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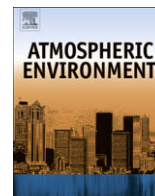


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Ultrafine particles near a major roadway in Raleigh, North Carolina: Downwind attenuation and correlation with traffic-related pollutants

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ABSTRACT

Ultrafine particles (UFPs, diameter < 100 nm) and co-emitted pollutants from traffic are a potential health threat to nearby populations. During summertime in Raleigh, North Carolina, UFPs were simultaneously measured upwind and downwind of a major roadway using a spatial matrix of five portable industrial hygiene samplers (measuring total counts of 20–1000 nm particles). While the upper sampling range of the portable samplers extends past the defined “ultrafine” upper limit (100 nm), the 20–1000 nm number counts had high correlation (Pearson $R = 0.7–0.9$) with UFPs (10–70 nm) measured by a co-located research-grade analyzer and thus appear to be driven by the ultrafine range. Highest UFP concentrations were observed during weekday morning work commutes, with levels at 20 m downwind from the road nearly fivefold higher than at an upwind station. A strong downwind spatial gradient was observed, linearly approximated over the first 100 m as an 8% drop in UFP counts per 10 m distance. This result agreed well with UFP spatial gradients estimated from past studies (ranging 5–12% drop per 10 m). Linear regression of other vehicle-related air pollutants measured in near real-time (10-min averages) against UFPs yielded moderate to high correlation with benzene ($R^2 = 0.76$), toluene ($R^2 = 0.49$), carbon monoxide ($R^2 = 0.74$), nitric oxide ($R^2 = 0.80$), and black carbon ($R^2 = 0.65$). Overall, these results support the notion that near-road levels of UFPs are heavily influenced by traffic emissions and correlate with other vehicle-produced pollutants, including certain air toxics.

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1. Introduction

Among the numerous components of vehicle-produced pollution, ultrafine particles (UFPs, diameter < 100 nm) are of interest as a potential health threat to populations living and working near major roadways. While the ultrafine fraction has a minimal contribution to current regulatory mass-based particulate matter measures (PM_{2.5} or PM₁₀, particles with diameters less than 2.5 μm or 10 μm, respectively), UFPs typically dominate the particle number size distribution. Given their small size, UFPs have been shown to efficiently penetrate the respiratory system and even transfer to extrapulmonary organs, including the central nervous system (Oberdorster et al., 2004; Elder et al., 2006; Elder and Oberdorster, 2006). Other studies indicate that UFP exposure is linked to adverse effects on respiratory and cardiovascular health, with comparably stronger associations observed than for PM_{2.5} (Peters et al., 1997;

Gilmour et al., 2004; McCreanor et al., 2007; Araujo et al., 2008). These findings motivate further research to characterize UFP emissions and spatial variability in the ambient atmosphere.

Roadway traffic has been shown to be an important source of UFPs. The majority of particulate matter emitted through vehicle tailpipe exhaust occurs in the PM_{1.0} (diameter < 1 μm) size range, with mass median diameter generally between 100 and 200 nm (Kleeman et al., 2000; Robert et al., 2007a,b) and number median diameter around 20 nm (Janhall et al., 2004; Kittelson et al., 2004). At a near-road location, UFPs were observed to have a magnified response to roadway emissions in comparison to larger particle sizes (Molnar et al., 2002). Other studies have observed a strong spatial gradient associated with UFPs, exponentially decreasing with distance from major roadways (Hitchins et al., 2000; Zhu et al., 2002a,b; Beckerman et al., 2007; Pohjola et al., 2007). These results suggest that traffic is a major source of UFPs and heavily influences air concentrations in the nearby vicinity of a major roadway.

While UFPs are generally known to be strongly influenced by traffic emissions, the characteristics of traffic-produced UFPs, relationship to co-emitted species, and spatial scale of impact are

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areas of needed research. This study reports new findings from the large-scale Raleigh Near-Road Study that occurred in the summer of 2006. A unique strength of this sampling campaign was the simultaneous real-time measurement of UFPs at five locations, allowing for parallel time series analysis of traffic impact at distances ranging 20–300 m from the road. In addition, UFPs are compared with real-time measurements of other traffic-related species, traffic counts, and meteorology.

