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Near Roadway Air Quality: Synthesizing the Findings from Real-World Data

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Title: Near Roadway Air Quality: Synthesizing the Findings from Real-World Data

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List of abbreviations and definitions:

ANOVA- Analysis of variance

CARB - California Air Resources Board

CO - Carbon monoxide

EC - Elemental carbon and surrogates

NO - Nitric oxide

NO_2 - Nitrogen dioxide

NO_X - Oxides of nitrogen

PM - Particulate matter

PM_{10} - Particulate matter less than 10 µm in aerodynamic diameter

$PM_{2.5}$ - Particulate matter less than 2.5 µm in aerodynamic diameter

UF1 - Ultrafine particle number (> 3 nm aerodyanimc diameter)

UF2 - Ultrafine particle number (> 15 nm aerodynamic diameter)

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Abstract

Objective: Despite increasing regulatory attention and literature linking roadside air pollution to health outcomes, studies on near roadway air quality have not yet been well-synthesized.

Data Sources and Extraction: We employ data collected from 1983 to 2007 reported in 37 roadside monitoring studies, encompassing more than 600 air pollutant concentration measurements collected at various distances from roads.

Data Synthesis: Two types of normalization, background and edge-of-road, were applied to the observed concentrations. Local regression models were then specified to the concentration-distance relationship and analysis of variance was used to determine the statistical significance of trends.

Conclusions: Using an edge-of-road normalization, most pollutants decay to background by 80 – 600 m from the edge of road; using the more standard background normalization, most pollutants decay to background by 160 - 800 m from the edge of road. For pollutants where both techniques estimated distance-to-background, edge normalization estimated a shorter distance-to-background in 71 percent of cases. Differences between the normalization methods arose due to the likely bias inherent in background normalization, since some reported background values tend to under-predict (be lower than) actual background. Shifts in pollutant concentrations with changes in distance from the road fell into one of three groups: at least a 50 percent decrease in peak/edge-of-road concentration by 150 m, followed by consistent but gradual decay toward background; consistent decay or change over the entire distance range; or no trend with distance.

Introduction

Since the early 2000s, there has been increased regulatory interest in understanding and mitigating near-road air pollution problems in the United States. In 2003, California Senate Bill 352 mandated a quarter mile buffer zone between freeways and schools to reduce the health-related impacts of near road air pollution. In 2004, the Sierra Club litigated to prevent expansion of highway US 95 in Las Vegas, Nevada, citing concerns regarding near-road air pollutants; the lawsuit settlement agreement committed state and federal agencies to monitoring at several roadside locations and to pilot mitigation strategies at nearby schools (Federal Highway Administration 2007). A 2005 California Air Resources Board (CARB) land use guide recommended siting “sensitive land uses” further than 500 feet from a freeway or high-traffic road (CARB 2005).

Concern over near-road pollution is motivated by a growing body of literature examining associations among pollutant concentrations, health impacts, and road proximity. To date, empirical findings on health effects related to near-road pollutant exposures are mixed (Brugge et al. 2007; Edwards et al. 1994; Gauderman et al. 2005; Kim et al. 2004; Lin et al. 2002; McConnell et al. 2006; Pearson et al. 2000; Pedersen et al. 2004; Peters et al. 2004; Savitz and Feingold 1989; Wjst et al. 1993; Wyler et al. 2000; Zmirou et al. 2004), and there have been few attempts to synthesize what is known about real-world near-road pollutant concentrations. This study begins to fill this gap by synthesizing and evaluating approximately three decades of published real-world monitoring data and characterizing the relationships that exist between pollutant concentrations and road proximity.

Two significant meta-analyses of near-road air quality have been undertaken in recent years. In the first, Brugge et al. (2007) reviewed cardiopulmonary health risks associated with near-road exposures and concluded, from a review of eight studies, that ultrafine particle

number, black carbon, carbon monoxide (CO), and oxides of nitrogen (NO_X, including nitric oxide [NO], and nitrogen dioxide [NO_2]) are elevated near roadways and the most important exposure zone extends to those individuals residing 30 m from freeways. In the second study, Zhou and Levy (2007) performed a meta-analysis to determine important parameters affecting the "spatial extent" of impacts resulting from mobile source air pollution. They reviewed 33 studies; 18 were monitoring studies; the remainder involved dispersion modeling, land use regression, biomonitoring and epidemiology. Spatial extent was defined as the distance at which roadway effects were no longer observable; it focused on measures of pollution concentration or health impacts. Their findings varied as a function of the spatial extent, the pollutant type, and local meteorology. Overall, they observed that the concentration-based spatial extent of mobile source impacts ranged from 100 – 500 m from roads. One limitation to this study, as noted by Zhou and Levy, was that results for particulate pollutants were not disaggregated by particulate size and mass fraction, a limitation that has been addressed in this study.

This paper advances understanding of the dispersion of near-road air pollutant concentrations by synthesizing findings from 37 monitoring studies undertaken between 1978 and 2007. The findings document, by individual pollutant type, the distances over which near-road concentrations decay to background. Concentration measurements are normalized using two techniques: normalizing to a background and an edge-of-road concentration. We find that normalizing on the basis of the edge-of-road concentration offers advantages to normalizing by the background concentration because the definition of background concentrations will differ across studies. The findings also complement other work describing the physical and atmospheric processes governing the shape and rate of decay curves for individual pollutants (Zhou and Levy 2007); such prior work has dealt mostly with chemical reactivity and dispersion

impacts on the atmospheric transport and fate of given pollutants such as NO_2 and ultrafine particles (Zhang et al. 2004). Finally, our results can help validate modeling tools or assess under which conditions model estimates are most robust. Pollutant exposure is determined by many factors such as time of day and location of activities (Sexton and Ryan 1988); to the extent that exposure occurs in the near-road environment, this study provides a stronger scientific context for designing buffer zones to avoid exposure to higher pollution concentration levels.

Methods

Data assembly and preparation. To populate our database, we included reported distance/concentration pairs from all monitoring studies that included information or findings on near-road concentration gradients, including upwind conditions to help establish background values. Although a diversity of measurement approaches and technologies have been used to assess near-road concentrations, the most frequently applied method was to arrange pollutant monitoring equipment along a vector approximately perpendicular to the road. Distances and pollutants varied among studies, as did motivating factors. Some studies collected data solely to observe near-road conditions; other studies were designed to improve model verification or calibration. Collected measurements typically involved measurement campaigns conducted over several weeks or longer.

The literature confirms what one might intuitively expect: meteorology—wind speed and direction—strongly affects near-road pollutant concentrations. When wind flows from the road to receptors, concentration gradients are more pronounced and extend to a greater distance than when wind is parallel to or away from receptors (Hitchins et al. 2000). In general, concentrations decay to background within a few hundred meters downwind of a road, although studies

measuring pollutants solely in the evening hours indicate that higher concentrations persist beyond 500 m (Zhu et al. 2006).

Our analysis unit was one distance/concentration pair (e.g., a single CO measurement at 30 m from the edge of road). We identified 619 such pairs from 37 papers (Ashbaugh et al. 1996; Cahill et al. 1994; Clements et al. 2008; Cyrys et al. 2003; Gidhagen et al. 2004; Gilbert et al. 2003; Hagler et al. 2008; Harrison et al. 2003; Hitchins et al. 2000; Hoek et al. 2002; Janssen et al. 1997; Kim et al. 2004; Kuhn et al. 2005a; Kuhn et al. 2005b; Lena et al. 2002; Monn et al. 1997; Nanzetta and Holmen 2004; Nitta et al. 1993; Ntziachristos et al. 2007; O'Donoghue and Broderick 2007; Pirjola et al. 2004; Pleijel et al. 2004; Reponen et al. 2003; Rodes and Holland 1981; Roorda-Knape et al. 1998; Sabin et al. 2006; Shi et al. 1999; Singer et al. 2004; Smargiassi et al. 2005; Tiitta et al. 2002; Vardoulakis et al. 2005; Weijers et al. 2004; Wu et al. 2003; Zhu et al. 2002a; Zhu et al. 2002b; Zhu et al. 2006; Zhu et al. 2008) published as of mid-2008; the literature represents wide geographic, meteorological, and traffic operational variation. (An annotated bibliography of all studies used in the analysis is included in Supplemental Material.)

A recent 2009 publication (Hu et al. 2009) that is not included in our database and that complements our study found differences between daytime and nighttime near-road pollutant concentrations. Our study synthesized available (as of mid-2008) literature on near-road monitoring, virtually all of which included daytime-only data or data which included daytime measurements. Since on-road activity tends to be greatest during daytime hours, we generally expect the daytime component of the measurements in our database to dominate reported concentrations. There is some indication however, that nighttime near-road ultrafine particle number concentrations can occasionally exceed daytime conditions, despite reduced traffic volumes (Hu et al. 2009).

Our final database includes distance/concentration pairs that spanned 104 unique measurement sets. A measurement set is defined here as a group of distance/concentration pairs originating from the same study and representing one pollutant under one set of measurement conditions. Many studies reported results from different observation days, seasons, or traffic conditions. If these data were available from the study results, we recorded them as separate measurement sets for analysis. Approximately 65 percent of included studies involved measurements near freeways or highways; the remainder involved measurements near arterial and/or local roads.

Most of the observations collected from the studies were obtained within 150 m of the roadway (Figure 1a) and focused on particulate matter (PM) mass (31%), particle number concentrations (17%), elemental carbon and surrogates (EC) (16%), and all oxides of nitrogen (15%) (Figure 1b).

Field measurements were grouped by pollutant type or surrogate (EC includes black carbon, black smoke, and the reflectance of PM filters). Ultrafines were also grouped, but as a separate category. The term “ultrafine” typically refers to particles less than 100 nm in diameter (Donaldson et al. 2001), and particle number concentrations (as opposed to mass concentrations) are typically used to quantify ultrafine roadside concentrations. We categorized particle number concentration into three groups: UF1 particle number denotes data collection beginning at 3 nm, UF2 particle number signifies data collection beginning at 15 nm, and fine particle number begins at 300 nm (0.3 µm, just above the ultrafine classification).

Normalization. Monitored concentration data is typically normalized to wind speed or traffic volume (Zhu et al. 2008), to a reference near-road distance (Pleijel et al. 2004; Reponen et al. 2003), or by subtracting out background concentration (O'Donoghue and Broderick 2007;

Weijers et al. 2004). There are problems in normalizing to traffic volume or meteorological conditions when aggregating data across numerous studies. First, many studies do not provide sufficient information (e.g., temporal resolution) in either reports or papers to derive similar measures of traffic or meteorological conditions elsewhere. Second, even when data can be gathered, studies frequently aggregate or resolve data to the units most useful for that particular study interest. For example, daily traffic might be used for cumulative effects, whereas peak hour traffic might be applied for a study interested in acute effects. We have chosen two types of normalization procedures that can easily be replicated in future studies and rely on factors that are usually readily available from, or described in published work.

The first, normalizing to background yields the relative concentration of pollutants measured in the near-road zone compared to nearby concentrations unaffected by (typically upwind of) the road. This normalization can directly identify whether and where measured concentrations fall to background levels. The normalization divides observed near-road concentrations by the reported background value; as values approach one, near-road concentrations approach background.

The second approach, normalizing to edge-of-road yields the relative concentration of pollutants in the near-road zone compared to concentrations measured at the point of expected maximum roadway influence: the roadway edge. This type of normalization indirectly allows assessment of whether and where measured concentrations fall to background levels. The edge-of-road normalization approach has two benefits relative to background normalization. First, it enables use of data from (many) studies for which no background measurements were published (and sometimes not recorded). Second, it avoids data comparison problems—since there is no standard protocol in use to measure near-road background concentrations, background

concentrations reported in monitoring may result from a variety of measurement approaches and locations relative to the road being studied.

Edge-of-road normalization involves dividing all concentrations in a measurement set by the edge-of-road concentration. The resulting values indicate whether and at what distance from the road the normalized values reach a stable value. In concept, stability is indicative of near-road concentrations approaching or reaching background, although it is numerically possible (though physically less likely) that stability could also represent a steady concentration above background. (See Supplemental Material for further edge normalization details.)

Previous work indicated that differences in meteorology (Kuhn et al. 2005b; Ntziachristos et al. 2007), traffic volume and fleet composition (Gramotnev and Ristovski 2004; Zhu et al. 2004), and other factors (Baldauf et al. 2008) can contribute to differences in observed concentrations of traffic-related air pollution. However, to the extent that the *shape* of the concentration decay curve is expected to be roughly similar across multiple studies for a given pollutant, dividing by the roadway edge concentration should preserve the shape while removing the absolute magnitude of the observations. This intuition has been confirmed by recent work on the influence of roadway configuration and sound/vegetation barriers on observed concentrations (Baldauf et al. 2008) and in other normalized comparisons undertaken by Zhu et al. (2008) for three facilities normalized to unit wind speed and traffic volume. Others have taken a similar approach with more limited data; for example, Pleijel et al. (2004) compared Swedish and Canadian monitoring data for NO_2 by dividing all observations by the NO_2 concentration at 10 m from the road.

