-by-second UFP data. Averaging the results by ventilation type for each probe vehicle run, the maximum correlations are found at lags of 1.0, 6.5, and 134.8 for Windows down, Windows up-Vent open, and Windows up-Vent closed conditions, respectively. Thus, in-vehicle concentrations follow outside-vehicle concentrations most closely at 1 s lags with the windows down, about 6 s lags with the windows up and the vent open, but over 2 min lags with the windows up and the vent closed. These lags are indicative of the much lower air exchange rates with the vent closed than open (with the latter being more similar to the windows down condition). These lags show a somewhat wider range than was observed by Zhu et al. (2007), who measured 30–60 s lags for in-vehicle UFP concentrations following outside-vehicle concentrations. However, a modeling study by Xu and Zhu (2009) found time delays of up to 200 s for vehicle cabins with low air exchange rates.

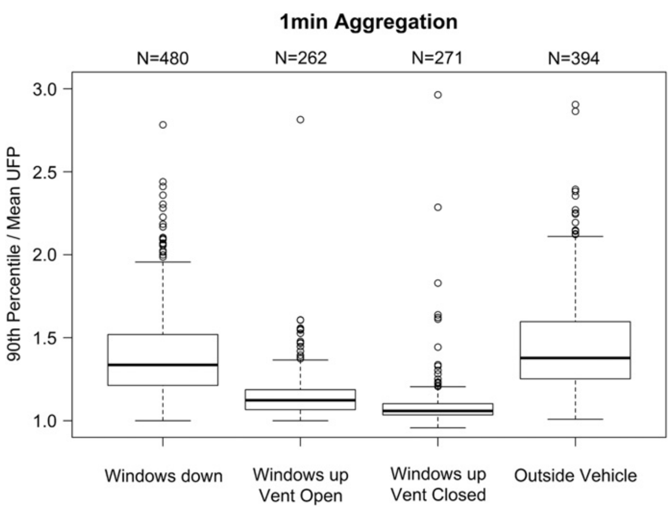

Fig. 4. UFP concentration peaking and ventilation conditions.

3.5. Regression analysis

As a final step we perform a regression analysis with the in-vehicle passenger-side UFP concentrations as the dependent variable. The UFP concentrations are natural log-transformed because of strong positive skew, which is consistent with previous research on UFP (Fruin et al., 2008; Boogaard et al., 2009; Aggarwal et al., 2011). The independent variables tested include the probe vehicle (dummy), relative humidity (%), temperature (°C), and wind speed (kph) at the weather station, road grade (%), ventilation conditions (4-factor dummy: windows down, windows up-vent open, windows up-vent closed-A/C off, and windows up-vent closed-A/C on), and traffic variables (traffic flow in vehicles per hour, traffic density in vehicles per lane-km, and probe vehicle speed in kph). The traffic variables are tested as linear and squared terms, including first-order lags of each. The 20-s log-transformed UFP measurements show strong autocorrelation with a Durbin–Watson test statistic of 0.27, significant at $p = 0.01$. To adjust for autocorrelation, regression is performed using maximum likelihood estimation of a generalized least squares (GLS) model that includes first-order

Table 3
Segmented regression analysis.

	Windows down		Windows up, vent open		Windows up, vent closed	
	Estimate	LR	Estimate	LR	Estimate[b]	LR
Speed (kph)	0.0023	6.87	0.0012	4.14	−0.0080	27.18
Speed^2					0.000056	
Flow (1000 veh hr^{-1})					0.0380	11.65
Density (veh/ln-km),					−0.0085	8.91
Density^2					0.000087	
Grade (%)			0.0106	6.99		
Vehicle dummy	[a]	9.89	[a]	7.82		
Temperature (°C)	−0.0552	4.54			−0.1085	15.87
Humidity (%)					−0.0184	11.20
Wind speed (kph)	0.0574	4.59				
N	1327		703		698	
Autocorrelation coef.	0.860		0.922		0.956	
D–W statistic	1.88		1.61		1.54	
Residual std. error	0.748		0.474		0.572	
Mean UFP (pt/cc)	17,829		13,331		3976	
Ln(UFP)	9.8		9.5		8.3	

[a] Vehicle dummy effects ranged up to 0.558.
[b] All traffic variables are lagged in the third model.

serial correlation (within probe vehicle runs). The regressions are based on all of the available data (i.e. they *do not* exclude the suspected high-emitting vehicle episodes) – a conservative approach because these episodes will exacerbate the model error.