2. Methods

2.1. Site description and schedule

An intensive near-road monitoring campaign took place alongside a major roadway (~125,000 vehicles per day) in Raleigh, NC during the summer of 2006. This study has been previously described in detail (Baldauf et al., 2007); thus, only the monitoring components pertinent to this analysis will be discussed. An aerial view of the monitoring site is provided in Fig. 1, including marked locations of monitoring instrumentation that will be discussed in this paper. Total counts of UFPs were measured along a transect perpendicular to the roadway, with four sites located at 20–300 m NE of the roadway and an “upwind” site located approximately 50 m to the SW. This transect was located in a clearing with free-flowing air transport, with only one major obstacle (a two-story building located at 120 m from the road) possibly impacting UFP concentrations. Additional atmospheric measurements conducted at 20 m from the roadway included oxides of nitrogen (NO, NO₂), carbon monoxide (CO), black carbon (BC), PM_{2.5}, PM₁₀, and air toxics (benzene, toluene, naphthalene, methylnaphthalene). For this current analysis, only the two air toxics with highest measured concentrations (benzene and toluene) will be discussed. Meteorological measurements (wind speed, wind direction, temperature) were conducted at 5 m, 20 m, and 300 m from the nearest travel lane.

Field sampling was conducted over July 27–August 22, 2006, although not all instruments were operating throughout the entire

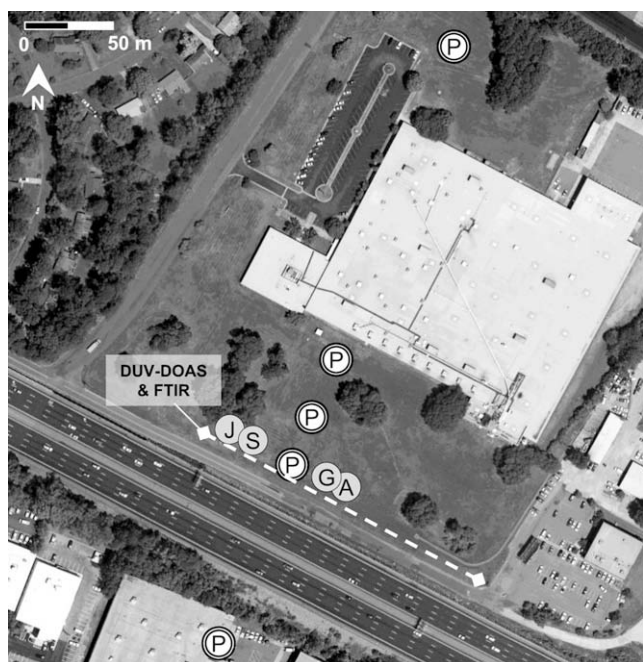


Fig. 1. Near-road sampling campaign in Raleigh, North Carolina including approximate locations of the P-Trak samplers (P), SMPS (S), GRIMM (G), Jet-REMPLI (J), Aethelometer (A), and open-path DUV-DOAS and FTIR instruments.

period. The numerous measurements in this study had various on and off time periods, limited by factors such as available power, personnel, daily maintenance/calibration, or instrument troubleshooting. The intensive portion of the study took place over August 3–10, with nearly every sampler running simultaneously. While field sampling data were collected at intervals from <1 s to 1 min, all air concentration data used in this analysis were averaged at 10-min intervals to account for variable transport time between monitoring sites (e.g. 4.5 minute transport time estimated from 20 to 300 m at 1 m s⁻¹).