Local regression. Locally weighted regression (loess) was used to regress concentration on distance for both sets of normalized data Loess is a robust smoother that does impose a

functional form on the relationship between the dependent and independent variables (Cleveland and Devlin 1988). The smoother uses a specified data window that moves along the x axis of a scatterplot. At each data point a fitted value is calculated using the subset of the data contained within the moving window. The size of the subset is defined as a percentage of total data and is referred to as the smoothing parameter; larger smoothing parameter values produce smoother concentration vs. distance curves. Local regression has previously been applied to near roadway data by Gramotnev and Rivoski (2004). However, the authors did not specify the value of the smoothing parameter used. We set the smoothing parameter by visual inspection. Parameter values of 0.75 and 0.70 (background normalization and edge normalization, respectively) produced smoothed curves sufficient for the purpose of our research. A degree of one, indicating locally linear fits, was used in all cases.

Analysis of variance. Discussion of statistical significance is rare in the near-road literature. In studies that do conduct statistical analysis, paired *t*-tests comparing observed concentrations to a reference group typically located closest to the roadway are used (Roorda-Knape et al. 1998; Smargiassi et al. 2005). Sabin et al. (2006) used paired t-tests and an analysis of variance (ANOVA) to test differences in the dry deposition rates of metals between downwind locations. However, there is some question as to whether near-road pollution concentrations (in addition to other meteorological and traffic measurements) meet the normality criteria for a *t*-test, and in at least one study the non-parametric Mann-Whitney test has been used (Zhu et al. 2004). The distribution of observed near-road concentrations may be skewed since there are generally a large number of low-concentration observations. If deviation from normality is very large, then ANOVA results may not be robust. To overcome this possibility, we applied both parametric and non-parametric tests. We performed an ANOVA to identify the magnitude and

significance of changes in concentration by distance from road and augmented the ANOVA results with the nonparametric Kruskal-Wallis test (Myers and Well 1991, p. 103-4).

Results and Discussion

Background normalization. The background-normalized concentrations are shown in Figure 2, and near-road concentrations and distance-to-background values are summarized in Table 1. In Figure 2, the range on the y-axis has been constrained to five times above background concentration. The only pollutant exceeding this is CO, which was observed to reach 20 times above background at the roadway edge. The range on the x-axis in Figure 2 has been constrained to 0 – 450 m from the edge of road. We also excluded data from two studies because the sampling and vehicle fleet characteristics were very unique relative to the rest of the studies. One study measured concentrations only at night; another study measured near-road conditions in 1978, when vehicle emissions and near-road concentrations were substantially higher than the values reported in other studies (Rodes and Holland 1981; Zhu et al. 2006). Organic carbon and sulfur are not shown in Figure 2 due to limited data. (See Supplemental Material for further discussion of omitted data.)

Changes in pollutant concentrations over distance generally fell into three groups. The first showed rapid initial concentration decay—defined here as at least a 50 percent decrease in peak/edge-of-road concentration by 150 m—followed by consistent but more gradual decay toward background; the second consistently decayed or changed over the entire distance range, while the third showed no trend with distance.

One pollutant, ozone, which is shown in the second panel, displayed an increasing trend beginning far below background near the road, and gradually approaching background by 400 m from the edge. This trend was unique among the other pollutants included in the study. However,

ozone values were consistent with expected near-road titration due to interaction with direct vehicle emissions of NO.

All pollutants except for UF2 particle number reached background by approximately 400 m. The high concentrations shown in the first 100 m drops off by 400 m, even considering the between-study differences in methods and traffic characteristics; this is notable considering the wide variation in data and the inherent limitations of this normalization method (i.e., the lack of common protocols used to define background). The curves indicate (ignoring ozone) that concentrations of certain pollutants are elevated near roadways, and decrease as the distance increases, while other pollutants show no roadway influence. These background normalized results suggest that a range of approximately 160 – 400 m is sufficient to reach background concentrations for the majority of pollutants.

Edge normalization. The results for the normalization to roadway edge are shown in Figure 3 and summarized in Table 2. We were able to include more data in the edge normalization than background normalization since background measurements or estimates were not a prerequisite to normalizing in this method. Exponential fits of individual measurement sets were used to determine edge concentrations for EC, benzene, CO, metal deposition, non-alkane hydrocarbons, NO, NO_2, NO_X, ozone, and UF1/UF2 particle number. Previous work has shown that an exponential decay describes the atmospheric fate of pollutants which vary by distance (Gramotnev and Ristovski 2004; Pleijel et al. 2004; Zhu et al. 2002a; Zhu et al. 2002b). We used linear regression to estimate an edge concentration for pollutants that showed little variation with distance according to the supporting annotated bibliography (PM_{10}, $PM_{2.5}$, fine particle number, and alkanes). This will not affect the shape of the decay curve in the event that the concentration actually varied exponentially with distance. The estimated value is used simply to normalize the

rest of the measurement set. The linear estimate would typically be below the highest value of an exponentially decaying pollution concentration, and would lead to near road estimates of concentration greater than one (i.e. higher than the edge-of-road value). We omitted organic carbon, sulfur, and metal deposition from Figure 3; the data for these pollutants were too sparse to smooth without significantly increasing the smoothing parameter value. We also omitted ozone because of its increasing concentration with increased distance from the road and data from the Zhu et al. (Zhu et al. 2006) study, which included nighttime-only results. (See Supplemental Material for more detail on omitted data and edge normalization.)

Edge normalization provides the percentage decrease in pollution concentration as measured from the roadway edge to the distance of interest (Figure 3). For concentrations that varied by distance, the percentage of the near road high concentration at which leveling occurred represents a proxy of that pollutant's background concentration; this assumes that the roadway influence has dropped to approximately zero when no further changes occur in the smoothed curve.

If the fitted values level off and then decrease again, neither the distance to background concentration, nor the edge-of-road vs. background concentration ratio, is consistent across studies. Larger percentage declines in normalized concentrations indicate a greater contribution of the roadway vs. background to total near-road pollutant measurements (Table 5).

Figure 3 shows CO, benzene, EC, non-alkane hydrocarbons, NO, NO_X, NO_2, $PM_{2.5}$, and UF1 and UF2 particle number concentrations all decreased as distance from road increased. PM_{10}, alkanes, and fine particle number showed ambiguous or little to no trend with distance.

We again categorized rapidly decaying pollutants as those which decreased at least 50 percent from their peak value by 150 m. Several pollutants exhibited sharp declines within the

first 100 – 150 m before leveling off. NO and CO showed the greatest declines. Benzene, EC, NO_2, and $PM_{2.5}$ showed gradually decreasing trends. Benzene declined continuously to 300 m indicating that background concentrations were not reached over the plotted distance range. NO_2 trended similarly, but flattened between 104 m and 137 m before again trending down and reaching 60 percent of its near-road value by 600 m. UF2 particle number showed substantial declines over the entire plotted distance range, but did not appear to level until approximately 570 m from the road. Most of these pollutant concentrations appear to reach background by 80 m to 600 m from the edge of road.

Limitations and differences between normalization methods. We have introduced a new normalization technique to the literature, partially to offset limitations of using the standard background normalization. In the background normalization there is no standard protocol to establish near-road background values across studies. If, in a particular study, background is mischaracterized as either too high or too low, that study's results can obscure or overstate trends when pooled with other study findings. Different studies variously defined background as concentrations measured at the edge of the upwind lanes, some distance from the upwind lanes, the nearest stationary monitoring site, or other locations. In general, high background concentrations will tend to generate flatter gradients and low background concentrations will generate steeper gradients.

For example, background measurements of PM_{10} for a study in Macao, China were taken on a separate island at sites located 2-4 km away from the roadways under study (Wu et al. 2003). The resultant low background measurements tended to inflate the background-normalized Macao concentrations relative to other studies in our database that typically measured background just upwind of the roadway under study.

As another illustration, different background measurement protocols resulted in anomalous UF2 particle number findings. Particle number concentrations increase with measurement of smaller-diameter particles (Molnar et al. 2002; Nanzetta and Holmen 2004; Shi et al. 1999). However, in Figure 2, normalized UF2 particle number (>15 nm diameter) concentrations exceeded UF1 particle number (>3 nm diameter) concentrations. The explanation involves a study by Hitchins et al. (2000) which reported measured UF2 particle numbers. The authors did not take background measurements, but did report concentrations when the wind direction was from the receptors to the road—a background estimation approach that has been used in some other studies (Gidhagen et al. 2004). Additionally, Hitchins et al. (2000) reported concentration values under several different wind speed scenarios. The highest values of UF2 particle number were reported at the lowest wind speed, but the background value was given for conditions with a higher wind speed. In this case, normalizing by the reported background concentration resulted in exaggerated concentration values. If another study contemporaneously reported background and near road values, it would likely show lower normalized concentrations. This highlights the difficulties associated with pooling data from studies that frequently employ different measurement and reporting protocols. Depending upon the sample size across studies, a single study can substantially alter the position of a pollutant's background normalized curve by reporting a background concentration much higher or lower than the background values found in other studies.

In general, it is likely that the bias across studies is for some reported background values to under-predict (be lower than) actual background, due to lengthy averaging periods for background vs. near-road measurements, or use of monitoring locations at relatively unpolluted sites distant from areas immediately upwind of the roads studied. This bias would tend to

increase the background normalized values estimated here, and lengthen the estimated distance required to reach background (affecting results shown in Figure 2).

When normalizing by edge concentrations, the data yield their own background value by virtue of leveling off. The general assumption with edge normalization is that, when pooling data from numerous studies, if the regression approaches a horizontal line, this approximately signifies that roadway influence has diminished to background.

A limitation of the information derived from edge normalization is the possibility that concentrations level off at values which are site-specific—for example, due to abnormally high background concentrations or due to conditions that inhibit dispersion and result in stabilized concentrations above background. If site-specific situations caused the distance at which background was reached to vary across studies, or caused the stabilized value to be an unusually large fraction of the edge-of-road concentration, the regression results (Figure 3) may be biased (e.g., stabilized values higher than background, or the point of stabilization at a shorter distance than would be expected without unusually high background).

Overall, compared to background normalization, edge normalization showed a more rapid decline to background concentrations for non-alkanes, EC, NO, NO_X, and UF2 particle number concentrations and showed a less rapid decline for NO_2 and UF1 particle number concentrations (Table 1 vs. Table 2). These differences would likely be smaller if studies better matched background concentrations to the location of near-road measurements.

Analysis of variance. Some of the most frequently cited studies using real-world observations (Zhu et al. 2002a; Zhu et al. 2002b) show substantial pollutant reductions by 80 m from the road, only slightly shorter than the 100 m zone of highest exposure for some pollutants found in a recent meta-analysis (Zhou and Levy 2007).

Near-road concentrations have traditionally been modeled as a Gaussian plume (see, e.g., Pearson et al. 2000) with as much as 96 percent of the concentration having dissipated by 150 m (Green et al. 2009). Our work, however, suggests that decay regimes may be slightly more complex and possibly organized into those pollutants that, under certain conditions, decay rapidly, those that decay gradually, and those that do not decay. To test this hypothesis, we divided our data into three different groups organized by findings in the literature. Specifically, the first bin (0-80 m) represents the window of anticipated peak concentration, as evidenced by our synthesis and widely referenced work (Zhu et al. 2002a; Zhu et al. 2002b); the second bin (80-120 m) captures the window in which some of the literature has flagged as the end of the spatial extent of mobile source impact (Zhou and Levy 2007), and finally, the third bin (120 m and beyond) represents a reference for the distance range where the literature (modeled and monitored) suggests a substantial decline in observed roadway influence. Some pollutants have no data in the second bin, but this simply reduces the test to a comparison between the first and third groups. The null hypothesis in this case is that there is no difference in mean observed concentrations between observations near the road (i.e., the first distance bin) and observations further downwind of the road (distance bins two and three).

The mean values (coefficients) for the pollutant within distance category are reported in Tables 3 and 4 for background and edge normalized data, respectively. The coefficients in the second and third distance bins (i.e., 80 – 120 m and > 120 m) are mean changes relative to the first category. It should be noted that the means are not directly comparable to the loess plots, since the loess algorithm uses a weighting function to calculate its fitted values at each data point.

All coefficients in the second and third bin for both normalization methods are negative (except for ozone), since concentrations generally decrease when moving from the first to the second and third distance categories. Results from the Kruskal-Wallis test and ANOVA are generally in agreement, indicating that any deviations from normality are generally not severe enough to affect the ANOVA results.

Background normalized concentrations. Mean values of multipliers above background in the first 80 m from the road range from a factor of 2.1 for benzene to 12.4 for CO. Surprisingly, NO does not seem to vary significantly with distance from road. However, its sample size is small and drawn from one dataset with a long averaging time. Both of these issues could obscure detection of differences by distance. The Kruskal-Wallis test also supports the hypothesis that there is no difference between the groupings of NO (large *p*-value).