3.5.1. Pooled regression

First, as a test of the contribution of each independent variable, we perform a pooled regression using one traffic variable at a time. Based on the results of a Breusch–Pagan test, we also adjust for heteroscedasticity by using a weighting matrix in the GLS estimation that segments standard errors by Window condition (up/down). From these three estimated models (one for each traffic variable) we perform likelihood ratio tests for the inclusion of each group of independent variables.

Based on likelihood ratios, the ventilation dummy is by far the largest factor, with likelihood ratios of 127–129 (with 3 degrees of freedom, significant at $p = 0.01$). The three traffic variable groups (each of which includes squared and lagged terms for 4 degrees of freedom) are the next most significant, although much less so, with likelihood ratios of 17, 11, and 6 for Speed, Flow, and Density, respectively (only speed is significant at $p = 0.01$). The Vehicle dummy variable has likelihood ratios of about 8 ($p = 0.02$ with 2 degrees of freedom), the weather variables collectively have likelihood ratios of about 4 ($p = 0.21$ to 0.28 with 3 degrees of freedom), and road grade has likelihood ratios of 4–6 ($p = 0.01$ to 0.04 with 1 degree of freedom).

The estimated variance structure indicates strong autocorrelation, with autocorrelation coefficient estimates of 0.92 for all three models. Using this coefficient in the GLS estimation adjusts for most of the autocorrelation, resulting in a new Durbin–Watson test statistic of 1.74 (greatly improved, though still significant at $p = 0.01$ based on a Ljung–Box test). Heteroscedasticity is also indicated with estimated standard error strata of 0.47 for the Windows-up condition in all three models (compared to the base case of 1.0 for the Windows-down condition). The Vehicle dummy variable is uniform for each day of data collection, so there is strong correlation between the Vehicle and weather variables (see Table 1). This correlation makes the effects of each difficult to distinguish within a linear model.

3.5.2. Segmented regression

The pooled regression shows strong autocorrelation, heteroscedasticity by Window condition, and the dominance of cabin ventilation factors on in-vehicle UFP concentrations. To continue the investigation we perform segmented GLS regression by ventilation conditions, again incorporating first-order autocorrelation. We employ a stepwise modeling approach, beginning with the full model and removing variables one at a time based on the lowest likelihood ratios for inclusion. The final models accept all variables at $p < 0.05$. Again traffic variables are tested as linear and squared terms for both zero-lag and first-lag variables.

The estimated model results are shown in Table 3, along with Likelihood Ratios (LR) for the independent variables and other model attributes. Of the traffic variables, only probe vehicle speed is significant in the Windows down and Windows up-Vent open models, whereas all three traffic variables are significant in the Windows up-Vent closed model. Note also that the significant traffic variables are all first-lags in the third model. In the first two models, speed has a significant positive influence on UFP concentrations, with about half as large of an effect when the windows are up. In both the Windows down and Windows up-Vent open models traffic density was a significant (positive) variable at $p = 0.10$, but did not meet the $p < 0.05$ criterion.

The largest estimated autocorrelation coefficients are in the Windows up models, with Vent closed more auto-correlated than Vent open (as expected). The Durbin–Watson (D–W) test statistics included in Table 3 show large improvement[4] from those generated without an autocorrelation adjustment (0.38, 0.21, and 0.17 for the three models respectively). Based on a Ljung–Box test there is still significant autocorrelation in the two Windows up models at $p < 0.01$, likely due to higher-order autocorrelation (particularly associated with the extreme-concentration episodes).[5] The residual standard errors for the models indicate that, as expected, the error variance is higher in the Windows down conditions. Also note that the mean concentrations are much lower in the third model.

The third model in Table 3 has significant squared terms for speed and density. The signs on the speed and density parameters produce "U"-shaped curves, with minimal UFP effects at 49 veh (ln-km)$^{-1}$ density and 71 kph speed. This density value corresponds to vehicle headways of about 20 m. The 71 kph speed corresponds approximately to the point at which traffic flow breakdown occurs (see Fig. 5 below). Thus, instead of continually increasing UFP with Speed and Density (as suggested in the first two models), very low-speed or low-density conditions also have higher UFP concentrations than more moderate traffic conditions in the third model.