2.2. Instrumentation

The main focus of this research is evaluating particulate number concentration data collected through a matrix of five simultaneously sampling industrial hygiene instruments (TSI P-TRAK® 8525, referred to hereinafter as “P-Trak”). These samplers are a cost-effective and portable measurement of total particle number concentration in the size range of 20–1000 nm, operating by condensing isopropyl alcohol onto ambient particles and then counting optically by light-scattering detection. As the P-Trak upper size limit of 1000 nm extends beyond the defined “ultrafine” size range (<100 nm), a comparison with a co-located (20 m site) research-grade ultrafine particle analyzer [Scanning Mobility Particle Sizer (SMPS, TSI Model 3071) with Condensation Particle Counter (CPC, TSI Model 3934)] was performed to determine whether the P-Trak sampler represents the ultrafine range well. SMPS data (105 channels covering a size range of 9.8–414 nm) were collected every 3 min, 45 s, which was averaged per 10-min increment and correlated with the P-Trak measurements for the time period of August 5–10, 2006. High Pearson correlation coefficients ($R = 0.7–0.9$) were determined over the size range of 10–70 nm, including the 10–20 nm region below the P-Trak detection limit. Moderate correlation ($R = 0.5–0.7$) was observed for the size range of 70–100 nm, followed by low correlation for particles exceeding 100 nm in size ($R = 0.3–0.5$). Thus, it can be concluded that the P-Trak measurements well represent the general trend of UFPs and, for ease of terminology, will be referred to as UFPs throughout this paper.

Two other studies have evaluated the P-Trak sampler relative to the SMPS/CPC measurement. One recent study (Drake-Richman et al., submitted for publication) compared multiple (4–10) P-Trak samplers relative to a SMPS/CPC in three environments – a laboratory setting, a smog chamber with injected fresh diesel exhaust, and a field setting with a diesel-powered tractor running nearby. The P-Trak measurement of particle number concentration (D_p ranging 20–1000 nm) was found to have 95% accuracy relative to the SMPS/CPC and $\pm 3–12\%$ precision (Drake-Richman et al., submitted for publication). It should be noted that the P-Trak instruments used in this current study were also used in the Drake-Richman study reported above. A second study reported that the P-Trak underestimated near-road ultrafine particle concentrations relative to the SMPS/CPC although the correlation was high (Zhu et al., 2006). Given the consistent high correlation with the SMPS/CPC observed and the lack of consensus as to a need for a correction factor in a near-road environment, the P-Trak data reported for this study have no correction factor applied.

During the Raleigh Near-Road Study, the five P-Trak samplers were rotated daily to avoid any systematic bias in measurement. For each sampling day, the P-Traks recorded 20-s UFP counts from 5:30 a.m. to 5:30 p.m., with maintenance interruptions (refilling with isopropyl alcohol) occurring each day at 8:30 a.m. and 2:30 p.m. Additional particulate matter measurements used for comparison with the P-Trak data include PM_{2.5} and PM₁₀ measured optically (Grimm Model 107) and black carbon (Magee Scientific AE4 Aethelometer). The Grimm PM_{2.5} and PM₁₀ 1-min optical