For PM_{10}, the *F*-statistic from the ANOVA differs from the *p*-value from Kruskal-Wallis. PM_{10} is on average 1.5 times above background concentrations (0 – 80 m), and declines by an average of approximately 0.4 from the near road value beyond 120 m (27 percent decrease). Benzene shows the next smallest significant increment above background at 2.1, declining to 1.1 past 120 m (48 percent decrease). Each of the remaining pollutants that vary by distance both begin at a higher above-background increment than PM_{10} and decrease more sharply. Thus, the relationship of PM_{10} with distance appears to be weak, if it exists. $PM_{2.5}$, fine particle number, and sulfur do not vary by distance, and alkanes show a very weak statistically significant relationship with distance bin, decreasing by eight percent over the distance range greater than 120 m from the road.

Edge normalized concentrations. Concentrations were significantly different for CO, NO, NO_2, UF1 and UF2 particle number when comparing the second (80 – 120 m) and third

distance bins (> 120 m) to the first (0 – 80 m). NO_X concentrations were significantly different when comparing the second distance bin to the first. EC, metal deposition, non-alkane hydrocarbons, and fine particle number showed significant decreases in concentration when comparing the third distance bin to the first.

Benzene, alkanes, PM_{10}, $PM_{2.5}$, and sulfur all show insignificant *F* statistics. In contrast with the results normalized to background concentrations, NO concentration changes with road distance are significant when normalized to the edge-of-road value.

The Kruskal-Wallis *p*-value indicates significant differences among means by distance group across all pollutants ($p < 0.05$) except for benzene, ozone, sulfur, alkanes, and PM_{10}. Ozone is the only pollutant which shows disagreement between tests; however, the Kruskal-Wallis p-value (0.06) is approaching significance. The discrepancy could be the result of the skewed distribution of ozone concentrations.

Conclusions

This study synthesized near-road air pollutant monitoring data collected from 1983 to 2007. Table 5 summarizes the concentration gradients of each pollutant that showed a significant variation with distance in both ANOVA models (see Tables 3 and 4 for significance). The findings show that the influence of the roadway on air pollution concentrations decays to background between 80 – 600 m according to edge normalization and between 160 – 800 m based on background normalization. These ranges cover all background normalized pollutants except for CO, which declines continuously to 285 m (end point of available CO data), and all edge-normalized pollutants except for metal deposition which was too sparse to smooth. Edge-of-road concentrations were elevated from 1.7 – 20 times above background.

The trends indicated by both normalization methods are broadly consistent, not considering the specific distance at which background is reached. For example, as shown in Table 5, there is general agreement in terms of the increment at the roadway edge relative to background concentrations (at least to an order of magnitude, in the case of CO and NO_2, and much closer for EC, NO_X, and UF1 particle number).

Limitations of this study and the literature on which it is based motivate consideration of standardized approaches to near-road data collection. The data contained in this study was aggregated from studies with non-uniform sampling procedures and non-uniform locations (i.e., different roadway, geographic, and meteorological conditions). Additionally, the sampling periods employed by each investigator typically varied between several hours and several days. Concentrations averaged over longer time periods will vary less than those measured during shorter time intervals. Background concentrations were not always averaged over the same period as the associated near-road measurements, nor were they taken in similar locations relative to the road. The choice of background measurement technique can over- or under-state roadway increments for a single study; as evidenced by Table 5, the variability in reporting background concentrations may result in an overall bias to over-estimate the distance at which pollutant concentrations decay to background.

Key considerations for future near-road work include: standardizing the location and method of obtaining background measurements and reporting more completely on site conditions. Some studies at specific sites have assessed how changes in traffic volumes or meteorological characteristics affected near-road concentrations (Janssen et al. 2001; Ntziachristos et al. 2007; Rijnders et al. 2001). Greater and more consistent specification of site conditions in future work will broaden understanding of the key factors that contribute to near-

road concentrations. This study is based on published data as of mid-2008; we excluded data from two studies (Rodes and Holland 1981; Zhu et al. 2006) not directly comparable to the rest of the published literature. Some of the excluded data (Zhu et al. 2006), combined with findings from a more recent publication (Hu et al. 2009), indicate that nighttime or pre-sunrise conditions can substantially lengthen, to perhaps two or three thousand meters, the distance at which near-road pollutant concentrations decay to background. Further work is needed to integrate daytime and nighttime measurement results, and to assess their relative importance given daytime and nighttime differences in travel activity, near-road pollutant concentrations, and factors affecting human exposure.

In addition to integrating nighttime and daytime near-road findings, future work should update the findings presented here to reflect ongoing research. Additional near-road measurement results were published following assembly and analysis of the 2007 and earlier data presented in this paper (e.g., Barzyk et al. 2009; Buonanno et al. 2009; Hagler et al. 2009). Findings from recent studies are consistent with results presented here—they show that daytime near-road concentrations are generally indistinguishable from background within several hundred meters from the road.

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Tables

Table 1. Summary of background normalized data.

Group	Pollutant	Approximate multiplier above background concentration at edge-of-road	Approximate distance required to reach background concentration (m) [a]
Rapid: >50% drop by 150 m	Benzene	3	160
	CO	20	—[b]
	Metal deposition	3	175
	Non-Alkanes	2.2	160
	UF1 particle no.	4	187
	UF2 particle no.	5	775[c]
Less rapid or gradual decay/change	EC	1.8	400
	NO	—	400
	NO_2	3	360
	NO_X	1.7	400
	PM_{10}	1.5	140

[a]The approximate distances were derived from Figure 2; the distance point at which the smoothed line reached a value of one on the y-axis is cited here as background. [b]CO did not reach background within the 285 m for which data were measured [c]UF2 particle no. reached background concentrations outside of the range plotted in Figure 2.

Table 2. Summary of edge normalized data.

Group	Pollutant	Percentage decrease[a]	Distance (m)	Reached background
Rapid: >50% drop by 150 m	CO	90	120	Yes
	NO	63	80	Yes
	Non-Alkanes	65	150	Yes
	NO_X	50	100	Yes
	UF1 Particle no.	83	210	Yes
	UF2 Particle no.	87	570[b]	Yes
Less rapid or gradual decay	Benzene	40	300	No
	EC	40	90	Yes
	NO_2	40	600[b]	Yes
	$PM_{2.5}$	22	800[c]	No

[a]For pollutant concentrations that reached background: defined as percent decrease in edge-of-road concentration at the stabilization distance. For pollutants that did not reach background: defined as percent decrease in edge-of-road concentration at the furthest distance for which measurement data were available. [b]UF2 particle no. and NO_2 reached background outside of the range plotted in Figure 3. [c]Data for $PM_{2.5}$ extended to 800 m from the edge of road.

Table 3. ANOVA and Kruskal-Wallis results: background normalization.[a]

	ANOVA		Kruskal-Wallis	0 – 80 m		80 – 120 m		> 120 m	
	Df	*F*	*p*-value[b]	Coeff.[c]	n	Coeff.[c]	n	Coeff.[c]	n
Alkanes	(1,22)	4.31*	0.043	1.053	8			-0.0817*	16
Benzene	(1,5)	12.44*	0.034	2.1	3			-0.992*	4
CO	(1,9)	6.14*	0.014	12.41	7			-9.97*	4
EC	(1,45)	9.23**	< 0.001	2.4	32			-1.111**	15
Fine PM no.	(2,13)	3.49	0.14		4		2		10
Metal deposition	(1,13)	30.8#	0.0022	3.06	5			-1.947#	10
Non-Alkane hydrocarbons	(1,34)	69.6#	< 0.001	2.37	12			-1.250#	24
NO	(1,4)	1.852	0.132		1				5
NO_X	(1,13)	4.80*	0.0022	5.55	5			-4.26*	10
NO_2	(2,29)	5.59**	0.0019	2.88	17		9	-1.666**	6
Ozone	(1,4)	58.4**	0.0603	0.1429	2			0.561**	4
UF1 PM no.	(2,40)	6.10**	<0.001	4.78	28		4	-3.51**	11
UF2 PM no.	(2,54)	6.34**	<0.001	6.27	24	-3.51*	9	-4.03**	24
PM_{10}	(2,32)	3.05	0.00185	1.456	23		3	-0.445*	9
$PM_{2.5}$	(2,30)	0.477	0.618		19		2		12
Sulfur	(1,2)	0.0314	0.655				1		3

[a]The last three rows contain those pollutants determined to not vary with distance as judged by significant results from one or the other ANOVA models. Statistical significance is indicated as follows: * $p < 0.05$, ** $p < 0.01$, # $p < 0.001$. [b]The Kruskal-Wallis *p*-value is taken from the non-parametric Kruskal-Wallis test whose null hypothesis is that there is no difference in the mean ranks of the groups. [c]Refers to the regression coefficients extracted from a linear regression of normalized concentrations on distance bin. The coefficient in the 0 – 80 m bin was the model intercept which represents the mean normalized value in that range, while the coefficients in the other two distance bins represent mean changes relative to the first bin. Missing values in the table indicate insignificant results as judged by the omnibus *F* and the Kruskal-Wallis *p*-value, or no data, evidenced by a blank n for the cell.

Table 4. ANOVA and Kruskal-Wallis results: edge normalization.[a]

	ANOVA		Kruskal-Wallis *p*-value[b]	0 – 80 m		80 – 120 m		> 120 m	
	Df	*F*		Coeff.[c]	n	Coeff.[c]	n	Coeff.[c]	n
Alkanes	(1,22)	3.71	0.0659		8				16
Benzene	(2,10)	2.89	0.116		4		2		7
CO	(2,13)	9.14**	0.003	0.626	9	-0.538*	2	-0.510**	5
EC	(2,30)	5.46**	0.01229	0.804	15		1	-0.249**	17
Fine PM no.	(2,13)	3.88*	0.0184	1.018	4		2	-0.202*	10
Metal deposition	(1,13)	144.7[#]	0.00182	1.000	5			-0.615[#]	10
Non-Alkane hydrocarbons	(1,34)	70.5[#]	<0.001	0.794	12			-0.387[#]	24
NO	(2,38)	12.32[#]	<0.001	0.748	22	-0.691**	2	-0.394[#]	17
NO_X	(2,83)	35.4[#]	<0.001	0.884	38	-0.256[#]	16	-0.1851[#]	32
NO_2	(2,22)	3.94*	0.0369	0.737	8	-0.609*	2		15
Ozone	(1,4)	58.4**	0.0603	1.993	2			7.82**	4
UF1 PM no.	(2,45)	18.36[#]	<0.001	0.670	29	-0.305**	6	-0.405[#]	13
UF2 PM no.	(2,51)	28.9[#]	<0.001	0.784	21	-0.219**	9	-0.454[#]	24
PM_{10}	(2,45)	2.16	0.1130		24		7		17
$PM_{2.5}$	(2,36)	0.219	0.0695		15		4		20
Sulfur	(2,5)	0.042	0.799		2		1		5

[a]The last three rows contain those pollutants determined to not vary with distance as judged by significant results from one or the other ANOVA models. Statistical significance is indicated as follows: * $p < 0.05$, ** $p < 0.01$, [#] $p < 0.001$. [b]The Kruskal-Wallis *p*-value is taken from the non-parametric Kruskal-Wallis test whose null hypothesis is that there is no difference in the mean ranks of the groups. [c]Refers to the regression coefficients extracted from a linear regression of normalized concentrations on distance bin. The coefficient in the 0 – 80 m bin was the model intercept which represents the mean normalized value in that range, while the coefficients in the other two distance bins represent mean changes relative to the first bin. Missing values in the table indicate insignificant results as judged by the omnibus *F* and the Kruskal-Wallis *p*-value, or no data, evidenced by a blank n for the cell.

Table 5. Summary of Pollutant Profiles Under Both Normalization Methods.[a]

	Distance from road at which leveling begins or background reached (m)		Percentage of near-road high concentration at which leveling begins or background is reached		Edge-of-road multiplier above background concentration (multiples of background)	
	EN	BN	EN	BN[b]	EN[c]	BN
NO	**80**	400	37	—[d]	2.7	—
EC	**90**	400	60	56	1.7	1.8
NO_X	**100**	400	50	59	2.0	1.7
CO	**120**	—	10	5	10	20
Non-alkane hydrocarbons	**150**	160	35	45	2.9	2.2
UF1 particle no.	**210**	187	17	25	5.9	4
UF2 particle no.	**570**	800	13	20	7.7	5
NO_2	**600**	360	60	33	1.7	3
Metal deposition	—	175	—	33	—	3

Abbreviations: EN is edge normalization; BN is background normalization. [a]Table entries are sorted based on the edge normalized distance at which background concentrations are reached (**bold**). Pollutants that showed significant results in both ANOVA models are included. Although NO did not show significance in the background normalized ANOVA because of a small sample size, it is included here since there is strong graphic evidence that it varies with distance (Figure 3). [b]Calculated as the inverse of the edge-of-road multiplier above background concentrations. [c]Calculated as the inverse of the percentage of near road high at which leveling occurs. [d]Missing values indicate no data at edge-of-road (e.g., NO), no smoothed data for estimation (e.g., metal deposition), or similar limitations.