As mentioned above, there is correlation among the Vehicle dummy and the meteorological variables (Temperature, Humidity, and Wind speed), so the effects of each are difficult to distinguish with the models. There are also relationships among the traffic variables that can lead to competition in the regression model. Fig. 5 shows the fundamental traffic variables in three bivariate plots (with dashed lines at the minimum-effect Speed and Density from the third model). Traffic speed and density have strong negative correlation (correlation coefficients of −0.79 using the probe vehicle speed and −0.92 using the traffic speed). The 49 veh (ln-km)$^{-1}$ density is near a break in the linearity of the speed-density relationship in Fig. 5. Using a congestion threshold of 73 kph, flow is correlated with density in uncongested conditions with a coefficient of 0.84 (but only −0.08 in congested conditions). Flow is correlated with traffic speed with coefficients of −0.38 and 0.25 in uncongested and congested conditions, respectively. These results are consistent with traffic flow theory (May, 1989).

[4] D–W statistics range from 0 to 4, with 2 indicating no autocorrelation, 0 indicating perfect positive autocorrelation and 4 perfect negative autocorrelation.
[5] Removing these episodes increases the third model's D–W statistic to an insignificant 1.88.

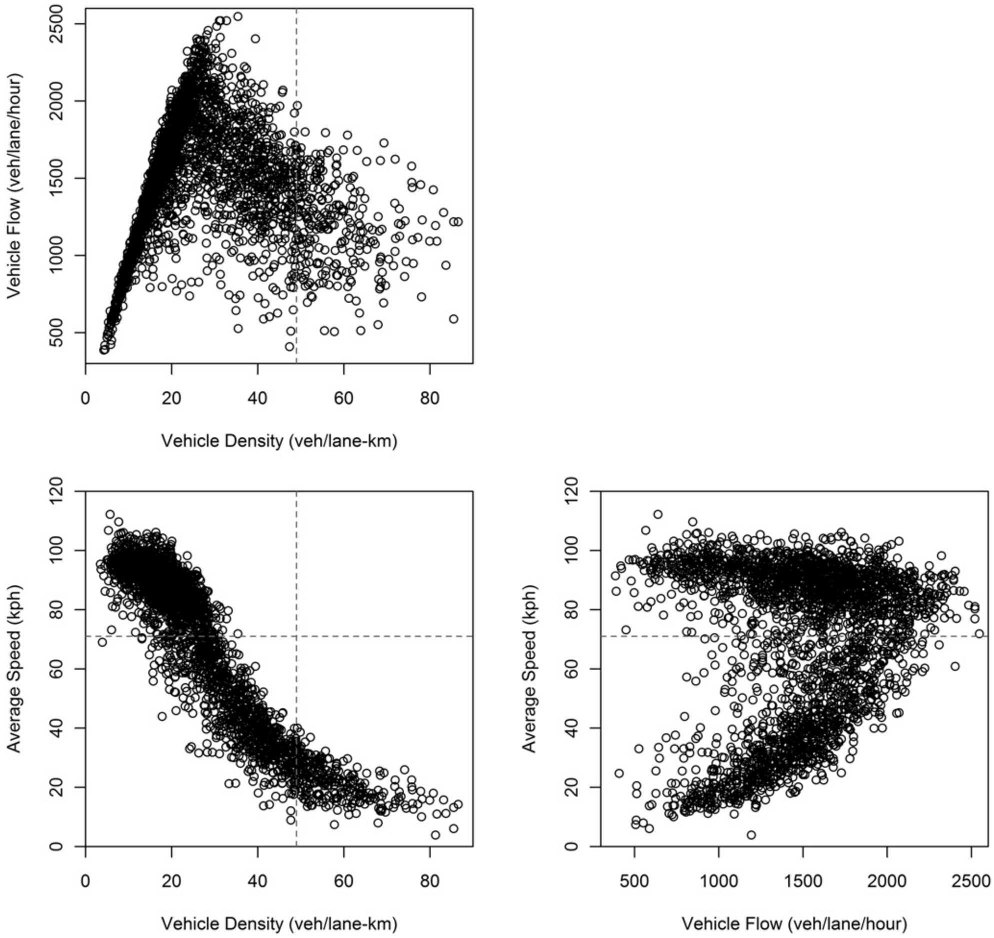

Fig. 5. Traffic variable fundamental diagrams.

Because of the traffic variable relationships demonstrated in Fig. 5, the net effects of traffic on UFP are smaller than indicated by the individual parameter estimates in the third model of Table 3. In uncongested conditions we see offsetting effects of increasing Flow and Density simultaneously. The highest Flows (and greatest Flow effect on UFP) are found near the minimum Speed effect at 71 kph. Moderately congested traffic states slower than 71 kph and below 49 veh (ln-km)$^{-1}$ density have off-setting UFP effects from the negatively-correlated Speed and Density. In heavy congestion above 49 veh (ln-km)$^{-1}$ density, the UFP-increasing effects of lower Speed and higher Density are partially offset by lower Flows.