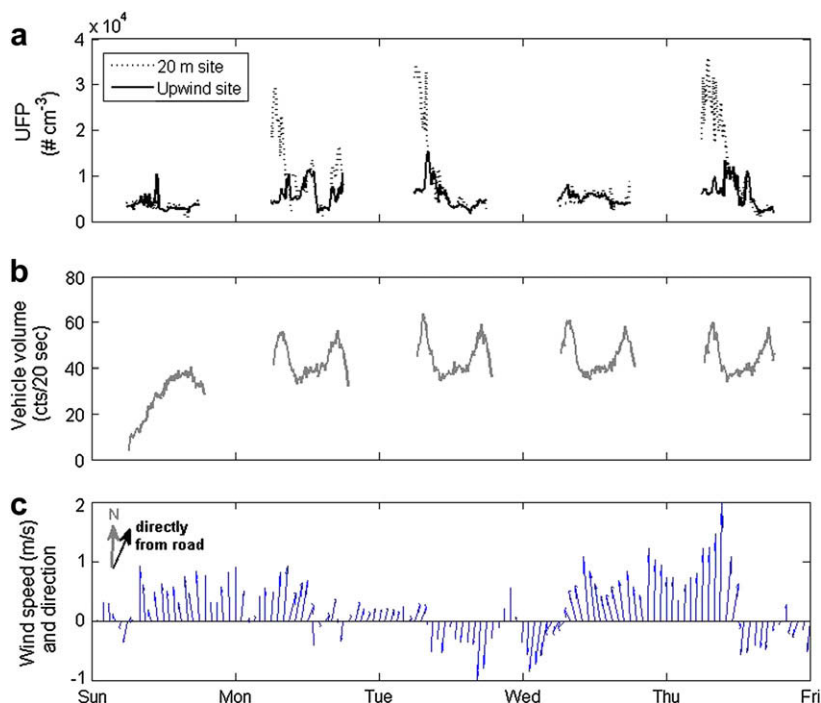


Fig. 2. A time series of P-Trak UFP number concentration (20–1000 nm, 10-min averages) measured over August 6–10 at (a) 20 m downwind and at an upwind site, (b) total traffic volume, and (c) hourly wind speed and direction. Wind vectors are oriented by wind direction and are scaled according to wind speed.

measurements were averaged and linear regression against 24-h average gravimetric $PM_{2.5}/PM_{10}$ data (AirMetrics Mini-Vol) yielded good agreement ($PM_{2.5} R^2 = 0.88$, $PM_{10} R^2 = 0.92$). Gas-phase data used in this analysis include open-path measurements of NO using Deep UltraViolet Differential Optical Absorption Spectroscopy (DUV-DOAS) that has been previously discussed in detail (Thoma et al., 2008) and CO using Fourier Transform Infrared Spectrometry (OP-FTIR). In addition, point measurements were made of NO₂ using a chemiluminescence-based analyzer (API Model 200A) and air toxics (benzene and toluene) using Jet resonance-enhanced multi-photon ionization with time-of-flight mass spectrometry (Jet-REMPI) (Oudejans et al., 2004). Other supporting measurements used in this analysis include wind speed and direction obtained by two methods (RM Young ultrasonic anemometer at 5 m and 20 m and HOBO cup and vane anemometer at 300 m) and traffic volume measured by video and processed using TigreEye™ software.

3. Results and discussion

3.1. Impact of traffic and meteorology

A clear signal of roadway emissions is evident when comparing P-Trak data collected at the 20 m site located NE of the roadway with a background site located at a 50 m distance to the SW of the road (Fig. 2). During morning rush hour with winds directly from the road (Monday, Tuesday, and Thursday), a nearly fivefold difference is observed between the upwind background site and downwind UFP counts. During a morning rush hour period with similar traffic but wind direction away from the 20 m site (Wednesday), low UFP levels are observed at 20 m. The influence of traffic loading is also evident, with low UFP levels observed on a weekend day with winds from the road and also in decreasing UFP levels after morning rush hour with no major change in wind patterns for several hours (Monday, Tuesday, and Thursday).

The lack of a strong evening rush hour signal in Fig. 2 appears to be mainly due to meteorology, given that the traffic volume is at a similar magnitude in the morning and evening rush hour. Only one day (Monday) appears to have noticeable UFP fluctuations after morning rush hour, which may be explained by low speed, meandering winds that occurred from noon through evening. The remaining weekdays either had afternoon wind flow transporting traffic emissions away from the 20 m monitoring site (Tuesday, Thursday) or along the road direction (Wednesday). The lack of a strong evening rush hour signal is similar to the findings for other vehicle-related species (e.g. NO, CO) measured during the Raleigh Near-Road Study (Baldauf et al., 2007; Thoma et al., 2008) and that observed during near-road measurements of UFPs in Sweden (Molnar et al., 2002; Janhall et al., 2004). Increased vertical mixing