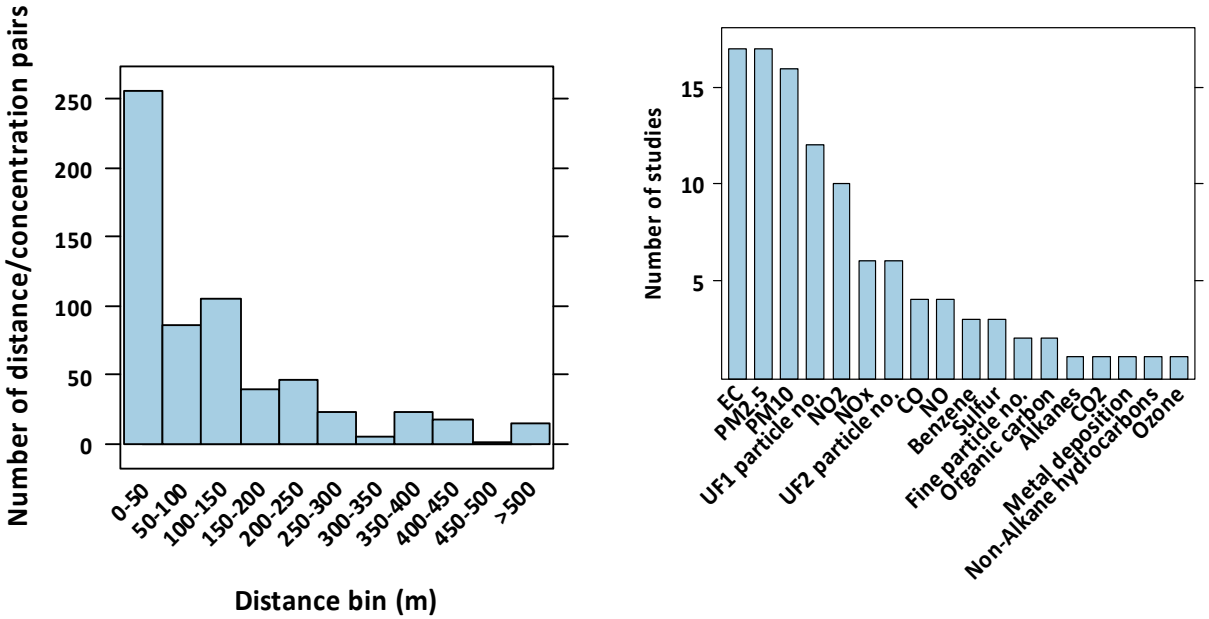
Figure Legends

Figure 1. Database summary: (a) observations grouped by 50 m distance bin and (b) studies grouped by pollutant. EC in (b) refers to direct and surrogate measures; measures of particle number (UF1, UF2, Fine) are discussed in the text. The sum of bars in (b) exceeds the 37 papers in our database—some papers studied several pollutants.

Figure 2. Local regression of background normalized concentrations on distance. The horizontal line indicates background concentration. A loess smoother (alpha = 0.75, degree = 1) is fitted to each pollutant which is placed into one of three groups. The regression sample size, n, is given in parentheses after each pollutant.

Figure 3. Local regression of edge normalized concentrations on distance. The horizontal black lines show a reduction from the edge-of-road concentration of 90% (at 0.1) and 50% (at 0.5). A loess smoother (alpha = 0.70, degree = 1) was fitted to pollutant data which was placed in one of three groups. The regression sample size, n, is given in parentheses after each pollutant. The n includes an estimated (not in the literature) edge-of-road value to facilitate normalization.

1 **Figures**



(a) **(b)**

Figure 1. Database summary: (a) observations grouped by 50 m distance bin and (b) studies grouped by pollutant. EC in (b) refers to direct and surrogate measures; measures of particle number (UF1, UF2, Fine) are discussed in the text. The sum of bars in (b) exceeds the 37 papers in our database—some papers studied several pollutants.

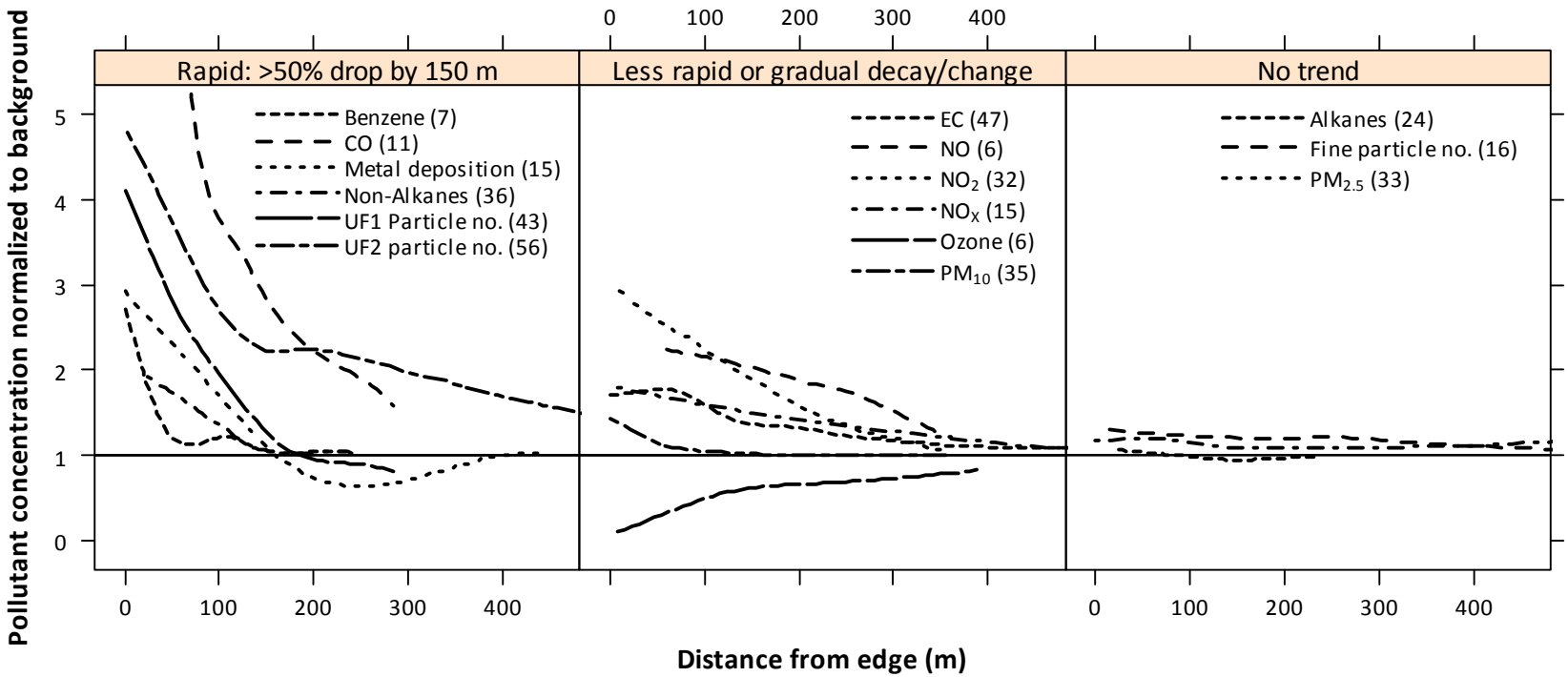
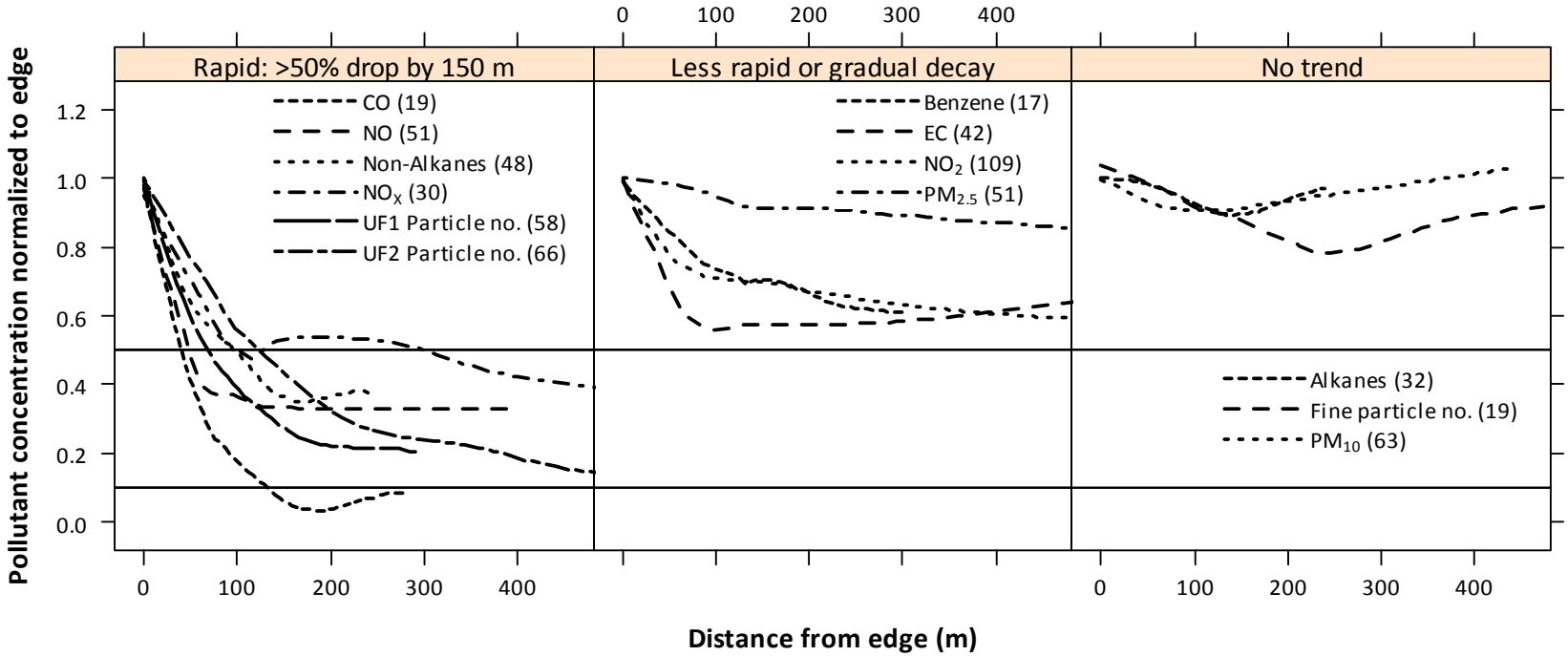


Figure 2. Local regression of background normalized concentrations on distance. The horizontal line indicates background concentration. A loess smoother (alpha = 0.75, degree = 1) is fitted to each pollutant which is placed into one of three groups. The regression sample size, n, is given in parentheses after each pollutant.



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1

Supplementary Material for "Near Roadway Air Quality: Synthesizing the Findings from Real-World Data"

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Annotated Bibliography

This annotated bibliography of the studies included in the meta-analysis is structured roughly chronologically. The chronology is broken if a particular group of studies was completed as a series, and other studies are included as necessary to provide additional context or to compare and contrast methods and results. Each description begins with the roadway name and location, and the dates that measurements were completed in underline and italics above the description of the study.

The earliest intuition that near roadway areas were areas of concern for air quality came with a group of studies in the 1960s and 70s. These were concerned with trace metal accumulation in soils and vegetation surrounding roadways. While some focused on lead (Cannon and Bowles 1962) others examined a suite of metals including cadmium, nickel, and zinc (Lagerwerff and Specht 1970), all known to be related to vehicular activity. They found increased contamination of soils near highways and intersections which decreased exponentially with distance.

An interest in the nature of air pollution concentrations in the same zone soon followed to complement the work on trace metals, emerging in the mid-1970s through work completed for CARB (Cahill et al. 1974) and the EPA. The difficulty of acquiring these early studies prevented their inclusion at the time of this writing.