Using $q = k^*v$ and an assumed relationship of $k = 54 - 0.43^*v$ (fitted from Fig. 5) with 3 lanes of traffic, the range of likely net traffic effects on UFP from the estimated parameters of the third model in Table 3 is -0.26 to -0.18. The effect is less negative (thus higher UFP) in high-speed, high-flow (but low density) conditions and low-speed, low-flow (but high density) conditions. This range of net traffic effects corresponds to an 8% change in UFP. These effects would be lower on a 2-lane freeway. The range of net traffic effects from the observed traffic data (again using the parameters of the third model in Table 3) is -0.31 to -0.21 (the 10th–90th percentiles) – a slightly larger 10% change in UFP.

This regression analysis combines several days of data using three different vehicles. We expect better (or more poorly) sealed cabins to provide more (or less) protection, and so these results can roughly be expanded by interpolation within the range of ventilation conditions. Segmenting the models by vehicle does not appreciably alter these results; although it reduces the sample size such that some of the traffic variables are no longer significant for the vehicles with fewer data (the Civic results are essentially unchanged).

4. Conclusions

Previous studies have shown significant relationships between on-road or roadside UFP concentrations and motor vehicle traffic. This study looks at traffic characteristics in more detail and shows that on a freeway short-term traffic states have only a small influence on in-vehicle UFP concentrations. Vehicle barrier effects are the primary determinant of in-vehicle UFP exposure concentrations, reducing both mean concentrations and peaking. In this study the vehicle cabin provides on average 15% protection with the windows down, 47% protection with the windows up and the vent open, and 83–90% protection with the windows up and the vent closed (more with the air conditioning on). The in-vehicle concentrations have more autocorrelation and less variance with the windows up and the vent closed than with the windows down or the vent open, and up to 2-min lagged effects when compared to outside concentrations.

Regression analysis reveals non-linear relationships between traffic variables and UFP, consistent with non-linear relationships among the traffic variables. Due to negative correlation between traffic speed and density, in a well-sealed vehicle cabin UFP concentrations are highest in high-speed, high-flow conditions (before traffic flow breakdown occurs) or high-density conditions (with low speeds and close vehicle spacing). A comparison of

Windows down and Windows up-Vent open conditions shows surprising similarity.

Although it could not be directly measured, qualitative analysis suggests that individual vehicles in the on-road fleet are another major factor influencing variations in UFP exposure concentrations. This has several implications. The first is that on-road air pollution exposure modeling can only estimate highly aggregate exposure levels unless individual vehicles modeled. Second, in support of the findings related to traffic density, inter-vehicle spacing is an important consideration for exposure concentrations of short-lived air pollutants such as UFP.

Our findings suggest that the most likely mitigation strategies for reducing on-freeway UFP exposure will be effective cabin shielding and targeting high-emitting vehicles. These are in contrast to general traffic congestion mitigation as an air quality improvement strategy. Congestion *per se* does not cause higher in-vehicle UFP exposure concentrations; the net effect of traffic is a complex combination of influences through pollutant emissions, dispersion, and vehicle penetration. In addition, it is suggested that future research efforts to model on-road exposure include detailed data on, and accurate representation of, vehicle fleet heterogeneity and inter-vehicle spacing and mixing.

To the best of our knowledge, this is the first study that combines in-vehicle UFP exposure measurements under varying vehicle ventilation conditions with simultaneous detailed traffic data. Because vehicle penetration is not independent of speed, these factors (traffic and ventilation conditions) need to be considered in concert. Some results unique to this study are the direct comparison of ventilation and traffic effects on UFP (showing clear dominance of the former – a result that had yet to be verified empirically) and the demonstration of opposing effects on UFP from traffic speed and density. We present a clear link between fundamental traffic flow diagrams (linking traffic speed, traffic flow, and traffic density) and the results of the UFP regression models; we show that changes in in-vehicle exposure are linked to unstable traffic conditions (around 71 kph) and high traffic densities (beyond the break of the linear speed-density relationship at 49 veh $(\text{ln-km})^{-1}$). These findings about traffic effects address gaps in the literature previously identified by Knibbs et al. (2011).

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