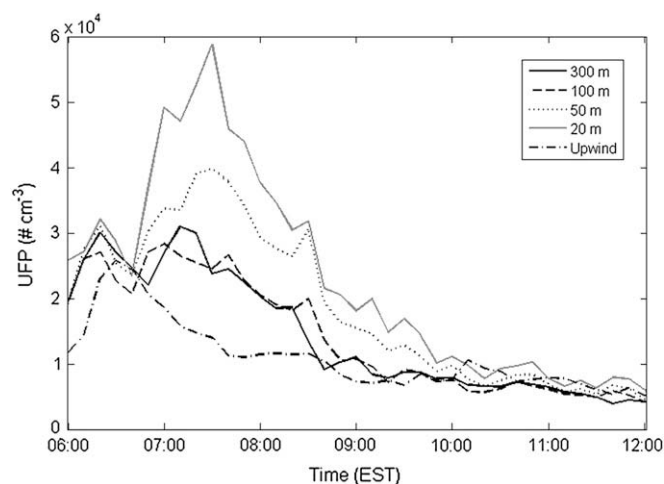


Fig. 3. UFP concentrations on Tuesday, August 14, 2006 at distances ranging 20–300 m away from a major roadway. The P-Trak samplers located at 20–300 m were downwind of the roadway during the shown time period.

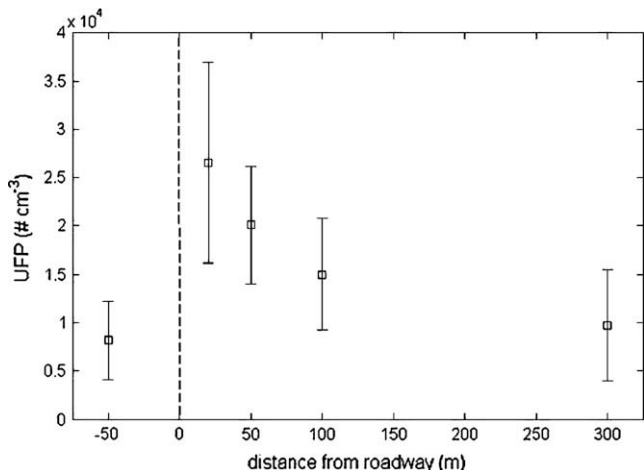


Fig. 4. UFPs measured by P-Trak analyzers during weekday rush hour (6:30–8:00 a.m.) with wind direction from the road (road location indicated by the dashed line). Averages and standard deviations shown represent 7 days (selected for downwind conditions) over the period of August 3–15, with each sampling location having >80% data completeness.

may be another factor contributing to lower near-road UFP levels in the evening (Janhall et al., 2004).

3.2. Horizontal gradient

The change in concentration of roadway-related air pollutants with increasing distance from roads has been a subject of interest in several roadway studies. The UFP data from the Raleigh Near-Road Study are unique in having simultaneous measurements along a transect, rather than limited duration sampling at one location at a time. An example of the five-site parallel time series of UFP data is provided (Fig. 3). During rush hour (6:30–8:00 a.m.), it is interesting that the 300 m site has elevated concentrations that reach a factor of 2 higher than the upwind location, suggesting that the region of impact extends beyond 300 m during heavy traffic. After the morning heavy traffic period ends, all four downwind sites drop to lower concentrations and by 10:00 a.m. are indistinguishable from upwind concentrations.

Given that the largest roadway UFP signal occurred during weekday morning rush hour under downwind conditions, 7 days fitting that description were combined and averaged for each sampling location (Fig. 4). As in the August 14th example (Fig. 3),

a strong exponentially decreasing horizontal spatial gradient is observed. On average, the 20 m site is a factor of 3.2 higher than the upwind location and 2.7 higher than the 300 m site. Several past studies have similarly reported exponentially decreasing downwind UFP concentrations with distance from a roadway (Hitchins et al., 2000; Zhu et al., 2002a,b; Beckerman et al., 2007; Pohjola et al., 2007).