Interstate 405 near Wilshire Blvd., Los Angeles, CA, 1978

Rodes and Holland (1981) sought to determine the influence of heavily travelled freeways on ozone and NO_X concentrations to determine standards for siting regional-scale pollution monitors. Determining the extent of the roadway influence was critical for siting ozone detectors. Since NO_X titrates ozone, detectors placed near roadways are not representative of

downwind exposures. These initial decisions to focus on regional scale pollution have however, left us in the current predicament where these exposures are not well classified. The authors studied the San Diego freeway in Los Angeles (200,000 veh/d) at six downwind locations (8, 32, 123, 198, 289, and 288 m from the freeway edge) and one upwind sampling location. Accounting for wind speed and direction, traffic counts and speed during, as well as ozone concentration during sampling, they characterized near road concentrations. They found that a simple exponential relationship was able to describe the decrease in NO_X concentrations as distance from the road increased and that the observed NO_X concentrations changed with high, medium and low background ozone concentrations. Gradients for NO and total NO_X were steep, decreasing by approximately 90 percent and 80 percent from their 8 m value by 123 m, respectively. Ozone levels were low and unchanged until 32 m, then increased to 123 m where leveling off occurred. The authors' final recommendations were to site NO_2 and ozone monitors at least 500 m downwind of a freeway which holds the roadway influence to less than 0.01 ppm.

Kannana-dori Ave. and Route 17, north of Tokyo, Japan, 1982 and Omekaido Ave., Western suburbs of Tokyo, 1983

Nitta et al. (1993), utilizing data from 1982 and 1983 for NO_2 and NO concentrations, attempted to quantify the effect of lightly travelled roadways (30,000 – 53,000 veh/d) in the suburbs of Tokyo, Japan on respiratory health. They also provide their emissions measurements. In general, concentrations immediately adjacent to the road were elevated above those further away. The gradient from 20 – 150 was more substantial for NO than for NO_2, but no background concentrations or meteorology data were given. Few studies are available which include data from the mid- to late-80s.

Arnhem and Wageningen, the Netherlands, 1994

In the Netherlands, Janssen et al. (1997) conducted roadside measurements on lightly (8,900 veh/d) and moderately (15,000 veh/d) traveled roadways in the cities of Wageningen (pop. 35,000) and Arnhem (pop. 130,000). One data point located 0.5 m from the edge of road was taken and compared to background levels for PM_{10}, $PM_{2.5}$, and black smoke. PM_{10} and $PM_{2.5}$ concentrations were on average 30 percent higher at the street than background while black smoke was 160 percent higher. Using elemental carbon concentrations estimated from black smoke measurements showed that the difference between street and background concentrations of PM could be attributed to elemental carbon. Since the Netherlands has a high proportion of private diesel vehicles the near road increment can be attributed largely to elemental carbon emissions from diesel fuel. A similar study was conducted in Amsterdam by Roemer and Wijnen (2001) who found that 24 hour averaged mass concentrations adjacent to the Einsteinweg (a 4-5 lane freeway) for PM_{10} and $PM_{1.0}$ were substantially less elevated above background (30 percent) than was black smoke (300 percent).

A13 Motorway, Delft and Rotterdam-Overschie, the Netherlands, 1995

Roorda-Knape et al. (1998) conducted near road monitoring as a companion to health studies by Bunekreef et al. (1997) and van Vliet et al. (1997). All were conducted in the province of South Holland. The authors sought to validate the use of distance from road as a proxy for air pollution exposure, and coupled near road measurements of $PM_{10,}$ $PM_{2.5,}$ black smoke, benzene, and NO_2, with indoor observations of PM_{10}, and NO_2 at schools that participated in the health study. The reflectance of PM_{10} filters was also measured at these schools as a surrogate for black smoke. Six residential areas were selected with high proportions of homes within 300m of roadways. For logistical reasons, only two of the sites (Delft and Rotterdam-Overschie) could

have detailed air quality monitoring completed, and both were located at different locations along the same freeway approximately 10 km from eachother. These are the two contained in the database. At Delft, sites were located at 15, 115, 165, and 305 m from the A13 motorway (average daily traffic = 120,000) while Rotterdam-Overschie had sites located at 32, 82, 133, and 260 m from the A13. More extensive measurements of NO_2 were taken to assess the variation in parallel to the road as opposed to only the horizontal gradient.

Averaging times varied by pollutant (continuous for benzene and NO_2, 15 min/hr for PM and black smoke). Black smoke and NO_2 concentrations were high close to the motorway and declined with distance, showing a curvilinear gradient which was more pronounced during periods of high exposure (> 33 percent of time downwind). No such gradient was found for PM_{10}, $PM_{2.5}$ or benzene. Indoor concentrations of black smoke and NO_2 in schools were less variable across different rooms in the same schools than were PM_{10} concentrations. Black smoke was correlated with truck traffic fraction and percentage of time downwind while NO_2 was correlated with traffic intensity, percentage of time downwind, and distance of the school from the freeway. The authors did not measure background concentrations in any of the cases, so no inferences can be made about the increments caused by the road above this level.

A38 Motorway, Birmingham, United Kingdom, 1996-1997

Comparable results were obtained in the United Kingdom by Shi et al. (1999) who examined the A38 motorway (30,000 veh/d) in Birmingham (pop. 10^6). A goal of the study was to determine the comparability of particle size distributions measured with an electrical low pressure impactor and an SMPS. They conducted measurements at the roadside (2 m from the edge), 32 m from the edge, and at two rooftop urban background sites. They found number

concentrations of ultrafine particles (in the size range 9.6 -352 nm) elevated nine times above background levels, while PM_{10} was elevated only by a factor of 1.5.

A38 Motorway, Birmingham, United Kingdom, 2000-2001

Harrison et al. (2003) attempted to quantify the contribution of various sources to the generation of roadside PM through analysis of the size distributions and chemical compositions of the particles. As lead has been phased out of gasoline, there has been a desire to identify other compounds which may be used as tracers of the contribution of road traffic.

Data was only taken at one distance from the road, and background concentrations were not measured as part of the study, but some data was used from a background monitoring site for comparison and was recorded in our database. PM_{10} was measured and is recorded in our database, but other PM size fractions included 2, 1, 0.2 and were not recorded. Polycyclic aromatic hydrocarbons (PAH) of various types were also measured but were not recorded since only a single point was taken, and no background measurements accompany the observations.

Particle number (measured using a CPC) and NO_X were used as indicators of traffic-related pollution. Particle number concentration was highly correlated with traffic, showing a similar diurnal pattern, and NO_X had been used similarly in previous work by the authors. Additionally, CPC and NO_X data were strongly correlated among themselves ($r = 0.84$). Other metals showing strong correlations with CPC number were copper, molybdenum, barium, lead, and iron. Subsequent regression analyses and a principal components analysis led to inconclusive results; however, smaller R^2's were noticed for many of the regressions (metals as the dependent variable, PM and CPC or NO_X as the independent variables) as PM size fraction modeled decreased. This indicates a potentially greater contribution to trace metals in the roadside environment from resuspension processes than vehicle exhaust. Of the PAHs measured,

methylphenanthrenes were most highly correlated with the traffic-related indicators, which are known to be markers of diesel exhaust.

I-80 Westbound near Davis, CA and Florin Rd. & Stockton Blvd., Sacramento, CA, 1994

The likely effect of roadway projects on the generation of PM_{10} "hotspots" —areas where air quality is particularly poor due to the contribution of specific sources in northern California was investigated by Cahill et al. (1994). They simultaneously measured traffic counts, meteorology, and pollution concentrations at both upwind and downwind locations for a major freeway and an urban intersection. A "sliding box" model was used to generate emissions factors per vehicle mile of travel at the three sites. An additional goal was a determination of the suitability of EPA's standard AP-42 PM_{10} emission factors for use on roadways in California. They determined that the intersection location had much higher potential to become a hotspot for PM_{10} generation than the freeway, due to increased amounts of material on the roadway surface. The AP-42 factors were found to overestimate the measured values. Experimental difficulties plagued the urban site, with measured mass from the particulate filters unable to reconcile with the aerosol composition. A follow up study was thus conducted by Ashbaugh et al. (1996). They generally found lower emissions factors than the previous study for the intersection location, and that its effects did not extend a great distance downwind with elevated PM_{10} concentrations falling to nearly background less than 100 m downwind. $PM_{2.5}$ concentrations showed no elevation near the intersection nor did they vary diurnally.

Sunrise Blvd. & Greenback Ln., Sacramento, CA, 1997

A final report in this series, by Ashbaugh et al. (1998) examined the effect of winter weather conditions on hot spot generation at a heavily trafficked intersection (average 4710

veh/hr during study). The results indicated that even during periods of near-zero wind, ambient air quality standards for PM_{10} and $PM_{2.5}$ were not exceeded near the intersection. Although measurements were taken at several distances from the road, the distances were not stated. A primary goal of the study was to quantify mixing behavior at various heights above the intersection, to see if hot spots would develop there. Accordingly, measurements were taken at 1, 3, and 9 m above the intersection.

Southeast Freeway, Bisbane, Australia, 1996

A study in Brisbane, Australia measured the horizontal profile of particle size distributions measured using an SMPS at two locations adjacent to the same freeway (Morawska et al. 1999). “Site 1” was influenced primarily by freeway traffic, while “Site 2” was positioned near the confluence of many secondary roads and an interchange to the freeway. Vertical concentration profiles at three office buildings were also taken. In general at Site 1, total particle number concentrations (in the size range 16 – 626 nm, measured by the SMPS) were elevated 10 m from the freeway at Site 1, but showed no trend from 60 – 210 m. Site 2 showed an increased difference between the site closest to the freeway (27 m) and the others (43 and 73 m) relative to Site 1, but still no decreasing trend after the first site. There was a steep cliff at the end of Site 2, however, which could have influenced the results. The presence of an interchange also likely contributed to the increased particle concentrations observed at this site. No traffic volume or truck fraction data were given for the freeway. Analysis of the size distributions of particles indicated the same source both closest to and farthest from the freeway at both sites.

Gateway Motorway, Tingalpa, Queensland, Australia, dates not specified

A later study by the same group also investigated particle concentration and size distribution near a freeway in Tingalpa, Australia (Hitchins et al. 2000). Two size ranges of particle concentrations were measured, one using an SMPS (15 – 697 nm) and the other using an aerodynamic particle sizer (500 – 20,000 nm). $PM_{2.5}$ concentrations were also estimated using a DustTrak aerosol monitor. With wind from the road towards the monitoring stations, at a distance of 150 m, total particle number concentration reached 50 percent of its near road (15 m) value. Under parallel winds, the decrease to half occurred sooner, within 50 – 100 m. $PM_{2.5}$ mass concentration trended similarly, but only decreased about 25 percent over the entire measured distance (approximately 400 m).

No background concentrations were measured as part of the study, but observations were presented for periods where wind was towards the road and thus observations showed no roadway influence. These were recorded in our database as background values, but were not given for all wind conditions and may overestimate elevations above background when combined with the measurements for low wind speeds.

Nijmegen, the Netherlands, 1999

As part of a measurement campaign monitoring on-road PM mass and number concentrations using a mobile measurement unit, Weijers et al. (2004) performed a series of near road measurements. During one day of their study, the measurement unit was driven away from unspecified heavily travelled roadways (> 10,000 veh/d) and onto a quiet side street where it sampled for 30 seconds. Measurements were obtained for PM_1, $PM_{>1}$, and particle number concentration. Particle number concentration and $PM_{>1}$ (as total suspended particulates) were recorded in our database. The measured values from “residential areas” in Nijmegen were used as the background concentration. While the results showed concentration gradients near the

roadway of similar shape for all PM mass measurements and particle number, the increment above background was far lower for PM mass than for particle number. The concentration of $PM_{>1}$ measured at 10 m from the road was 20 percent greater than background, while the ultrafine particle number was elevated 360 percent above background at 13 m from the road. $PM_{>1}$ decreased by 90 percent by 75 m, PM_1 decreased 90 percent by 120 m, and the distance corresponding to a 90 percent drop in particle numbers was 210 m.

VT5 Motorway and Savilahdentie, Kuopio, Finland, 1999

Tiitta et al. (2002) were motivated by a desire to properly site air quality monitoring stations and to produce data capable of validating roadside dispersion models for $PM_{2.5}$. The monitoring location for their study was located directly adjacent to an arterial road (Savilahdentie) but in close proximity (~500 m, p. 4058) to a freeway (VT5 motorway). Traffic ranges provided capture the range in flows expected on both road (low corresponds to arterial, high to freeway). Real-time traffic and meteorological (wind speed/direction, relative humidity, temperature) data were taken along with PM readings.

Noting that $PM_{2.5}$ sources are varied, their model of total $PM_{2.5}$ was broken into six separate terms, comprising both mobile (exhaust, non-exhaust) and stationary sources, urban and long-range transported background concentrations and resuspension processes not related to traffic flow. Mobile sources were further disaggregated by vehicle/fuel type and observational data was used as model input. The observed vehicular emissions were used then used to calibrate their model using multiple regressions. Additional data was used for the long-range transported and stationary components. Long range transport, vehicular (primary and non-exhaust) emissions, and other, accounted for 41, 33, and 26 percent of observed $PM_{2.5}$ concentrations,

although an application of the model to an independent dataset would be required for verification purposes.

The collected data was reported as well: traffic emissions caused an increase in $PM_{2.5}$ concentrations of 30 percent from the furthest to the closest distances from road. This agreed well with model predictions (33 ± 6 percent).

Hunts Point, Bronx, New York, NY, 1999

Lena et al. (2002) conducted measurements of $PM_{2.5}$ and elemental carbon at six intersections in the Hunts Point region of the Bronx which is heavily impacted by diesel truck traffic. (The sixth location an enclosed garden and will be reported as the background concentration on each of the intersection measurements). Wind direction and meteorology were not stated in the paper. Traffic volumes reported (both for cars and trucks) showed variability. Results reported here are representative means. They determined a relationship between truck density and elemental carbon concentrations ($R^2 = 0.84$) but that between truck density and $PM_{2.5}$ was much weaker ($R^2 = 0.52$). This highlights the importance of mobile sources to elemental carbon and regional sources for $PM_{2.5}$.

Avenida da Amizade Road, Rua do Campo Road, Rua de Ferreira do Amaral Road, and Rua da Ribeira do Patane Road, Macao, China, 1999

Wu et al. (2003) investigated the chemical composition of PM_{10} and $PM_{2.5}$, providing point-roadside data was given and was recorded in our database. Distances from road were only reported as 1 – 2 m, therefore, 1.5 m was recorded in our database as a point estimate.

Organic carbon was determined to be the largest component of both roadside and background concentrations of PM. However, the study did not attempt to account for sampling artifacts (adsorption of gaseous organic compounds, or evaporative loss of volatile organic

compounds) so the organic carbon data was not recorded in our database and is not necessarily reliable. Crustal materials exhibited high concentrations both near road and at background, indicating a low traffic-related component. Elemental carbon was a major near road component and was found to be higher next to roads with a higher proportion of diesel traffic, supporting its use as a diesel tracer. Both organic carbon and elemental carbon were reported for PM_{10} and $PM_{2.5.}$ Elemental carbon is known to reside mostly in the submicrometer range, and background values were given for both, so there should be no issue with normalization.

Avenue Leclerc & Place Basch, Paris, France, 2001

Vardoulakis et al. (2005) conducted this study in a highly urbanized area of Paris, France, with a mix of traditional six-storey buildings, modern high rises, and some detached homes. A week of "intensive" sampling was completed comprising vertical measurements taken along the sides of buildings, in addition to the roadside monitors at the study intersection (66,000 veh/d). Trees were found to disrupt the transport of benzene upwards beyond the second floor of apartment buildings.

Ten roadside monitors were placed along Avenue Leclerc to detect small spatial scale variation in pollution concentrations. Each monitor was placed exactly at the roadside or within the traffic stream (on medians). Strong correlations were found between all measured volatile organic compounds (VOCs) and CO, and regression analysis was used to model benzene concentrations based on CO. The authors found that wind speed and direction had a large effect on observed concentrations—higher concentrations of pollution were observed on the leeward side of the street. The highest benzene concentrations were observed in October during low-wind speeds, and the lowest in August due to low traffic density, again illustrating the complex interplay between the factors contributing to observed concentrations. Overall seasonal trends

were not visible. High variability was found due to differences in urban form and meteorology, leading to the realization that the siting of monitoring equipment is crucial. Measurements conducted where the intake of undiluted exhaust gas is possible are not representative of exposure. Dispersion models were tested for consistency with observations and were found to overestimate concentrations in all cases but this could be due to the area's complex geometry.

Concentration results were only presented for benzene. Other data was used to populate correlation matrices, to complete linear regressions, or was presented in high time resolution, making the extraction of an average for database input excessively difficult.

Interstates 580 and 880, State route 92, East Bay, San Francisco, CA, 2001-2002

The East Bay Children's Health Study was designed to assess the links between exposure to traffic related exhaust and respiratory illness in children attending 10 schools in the East Bay of the San Francisco Bay Area, which has generally good regional air quality. The schools were selected to represent locations upwind and varying distances downwind from the freeways under study, and neighborhood measurements were taken at three of the schools to assess outdoor concentrations at childrens' residences. Residences were found to be good indicators of school exposure for those upwind or far downwind, but near downwind homes underestimated or overestimated on the basis of distance from road.

Responses to questionnaires (n = 1,109) on health outcomes (asthma, bronchitis, and chest illness) were modeled via logistic regression using air pollution (PM_{10}, $PM_{2.5,}$ black carbon, NO_X, $NO_{2,}$ NO) monitoring data taken at each of the schools as independent variables. Since most students resided in the surrounding neighborhoods, school exposure was taken as a surrogate for personal exposure.

Kim et al. (2004) reported the epidemiological results. In general, NO_X, NO, and black carbon showed stronger associations with respiratory illness, gauged by increasing odds ratios, than PM_{10}, $PM_{2.5,}$ and NO_2. NO_X, NO, and black carbon were also highly correlated. An additional model using distance downwind from a major road instead of pollution concentrations showed similar or stronger odds ratios, indicating that the roadway is a key source of exposure to these pollutants.

In general black carbon, NO_X, NO, and NO2 to a lesser extent increased in concentration as the schools decreased in distance to a major roadway (either I-580 or I-880) within the range of 300 m. The authors reported concentrations taken at these schools and averaged over periods of tens of weeks. Reported concentration values therefore do not all originate from the same location on the same freeway. Each school was identified as being close to a particular freeway and thus was exposed to the annual average daily traffic associated with that freeway. Schools upwind or far downwind (schools 1, 3, 4, 6, and 7 from the study) did not have statistically distinguishable differences in pollutant concentrations. Background concentration values reported in the paper therefore reflect average for schools greater than 300 m from a major traffic source. Singer et al. (2004) showed that concentrations of NO_X and NO_2 measured at those schools were similar to those taken at the closest stationary monitoring station—within 350 m upwind, concentrations were similar to regional monitoring station data and from schools located >1000 m downwind of the freeway.

Singer et al. (2004) took hourly wind speed, direction, and temperature were taken primarily to validate classifications by wind direction. Sites to the east or northeast of freeways were predominantly downwind from traffic emissions although wind patterns shifted during the winter months. Near road effects were somewhat higher in spring when the school was directly

downwind of the freeway for almost the entire day and somewhat lower in fall, when nighttime winds shifted.

E4 Motorway, 28 km north of Stockholm, Sweden, 2003

Gidhagen et al. (2004) attempted to quantify the geographical and temporal variations of ultrafines to better model exposure levels for urban populations. By investigating a rural highway they expected low particle surface area (rapid dilution) and high vehicle speeds. These conditions favor the generation of ultrafines. The effect of temperature was also investigated, and aerosol dynamics were explicitly accounted for. The authors performed near road monitoring at 10 m and 63 m from the E4 motorway (52,300 ADT). The results showed that total NO_X and total particle number were highly correlated while the ratio of particle number to NO_X decreased as temperature increased.

Results suggest that, for assessing urban exposure, deriving emissions factors from roadside monitors between 10 – 15 m from the edge of road would be adequate data for Gaussian line modeling to assess exposure to total particles.

Interstates 405 and 710, Los Angeles, CA, 2001-2005

A series of studies conducted by researchers at UCLA and USC became heavily cited in the subsequent study of pollution concentrations near heavily travelled roadways. Zhu et al. (2002b) examined CO, black carbon, and particle mass and number concentrations at five locations downwind of the I-405 freeway (13,900 veh/hr during peak, <5 percent trucks) in Los Angeles, California. They found that with increasing distance, ultrafine particle size distributions shifted to greater diameters and number concentrations dropped dramatically. Number concentration decreased to approximately half of its value at 30 m between 90 m and 150 m and

reached background by 300 m. CO and black carbon tracked particle number well, leveling off by 150 m from the freeway.

Zhu et al. (2002a) conducted similar measurements at the I-710 freeway which has a large proportion of diesel truck traffic since it functions as a goods movement corridor for the ports of Los Angeles and Long Beach (12,000 veh/hr during peak, 25 percent trucks). Total particle number concentration (6 – 25 nm) decreased 80 percent in first 100 m before leveling off somewhat after 150 m and reaching background at 300 m.

Particle number, black carbon, and CO all exhibited a similar exponential decay pattern at both the 405 and 710 freeways while absolute concentrations of black carbon and CO differed, as would be expected given the difference in fleet composition on each section. I-405 generally had twice the CO concentration and a third of the black carbon concentration as I-710.

An identical study was carried out by Zhu et al. (2004) during the winter of 2002 at the same freeway locations and same distances. They found a distinct mode in the 20 – 30 nm size range for the 710.

To examine the effects of meteorology on observed concentrations, Zhu et al. (2006) conducted air quality measurements over seven nighttime periods (11pm – 4am) and compared them to their previous results from the I-405 freeway in Los Angeles (Zhu et al. 2002b). This study represents an important contribution to the near road literature since it explicitly recognizes the importance of meteorology. Particle number concentrations decreased more slowly during the night than during the day, to reach 40 percent of their original value at 30 m by 300 m, but even by 500 m the background level had not been reached. This implies that the nighttime contribution from freeways to local air quality is more substantial than during the day because of increased local traffic during this time (the day) which raises background levels of pollution.

Although traffic volume was only 25 percent of the daytime value, particle number concentrations were approximately 80 percent of their daytime value. This was the contribution of meteorology. Lower temperatures correspond to higher rates of particle production per vehicle. Speeds are also higher at night, which is proportional to particle number concentrations. There were more trucks at night than during the day. However, the most important factor appeared to be the relative humidity which increases the number concentration of the nucleation mode. None or very weak PM_{10} and $PM_{2.5}$ gradients were observed indicating that policies to limit either may be ineffective at reducing total particle number concentrations.

No trends were observed at upwind locations, indicating that using proxy indicators for traffic exposure may be inappropriate since actual AP exposure is highly dependent on wind and meteorological conditions that vary over the day and by season. Indeed, "…many epidemiology studies have relied on distance from major roads or self-reported traffic density as a proxy for exposures. It is clear … that none of these is a good measure of exposure to traffic exhaust" (Zhu et al. 2006, p. 2534).

Interstate 710, Los Angeles, CA, 2006

The I-710 was investigated again in the Winter of 2006 by Ntziachristos et al. (2007) who sought to classify the physical and chemical properties of particles near the freeway in detail including particle number, surface area and mass. The authors used a single location, 10 m from the edge of the shoulder to measure both gaseous and particulate concentrations. Size distributions were measured using a SMPS (16 – 638 nm) and particles were counted with a CPC (>6 nm). Gaseous pollutants were also measured.

They found that particle number concentration near the freeway was overwhelmingly in the less than 16 nm size range. This comports with the results of Zhu et al. (2004) and results

from a comparison of the SMPS and CPC numbers. Further, particle size distribution was bimodal, peaking in both the nucleation and accumulation modes which is indicative of roadways dominated by diesel vehicles.

Particle emissions dropped substantially on a day when the freeway was closed to traffic, but organic carbon concentrations did not drop as much nor did they correspond as closely to traffic volumes as elemental carbon and black carbon, indicating a secondary source and secondary formation mechanisms. All measures of particle intensity except one peaked at 4 pm when traffic was congested, strengthening the link between traffic and particle emissions. CPC number increased with time of day (11 am - 7 pm) because of the nucleation of small particles as temperatures dropped.

A further study goal was to investigate the appropriateness of using the dilution ratio (which compares the CO_2 ratio in exhaust gas to that at the sampling site) to decouple the influence of sampling site location, meteorology and fleet effects on measured values. This was accomplished by a comparison to two other studies.

Regression modeling was also undertaken, using surface area concentration and the particle volume in the nucleation and accumulation modes. Light and heavy organic carbon were associated with the nucleation mode and accumulation mode volume, respectively supporting previous evidence of a unique particle mode adjacent I-710 in the size range 20 – 30 nm. Further, predominantly gasoline fueled vehicles have shown an organic carbon profile shifted towards higher mass.

The deposition and composition of trace metals (including nickel, copper, chromium, lead, and zinc) in the vicinity of both freeways was also studied at approximately the same time. Sabin et al. (2006) investigated the chemical composition of particles deposited at three distances

from the Interstate 405, and one background location in 2003. The freeway appeared to be a source of particles greater than 6 µm in diameter. Deposition of metals reached background quickly, somewhere between 10 and 150 m. A simple modeling effort based on Gaussian plume dispersion showed good agreement with empirical results. The study was limited by the small sample size.

State Route 110, Los Angeles, CA, 2005

Kuhn et al. (2005a) and Kuhn et al. (2005b) studied State Route 110, a freeway with heavy duty truck traffic prohibited in the summer of 2004 and winter of 2005, respectively. The studies were motivated by the knowledge that the organic constituents of PM emitted from vehicles are semivolatile and that observed concentrations and size distributions can thus be affected by meteorology as well as the chemical properties of the compounds. The authors sought to characterize how size distributions shift due to volatility and to draw conclusions for exposure to PM. Two locations adjacent to the roadway were used in each paper (results were reported at 2.5 and 150 m) and the chemical composition and mass of particles in the coarse (2.5 – 10 µm), accumulation (0.18 – 2.5 µm), and ultrafine (< 0.18 µm) modes were measured between 12 pm and 7 pm on weekdays.