It is challenging to compare UFP gradient results between studies, given that each study had unique monitoring distances relative to the major roadway, took place in separate locations, and measured over different ranges of ultrafine particle sizes. In order to directly compare past research with the current findings, measured gradients of UFPs in various near-road studies are normalized to the UFP level at each study’s respective roadside site and plotted together (Fig. 5). All measurements appear to have a general exponential decay with increasing distance from the road, although the rate of decrease varies from study to study. Possible factors impacting the rate of decrease include background UFP levels at each site, meteorological conditions during each set of measurements, and differences in sampling techniques. All of these referenced measurements were conducted by moving from one location to the next, with varying degrees of repeat measurements taken in each study. Thus, uncertainty may be introduced due to possible changes in meteorology or source strength during a sample set. In contrast, the Raleigh Near-Road Study had simultaneous samples at all sites.

To quantitatively compare the UFP spatial gradient among past field measurements, a linear decrease is approximated for the first 100 m from the roadway (Fig. 5) and the decrease in UFPs with distance is calculated as a percentage drop per 10 m (Table 1). When available, the average UFP upwind concentration or concentration at sites ≥300 m from the road was subtracted from all downwind averages in order for the decrease in only “road source UFPs” to be calculated. Despite the variation in location, monitoring time period, and specific particle diameter ranges, the results are quite consistent with one another. The six studies (this study included) recorded an approximate 5–12% drop in UFPs per 10 m distance from the roadway, with an overall average gradient of 8.5% per 10 m. Although the measured particle size range differs from study to study, all monitoring instruments did sample within ultrafine range (<100 nm) where highest number concentrations are expected in the near-road environment (e.g. Zhu et al., 2002b). Also, the near-road exponential decay in particle counts has been observed to occur at the fastest rate in the ultrafine range, while accumulation mode particles (100–1000 nm) may actually

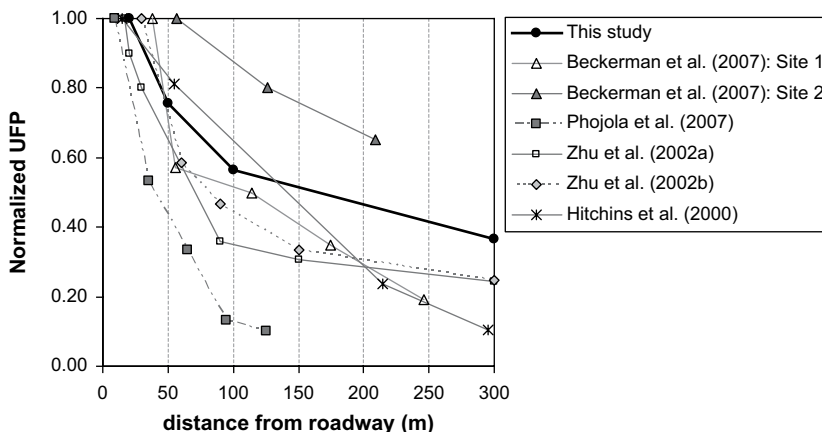


Fig. 5. Comparison of UFP gradient (up to 300 m in distance from the road) measured in this study and in previous research. The data points for each study are normalized the study’s specific UFP level measured nearest to the road (ranging 9–57 m). Each plotted line represents downwind conditions of a major roadway.

Table 1
Comparison of UFP gradients downwind of roadway.

Instrument	D_p (nm)	Location (city, country)	Time period and # of sampling days	Gradient ^a (% per 10 m)	Study
P-Trak	20–1000	Raleigh, USA	August 2006 (7 days)	8	This study
P-Trak	20–1000	Toronto, Canada	August 2004 (2 days)	7 ^b	Beckerman et al. (2007)
SMPS/CPC	3–50	Helsinki, Finland	February 2003 (2 days)	10 ^c	Pohjola et al. (2007)
SMPS/CPC	6–220	Los Angeles, USA	August–October 2001 (7 days)	9	Zhu et al. (2002a)
SMPS/CPC	6–220	Los Angeles, USA	May–July 2001 (9 days)	12	Zhu et al. (2002b)
SMPS/CPC	15–697	Brisbane, Australia	N/A (8 days)	5	Hitchins et al. (2000)