The results showed that higher wind speeds and drier conditions in the summer led to higher mass concentrations in the coarse mode than in winter. The accumulation mode masses were similar at both measurement locations, indicating that that size fraction represented an urban background aerosol. Concentrations broken out by species showed little variation between the two locations in winter except for organic carbon, elemental carbon, Cu, and Ba. However, organic carbon was affected by a sampling artifact in which gaseous organic material was absorbed by the measurement filter. In general, concentrations of all species were higher in the

summer except for organic carbon and elemental carbon which are known to track traffic emissions. They are thus affected by atmospheric inversion caused by colder temperatures.

Observed concentrations did not include a background value, and so are not extensive enough to determine when that value is reached. Recorded in our database were values of PM_{10}, $PM_{2.5}$, elemental carbon, and sulfur.

Interstate 80 near Sacramento, CA, 2000-2001

Nanzetta and Holmen (2004) also attempted to quantify the effects of meteorology and traffic volume on observed particle number concentrations. They investigated a section of I-80 near Sacramento, California (6330 veh/h during the study period with 10 percent heavy trucks) in both fall and summer months. A single monitor was placed downwind at 3 m from the edge of road and an upwind monitor was placed at 6 m from the roadway edge in winter and 9 m from the edge in summer. Ratios between downwind and background observations ranged from 14.6 in winter to 1.3 in summer in the size range 6 – 237 nm, and 6.5 in winter to 2.1 in summer in the size range 23 – 898 nm. These differences are due largely to temperature and relative humidity, since nucleation of new particles decreases as temperatures increase and relative humidity decreases.

Statistically significant relationships between observed concentrations and roadway parameters and meteorology were difficult to ascertain in many cases. While relationships were found with heavy duty vehicles and particle numbers in summer, no relationship with any vehicle types was found in winter. Temperature, relative humidity, and solar radiation were significantly correlated with upwind concentrations, while only temperature and wind speed showed significant correlations with downwind concentrations. Their study again highlights the

importance of daily and seasonal trends in assessing observed pollutant concentrations and points to a need for better time resolved traffic and concentration measurements.

Interstates 71 and 75, Cincinnati, OH, dates not specified

Reponen et al. (2003) studied the I-71 and I-75 freeways (135,000-150,000 veh/d, 10 percent trucks) in Cincinnati, Ohio, measuring particle number, elemental and organic carbon, and $PM_{2.5}$ mass concentrations. The measurements were conducted in support of a larger epidemiological effort, the Cincinnati Childhood Allergy and Air Pollution Study. Condensation nucleus counters, measuring in the size range 0.02-1 µm, showed a steep gradient in ultrafine particle concentrations when moving away from the freeway. Concentrations decreased by 50 percent between 50 and 150 m. Background concentrations were measured at 1600 m downwind from the freeway and were reached by 800 m. A minimal gradient was observed for coarse particles as measured by an optical particle counter (size range 0.3-20 µm). In general, however, "traffic particles could not be distinguished from the background levels beyond 400 m from the highway" (p. 562).

Exposure to traffic pollution was detected at distances up to 400 m using sulfur (believed to be a surrogate for diesel exhaust) as an indicator although results were somewhat inconsistent (and not discussed) between sampling periods. Elemental carbon, another indicator of diesel exhaust, exhibited elevated concentrations up to 400 m downwind as well.

Stockholm County, Sweden; Munich, Germany; and Rotterdam, the Netherlands, 1999

Hoek et al. (2002) sought to quantify the average annual concentration and spatial variability of $PM_{2.5}$ in Munich, Stockholm and the Netherlands. Approximately 40 sites were selected in each area and were broken down into background (urban/rural) and traffic. Urban

background was further disaggregated by proximity to city center. Results are reported as means and ranges for each type of site.

The authors found more spatial variability for $PM_{2.5}$ than previous literature (17 to 18 percent higher at traffic sites than at urban background locations), but the $PM_{2.5}$ absorption coefficient exhibited even larger variability (31 to 55 percent increase at traffic sites versus urban background concentrations), supporting previous findings suggesting that measuring only $PM_{2.5}$ (or PM_{10}) concentrations as a traffic-related air pollution indicator can underestimate the contrast between background and traffic-related emissions exposure. Detailed data on distance from road was not given, but mean distance from road for all sampling sites within each of the three areas ranged from 112 to 139 m. In Stockholm and the Netherlands the maximum distance from road was 450 m while for Munich it was 930 m.

The sampling approach utilized four two week mean samples at each site taken over a period of a year and calibrated using a monitor in continuous operation at one particular site in the study area. The authors suggest that similar approaches could be used in other cities to assess spatial variations in pollution that could contribute to differential epidemiological risks.

Using similar methods, a later paper by the same group (Cyrys et al. 2003) investigated correlations between annual averages of elemental carbon, $PM_{2.5}$, PM_{10}, and PM absorption coefficients in the three study areas to determine the extent to which absorption coefficient of PM filters can be used as a surrogate for elemental carbon concentrations. They conducted direct measurements of elemental carbon using a two part process whereby organic carbon is first removed by thermal desorption and the remaining elemental carbon is oxidized to CO_2 and water. This is comparatively more laborious than the much simpler method of measuring filter absorbance using a single instrument which determines how reflective the filter on which PM is

collected is used. They found that contrasts between traffic and background were greater for elemental carbon and $PM_{2.5}$ absorption coefficients than PM mass concentrations, and that the $PM_{2.5}$ absorption coefficient better tracks spatial variability in combustion derived particles than $PM_{2.5}$. On net, these findings support the use of PM filter absorbance as an inexpensive surrogate for elemental carbon concentrations.

Highway 15, Montreal, Quebec, Canada, 2001-2003

A pair of studies conducted by Gilbert and colleagues at Health Canada and Environment Canada (Gilbert et al. 2005; Gilbert et al. 2003) sought to characterize the exposure profile of NO_2 adjacent roadways in Montreal, Quebec.

Gilbert et al. (2003) conducted measurements at the Decarie Highway in Montreal (185,000 veh/day) over a week in September, 2001. They found that the major NO_2 decrease occurred within 200m of the highway. An estimated regression model including the logarithm of distance from roadway and a binary variable for upwind/downwind location had an R^2 of 0.97. Downwind concentrations were significantly greater than upwind concentrations at similar distances, although no clear “background” value was provided so it is not clear when or whether NO_2 concentrations became uninfluenced by contributions from traffic. This is certainly different than the findings showing a precipitous drop of particle number concentrations near freeways and no influence in the upwind direction. Here the decline was much more gradual at increasing distances and almost even between upwind/downwind samplers. This could have been due to the temporal nature of the study (values averaged over 7 days as opposed to just peak periods). The authors conclude that distance from a major road may adequately characterize exposure to NO_2.

Gilbert et al. (2005) developed a multiple regression model to predict NO_2 concentrations based on land use characteristics. The results of 67 monitoring locations were included in the

analysis. The final model included positive associations with traffic count on the nearest highway, length of highways within 100 m, the length of major roads within 100 m, length of minor roads within 500 m, population density within 2000 m, and negatively associated with the area of open space within 100 m and distance from the nearest highway. Final model R^2 was 0.545. The authors suspect that higher values would have been obtained if wind speed and industrial sources were better accounted for.

Metropolitan Highway and various urban arterials, Montreal, Quebec, Canada, dates not specified

Smargiassi et al. (2005) also conducted near road measurements in Montreal and sought to assess the spatial and temporal variability of elemental carbon (measured using the $PM_{2.5}$ absorption coefficient) , $PM_{2.5}$, and NO_2. They also assessed the effect of traffic density on the observed concentrations. A primary study goal was to draw conclusions regarding the exposure of individuals residing adjacent to urban roads to particulates.

Four sites were chosen with differing traffic intensity but free from stationary sources of pollution. The first represented background concentrations—a quiet residential street. Two and three were on urban arterials, while four was adjacent to the Metropolitan Highway. Sites were separated by approximately 1 km, would therefore show an influence only from their closest roadway. Thus, this study, instead of moving further from roads, held distance from road constant and increased the traffic volumes. All three traffic sites were located “less than 10 m from the curb.”

A gradient was observed with increasing traffic volume for $PM_{2.5}$ absorbance and NO_2. $PM_{2.5}$ was approximately equal across all sites, supporting the regional nature of $PM_{2.5}$

emissions. Although results showed spatial gradients in the urban area studied, the authors were unable to draw definitive conclusions about personal exposure.

M50 Motorway, Dublin, Ireland, 2003

A pair of studies published by colleagues at Trinity College in Dublin, Ireland, examined the concentrations of 11 hydrocarbons at three distances downwind and upwind (six receptors total, each at 25, 120, and 240 m from the edge of road on both sides) of the primary beltway in Dublin, Ireland, the M50. The data collection and results are described in O'Donoghue and Broderick (2007) and the data is compared to dispersion modeling using CALINE4 and COPERT III in Broderick and O'Donoghue (2007). The studies were motivated by a lack of near road hydrocarbon measurements previously reported in the literature.

Consistent meteorology in terms of temperature and wind speeds persisted over the sampling campaign, which lasted five weeks and took place during the morning peak period (7 am – 10 am). Small modulations in wind speed (0.85 m/s average when from the east versus 1.28 m/s from the west) were enough to change the rate of decay to background concentrations for the non-alkenes monitored (*iso*-pentane, *n*-nentane, ethane, propene, 1,3-butdiene, acetylene, and benzene). By 240 m from the road under all wind conditions, background concentrations were close to being reached. For the lower wind speeds, between 20 and 35 percent of the concentration at the nearest monitor remained by 240 m. If an edge concentration would have been interpolated, however, this number would have been smaller. Monitored alkenes (ethane, propane, *iso*-butane, and *n*-butane) did not vary with distance from road.

Benzene was grouped with other measurements of that pollutant into its own category, while the other measurements were grouped together as alkanes or non-alkane hydrocarbons in

our database, since no other studies overlap with the non-Benzene compounds measured by these researchers.

Interstate-440, Raleigh, NC, 2006

Hagler et al. (2008) investigated the relationship between traffic and ultrafine particle number concentrations. They found variability in terms of absolute particle number measurements, but a similar pattern of decay on each observation day. They noted that benzene, toluene, NO, CO, black carbon correlated with the decay of ultrafines, but that secondary pollutants did not correlate (PM_{10}, $PM_{2.5}$, NO_2). They did not quantitatively assess the influence of each of the parameters on observed concentrations.

Highway 71, Interstate 35, FM-973, Austin, TX, 2007

Clements et al. (2008) and Zhu et al. (2008) performed an extensive measurement campaign adjacent to Highway 71 (arterial), Interstate 35 (freeway), and a truck traffic-dominated freeway (FM-973). A specific study goal was to determine how the traffic composition and freeway design affects observed concentrations. Data collection methods mixed stationary monitoring units with a mobile platform that stopped at fixed distances from the roads.

Clements et al. (2008) reported those measurements from the arterial. In addition to ultrafine particles (< 100 nm), criteria pollutants (CO, NO_X, $PM_{2.5}$, VOCs) and several other organic compounds were measured—polyaromatic hydrocarbons, alkenes, hopanes, and several carbonyls. Traffic-related pollution generally decayed to background concentrations by 150 m. Some complex relationships with distance were noted for specific carbonyls (Acetaldehyde) and alkenes (C28) which increased with distance. These were not recorded in our database.

Suggested reasons include chemical reaction with other exhaust species. Other alkenes and carbonyls showed the expected decreasing relationship.

Zhu et al. (2008) reported the ultrafine particle number results observed at Highway 71, and the normalized results for all three roadways. The data were normalized to unit traffic intensity and windspeed, which is only possible if the primary data are accessible. Their results showed good agreement between roads when normalized ultrafine particles were plotted with distance, strengthening the importance of traffic intensity and windspeed as factors affecting observed near road concentrations.

Data Reduction

To ensure that observations were considered together with pollutants they were meant to represent, we performed several grouping operations to collapse the data, described below.

Elemental carbon can be measured by many different means. Cyrys et al. (2003) notes the importance of this pollutant as a surrogate for diesel particulate matter (DPM). They noted further that the terms black carbon and soot are used interchangeably with elemental carbon, but that the two pollutants have somewhat different behavior. Nevertheless, we too grouped elemental and black carbon together. Direct measurement of elemental carbon is complex, but the reflectance of particulate filters is correlated with elemental carbon, regardless of the upper size cut of the filter used (whether reflectance is measured from PM_{10} or $PM_{2.5}$ filters, the two recorded in our database) since elemental carbon resides mostly in the submicron range (Fischer et al. 2000). Measurements of black smoke also are correlated with elemental carbon. We combine all of these indicators together, labeled as “elemental carbon” in figures but frequently referred to as “elemental carbon and surrogates” in the text.

Total suspended particles (TSP) are converted to PM_{10} using a factor of 0.55 to estimate PM_{10} concentrations following Levy et al. (2000).

Alkanes (ethane, propane, *n*-butane, and *iso*-butane) and non-Alkane hydrocarbons (ethane, acetylene, *iso*-pentane, benzene, propene, *n*-pentane, and 1,3-butadiene) were studied by O'Donoghue and Broderick (2007) who noted that those groupings tended to behave similarly—alkanes did not vary substantially with distance, while the non-alkanes did. We extract benzene from the non-alkanes to place it into its own group, but keep the others grouped together since they were the only measurements taken of those pollutants. If future studies produce more of these measures, they can easily be split out into their own groups.

The term "ultrafine" typically refers to particles less than 100 nm in diameter (Donaldson et al. 2001), and particle number concentrations (as opposed to mass concentrations) are typically used to quantify ultrafine pollution. Detectors used for counting particle number varied among scanning mobility particle sizers (SMPS) fitted with condensation particle counters (CPC), stand-alone CPCs, condensation nucleus counters, and optical particle counters. The size range measured by each can be set by the analyst, and since the number concentration tends to increase depending on the low end of the measured range (Molnar et al. 2002),[1] we grouped studies into three categories according to that value.

We considered "UF1 particle number" to begin at 3 nm, "UF2 particle number" included size ranges beginning at 15 nm, supported by findings that size distributions typically achieve maxima in the range greater than 15 nm (Molnar et al. 2002; Nanzetta and Holmen 2004). The final category, entitled "Fine particle number" included studies which counted particles beginning in the 300 nm size range (~ 0.3 µm, just above the ultrafine classification). The size

[1] For example, Shi et al. (1999) found comparatively little variation for particle number in the size range greater than 72 nm when compared to that greater than 30 nm, and Nanzetta and Holmen (2004) found higher concentrations in the range from 6 – 237 nm than 23 – 898 nm.

range measured for each study that analyzed particle number concentrations is shown in Table S-1; many studies measured multiple size ranges. While the SMPS can measure size distributions as well, these data are not suitable for our database, since the output is not a single measurement at a distance from the road, but rather a distribution.

TABLE S-1. Particle Size Ranges Counted in Studies Which Recorded Particle Numbers

Particle Number Classification	Size Range Measured	Study
UF1	> 3 nm	Gidhagen et al. (2004)
UF1	> 6 nm	Ntziachristos et al. (2007)
UF1	6 – 220 nm	Zhu et al. (2002a); Zhu et al. (2002b)
UF1	6 – 237 nm	Nanzetta and Holmen (2004)
UF1	4 – 163 nm	Gramotnev and Ristovski (2004)
UF1	6 – 300 nm	Zhu et al. (2008)
UF1	> 7 nm	Weijers et al. (2004)
UF1	7.4 – 290 nm	Zhu et al. (2006)
UF1	> 8.2 nm	Kuhn et al. (2005a); Kuhn et al. (2005b)
UF1	9.6 – 352 nm	Shi et al. (1999)
UF2	15 – 697 nm	Hitchins et al. (2000)
UF2	16 – 638 nm	Ntziachristos et al. (2007)
UF2	20 – 1000 nm	Reponen et al. (2003)
UF2	23 – 898 nm	Nanzetta and Holmen (2004)
UF2	20 – 1000 nm	Hagler et al. (2008)
Fine	500 – 20,000 nm	Hitchins et al. (2000)
Fine	300 – 20,000 nm	Reponen et al. (2003)

Omitted Data

It was not possible to conduct a local regression on each pollutant type. Generally, the regression failed on those pollutants which had a small sample size or large distances between observations. Organic carbon and sulfur were omitted from the background (Figure 2) and edge normalized plots (Figure 3), due to limited data, and metal deposition was omitted from the edge normalized plot since the data was too sparse to be smoothed at the more restrictive value of the smoothing parameter used to generate that figure (0.70 as opposed to 0.75 for Figure 2).

Data from two studies was removed from the background normalized plots in the article (Figure 2). Rodes and Holland's (1981) data for oxides of nitrogen (NO, NO_2, and total NO_X) which were taken during a period with substantially different fleet characteristics than existed at the date of the majority of the studies in the database. Zhu et al.'s (2006) data for nighttime concentrations of UF1 particle number were taken under substantially different meteorological conditions than every other study included in the analysis and were also excluded from the edge normalized plots (Figure 3) to ensure that nighttime-only data were not included for analysis. To justify the exclusion of these data, consider Figure S-1, which expands upon ideas presented in the section "Limitations and Differences Between Normalization Methods" in the main article and looks at differences between background and edge normalization for NO.

Two sets of measurements with background concentrations are included in the pooled data for NO. One was completed in the late 1970s (Rodes and Holland 1981) and the other in 2004 (Kim et al. 2004). The entire set of measurements from the 1970s is high, ranging from 290 ppb at 8 m from the road, to 5 ppb at 388 m with a 1 ppb background value. The 2004 study shows a much lower roadway increment (17 ppb background with 38 ppb at 60 m from the road). In the 1970s study, concentrations were about 150 – 200 times greater than background at 8 m from the road, whereas in 2004, they are only approximately twice background. The normalization to edge appears to create a more common comparison. Normalizing by background, the mean observation from the 1970s for NO is 57 times greater than that for 2004. The same comparison for edge normalization shows that the mean observation for 2004 is 2.4 times greater than that for the 1970s (since the 1970s data shows a greater percentage decrease as it approaches background concentrations). Since the ratio of means differs by an order of magnitude between normalization methods it is more appropriate to include the Rodes and

Holland (Rodes and Holland 1981) data in the edge normalized assessment where the means are much closer in magnitude.

While the difference is not as stark, the mean background normalized UF1 particle number concentration from the nighttime Zhu et al. (Zhu et al. 2006) data for are 1.75 times greater than the other measurements in the database, and edge normalized measurements are only 1.30 times greater. In this case, the nighttime concentrations are generally higher when normalized to background and also decay to background concentrations more slowly than non-nighttime concentrations. These data were excluded from both the background and edge normalized assessments since they were taken exclusively during nighttime conditions, different from every other reviewed study.

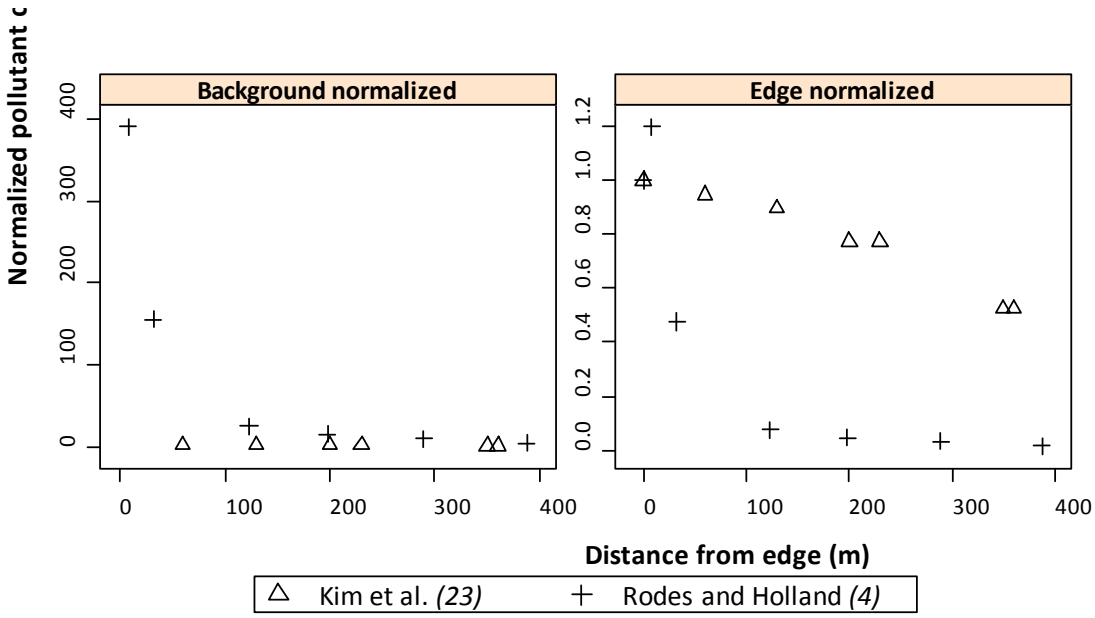

FIGURE S-1. Background normalized NO data.

If all information contained in the database were equal, only one type of normalization would be required to compare studies. Perfect information about the decay profile would constitute an edge concentration, and several measurements at a perpendicular transect, with the final measurement representing a background value. From this data, the near road multiplier

above background concentrations would be known, as would the percentage decreases with distance and the distance at which background concentrations are reached. However, many studies are missing information on background concentrations, for example, or only took concentration measurements at two points, making an exponential fit impossible. Employing both normalization methods prevents the unnecessary exclusion of some data.

Edge Normalization

Edge normalization estimated an edge-of-road concentration according to the following algorithm. If a measurement point was present in the first 3 m from the road, it was taken to represent an edge-of-road concentration. Otherwise, if there were only two data points in the set they were simply connected and their intercept with the vertical axis was used as the edge concentration. If results from the annotated bibliography showed that there was little or no relationship between pollutant concentration and distance, linear regression was used on the entire set of measurements from that series to determine the edge concentration. If three or more near road points were given and the background normalization and literature agreed that the pollutant varies with distance, an exponential decay model was utilized which varied according to distance. We fitted an exponential decay model to each set of measurements that met our criteria; the general model took the form shown in Equation 1,

$$\mathrm{C} = \beta_0 + \beta_1 e^{-\mathrm{D}/\theta} + \epsilon \qquad (1)$$

where C is the concentration value predicted based on D, distance, ϵ is a normally and independently distributed error term,[2] and β_0, β_1, and θ, are estimated model parameters whose

[2] It is likely that the error term is in fact heteroskedastic, since its variance should increases as distance decreases, corresponding to a higher influence of the variation associated with roadway activity and as the magnitude of observed concentrations increases. Since the model is being used to describe a previously identified physical relationship, and not in an exploratory manner, and since we are not assessing model performance, this should not be

initial values were estimated using an algorithm described by Venables and Ripley (2002, p.216).[3] Representing concentration as a function of distance, the algorithm proceeds as follows. A quadratic is fit to distance, and three values for concentration, C_0, C_1, C_2, are determined at equally spaced distance points: $D_0, D_1 = D_0 + \delta, D_2 = D_1 + 2\delta$. Substitution of these points into Equation 1 yields an initial value of θ, i.e. $\theta_0 = \delta / \ln\left(\frac{C_0 - C_1}{C_1 - C_2}\right)$. Ordinary least squares regression of concentration on $e^{-D/\theta}$ can then be used to determine initial estimates of β_0 and β_1. Letting the parameters vary by modeled dataset recognizes that while distance is the most important independent variable[4] the precise shape of the curve will vary according to site- and study-specific details.

Numerical methods are required for determination of the final coefficients for the nonlinear regression since the equations for their prediction are typically themselves nonlinear. R (R Development Core Team 2008) offers three algorithms for nonlinear model estimation under the function `nls()`: Gauss-Newton, Golub-Pereyra, and "NL2SOL" from the package "Port." An applied approach was taken to selection. Not all algorithms could be applied to each set of measurements—some failed to converge, or iterated indefinitely without finding acceptable parameters. Trial and error revealed that Golub-Pereyra functioned adequately most often and thus was used in the broad majority of cases while NL2SOL was able to estimate the remaining models. Two datasets from the series of non-Alkane measurements were not amenable to estimation regardless of algorithm, but showed relationships with distance upon visual examination of the data clear from road, with the nearest to road concentration high and the other

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an issue. Nevertheless, additional studies utilizing this type of normalization should examine this issue in greater detail, possibly applying a generalized model which allows the variance to be defined as a function of the predicted variable.

[3] Implemented in R as "negexp.SSival" in package MASS.

[4] Confirmed by previous results which modeled land use and traffic emissions as well as atmospheric dispersion models (see, e.g., Cohen et al. 2005; Gilbert et al. 2005; Gilbert et al. 2007; Kwon 2005; Levy et al. 2003; Luginaah et al. 2006; Ross et al. 2007; Sapkota and Buckley 2003).

two measured concentrations effectively equal. Three data points were available for each of these two remaining datasets, and the two closest to the road were fit linearly which provided a reasonable approximation for the edge of road concentrations. In all other cases, modifying the solution algorithm allowed the determination of a regression equation, even considering the small sample sizes (n = 3) in some cases. For each fitted model, the prediction at zero distance—the edge of road concentration—was assigned a value of unity, and the subsequent measurements were normalized to it. These two normalization procedures—background and edge—allow all of the data to be assessed, regardless of other shortcomings.

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