^a Gradient value applies to distances up to ~ 100 m from roadway and is calculated as follows: $\Delta N/\Delta x = [(N_{\text{roadside}} - N_0)/N_{\text{roadside}}] \times [1/(X_{\text{far}} - X_{\text{roadside}})] \times 100\%$, where N values have concentration upwind or at ≥ 300 m (N_0) subtracted.

^b This study reported gradients at two sites; only one site's gradient is shown that more closely fit the 0–100 m distance.

^c No upwind or ≥ 300 m data available to subtract out background UFPs.

gradually increase with distance (Zhu et al., 2002a). Thus, all studies may be observing the similar loss of ultrafine particles over the first 100 m distance.

The UFP gradient results in this study are specific to open transect conditions, which provides a good base case analysis in understanding the spatial impact of roadway emissions. Recent findings of collaborators in the Raleigh Near-Road Study show that roadside topography, such as noise barriers and vegetation, may have a substantial impact on downwind particulate concentrations (Bowker et al., 2007; Baldauf et al., 2008). Because major roadways tend to be located in urban environments with variable near-road topography, further research is needed to translate the clear transect results to a more complex environment.

3.3. Correlative characteristics

The Raleigh Near-Road Study allows a unique opportunity to compare measurements of UFPs using the lower cost P-Trak sampler with state-of-the-art instruments measuring specific roadway pollutants in real time. This analysis provides insight into how well the P-Trak sampler, or bulk 20–1000 nm particle counts in general, correlates with vehicle-related pollutants. A comparison of the 20 m site P-Trak UFP counts with other roadway pollutants (NO_x , CO, $\text{PM}_{2.5}$, PM_{10} , benzene, and toluene) reveals varying degrees of linear correlation (Fig. 6). A strong relationship is observed with NO and CO ($R^2 > 0.70$), although NO_2 has a comparatively much weaker correlation with UFPs ($R^2 = 0.34$). The strong UFP–NO correlation observed in this study agrees with past findings by Janhall et al. (2004). This finding also supports previous analysis of near-road NO_x measured during the Raleigh Near-Road Study which found that NO more closely tracks roadway activities than NO_2 (Thoma et al., 2008).

In addition to NO and CO, black carbon, benzene, and toluene also appear to follow the UFP trends, with correlation coefficients of 0.65, 0.76, and 0.49, respectively. Linear regression equations were calculated for the pollutants that correlated extremely well ($R^2 > 0.70$) with UFPs (NO, CO, benzene) and the linear fit equations are provided in Table 2. NO and benzene are observed to have a negligible y-intercept in comparison to their overall average concentration while CO has a significant offset of 0.16 ppm, assumed to represent the regional ambient background. These predictive equations are likely applicable to future near-road studies, given similar traffic characteristics (fuel type, vehicle mix) and distance from the road (20 m). These results do not necessarily lead to the conclusion that gaseous species highly correlated with UFPs have similar near-road spatial gradients, as UFP levels may be altered by particle-specific post-emission processes such as coagulation and condensational growth.

Finally, it is evident that UFP number count poorly relates to concentrations of $\text{PM}_{2.5}$ (Fig. 6g) and PM_{10} (Fig. 6h), with correlation coefficients < 0.1 . A previous roadside study by Molnar et al. (2002) had similar results, finding essentially no relationship between the ultrafine (< 100 nm) particles and $\text{PM}_{2.5}$. As the mass-based $\text{PM}_{2.5}$ depends disproportionately on the number of larger particles, this lack of correlation suggests that, in the near-road environment, the ultrafine mode is more heavily influenced by roadway traffic than larger-sized particles.

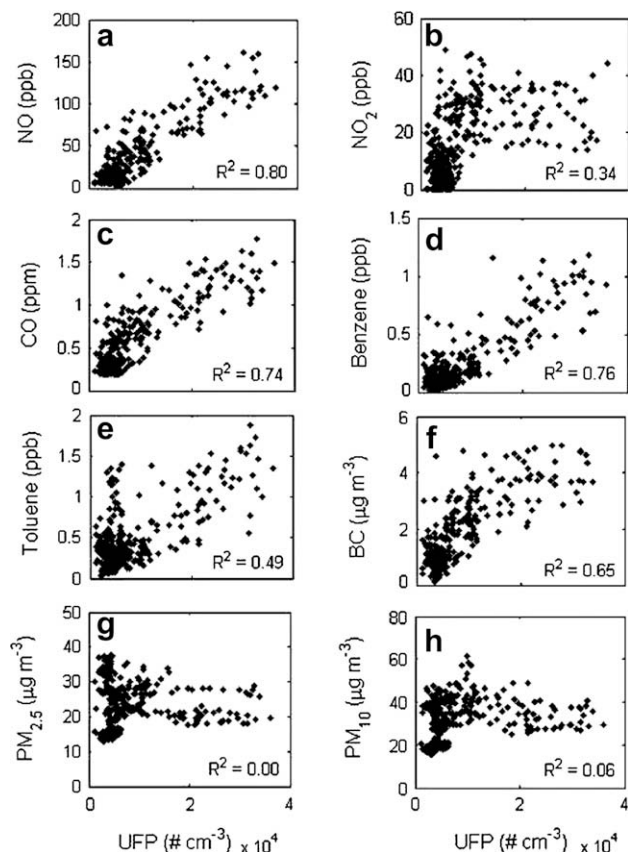


Fig. 6. P-Trak UFP number count (10-min average, August 5–10, 2006) compared with (a) NO, (b) NO_2 , (c) CO, (d) benzene, (e) toluene, (f) black carbon, (g) $\text{PM}_{2.5}$, and (h) PM_{10} . All measurements were at 20 m from the roadway. Also shown are linear regression correlation coefficient values, with UFPs set as the independent variable.

Table 2
Linear regression equations with the P-Trak as an independent variable.

Component	Units	N^a	Average concentration	Equation ($y = mx + b$) $x = \text{UFP} (\# \text{ cm}^{-3}) \times 10^{-4}$
NO	ppm	338	0.03	$= 0.043x - 0.002$
CO	ppm	388	0.49	$= 0.43x + 0.16$
Benzene	ppb	410	0.20	$= 0.29x - 0.02$

^a Each data point represents a 10-min period during August 5–10, 2006.

4. Conclusions

During an extensive near-road field campaign conducted in Raleigh, North Carolina in 2006, ultrafine particulate matter was measured using industrial hygiene P-Trak samplers (measurement range 20–1000 nm) simultaneously at five locations perpendicular to a major roadway. The bulk 20–1000 nm number counts had high correlation with size-segregated ultrafine counts (10–70 nm) measured by a co-located SMPS/CPC, supporting the use of the P-Trak data as representative of UFP trends. The five parallel time series data shows that weekday morning commute emissions elevate ambient UFP levels more than 300 m downwind of the roadway. A strong downwind horizontal gradient is apparent, estimated at an 8% drop per 10 m distance.

An intercomparison between pollutants measured at 20 m from the road indicates that UFPs have moderate to strong correlation with roadside concentrations of air toxics (benzene, toluene), black carbon, NO, and CO. The inter-pollutant relationship also implies that epidemiological research seeking to isolate pollutant-specific impacts of traffic emissions may have challenging confounding variables. Finally, it is important to note that this study agrees with past findings of a poor relationship between UFPs and levels of regulatory mass-based particulate matter (PM_{2.5} and PM₁₀). This finding suggests that while traffic contributes to regional levels of PM_{2.5} and PM₁₀, the local-scale impact (hundreds of meters) is most strongly evident in the ultrafine size fraction.

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