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Estimation of on-road NO₂ concentrations, NO₂/NO_x ratios, and related roadway gradients from near-road monitoring data

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Abstract

This paper describes a new regression modeling approach to estimate on-road nitrogen dioxide (NO₂) and oxides of nitrogen (NO_x) concentrations and near-road spatial gradients using data from a near-road monitoring network. Field data were collected in Las Vegas, NV at three monitors sited 20, 100, and 300 m from Interstate-15 between December, 2008 and January, 2010. Measurements of NO₂ and NO_x were integrated over 1-hour intervals and matched with meteorological data. Several mathematical transformations were tested for regressing pollutant concentrations against distance from the roadway. A logit-ln model was found to have the best fit ($R^2 = 94.7\%$) and also provided a physically realistic profile. The mathematical model used data from the near-road monitors to estimate on-road concentrations and the near-road gradient over which mobile source pollutants have concentrations elevated above background levels. Average and maximum on-road NO₂ concentration estimates were 33 ppb and 105 ppb, respectively. Concentration gradients were steeper in the morning and late afternoon compared with overnight when stable conditions preclude mixing. Estimated on-road concentrations were also highest in the late afternoon. Median estimated on-road and gradient NO₂ concentrations were lower during summer compared with winter, with a steeper gradient during the summer, when convective mixing occurs during a longer portion of the day. On-road concentration estimates were higher for winds perpendicular to the road compared with parallel winds and for atmospheric stability with neutral-to-unstable atmospheric conditions. The concentration gradient with increasing distance from the road was estimated to be sharper for neutral-to-unstable conditions when compared with

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stable conditions and for parallel wind conditions compared with perpendicular winds. A regression of the NO_2/NO_x ratios yielded on-road ratios ranging from 0.25 to 0.35, substantially higher than the anticipated tail-pipe emissions ratios. The results from the ratios also showed that the diurnal cycle of the background NO_2/NO_x ratios were a driving factor in the on-road and downwind NO_2/NO_x ratios.

Keywords

Near road; NO_2 ; oxides of nitrogen; nitrogen dioxide; dispersion

1. Introduction

The respiratory effects of nitrogen dioxide (NO_2) exposure are well known [EPA (2016), Hei (2010), and references cited therein]. Therefore, a number of cities have measured concentrations of NO_2 near major roadways with the goals of characterizing human exposure to ambient NO_2 and/or understanding how meteorology and the built environment influence NO_2 concentrations near roadways (e.g., Beckerman et al. 2008; Bignal et al. 2007; Cape et al. 2004; Carslaw 2005; Carslaw and Beevers 2005; Durant et al. 2010; Gilbert et al. 2003; Gonzales et al. 2005; ES Kimbrough et al. 2013; Maruo et al. 2003; Massoli et al. 2012; McAdam et al. 2011; Monn 2001; Pleijel et al. 2004; Polidori and Fine 2012; Singer et al. 2004; Smargiassi et al. 2005; Uemura et al. 2008; Westerdahl et al. 2005; Yli-Tuomi et al. 2005; Zou et al. 2006). However, only a few studies have measured NO_2 concentrations on roadways (Benson 1984) or in vehicles (Baldauf et al. 2013). These studies have reported reductions in concentrations between an on-road or near-road monitor and a downwind monitor with approximate gradients (i.e., change in concentration with change in distance from the source) of 0 to 12 ppb decrease in NO_2 concentration per meter of distance away from the roadway. However, the decline in concentration over distance from the road is not linear, as Karner et al. (2010) observed by regressing near-road concentration measurements from several studies, normalized by either background or on-road concentrations, against distance from the road.

Differences among conditions make it difficult to compare the near-road ambient concentration gradients reported in previous studies. For example, near-road monitoring locations have varied substantially among previous studies (Baldauf et al. 2009; Beckerman et al. 2008; Bignal et al. 2007; Cape et al. 2004; Carslaw 2005; Carslaw and Beevers 2005; Durant et al. 2010; Gilbert et al. 2003; Gonzales et al. 2005; ES Kimbrough et al. 2013; Maruo et al. 2003; Massoli et al. 2012; McAdam et al. 2011; Monn 2001; Pleijel et al. 2004; Polidori and Fine 2012; Singer et al. 2004; Smargiassi et al. 2005; Uemura et al. 2008; Westerdahl et al. 2005; Yli-Tuomi et al. 2005; Zou et al. 2006). Near-road monitors in these studies were positioned anywhere between 0 and 50 m from the road, while the downwind monitors were positioned anywhere between 10 m and more than 1 km away from the road. Furthermore, these studies were conducted under different conditions including season, climate, traffic level, fleet mixture, topography associated with each city (e.g., New York, Raleigh, Los Angeles, Tokyo, Zurich, Toronto), level of available O_3 , monitor type (passive, chemiluminescence, or tunable infrared laser differential absorption spectrometer), and

number of monitors. Additionally, U.S. NO_2 levels measured after 2005 are likely to have been influenced by mobile source emissions reduction policies outlined in the Tier 2 Motor Vehicle Emissions Standards of 2000 (40 CFR Parts 80, 85, and 86) and 2007-2010 phase-in of heavy-duty highway engine emission standards for oxides of nitrogen (NO_x) (40 CFR 86.007-11). The majority of previous near-road studies were conducted prior to the implementation of major mobile source emission reduction policies, thus, may not reflect recent changes in diesel emissions.

Estimating the on-road concentrations of NO_2 from near-road monitors is complicated by atmospheric chemistry. NO_2 comprises a small fraction of tailpipe emissions of oxides of nitrogen (NO_x), while the majority of tailpipe NO_x consists of nitric oxide (NO). Emissions measurements illustrate that median NO_2/NO_x ratios are less than 0.1 for low emissions vehicles including hybrid and compressed natural gas buses but can be as high as 0.7 in vehicles with diesel particulate filters or trucks with unknown emissions control technology (May et al. 2014; Shorter et al. 2005); Dallmann et al. (2012) published data to suggest that emissions of NO_2 are 7% of total NO_x emissions for heavy duty diesel trucks. In the near-road environment, NO_2 is relatively non-reactive, while NO can rapidly react with ambient ozone (O_3) to form additional NO_2 (Kota et al. 2013; Wang et al. 2011). This reaction can occur on the roadway, altering the on-road distribution of NO and NO_2 from what would be expected from tailpipe emissions alone. NO_2 and NO rapidly interconvert in the near road environment. Oxidation of NO and NO_2 occurs on time scales of minutes to hours, and the reaction rate is limited by available O_3 . Furthermore, during stable conditions, downward transport of O_3 is restricted, so that there may be less available O_3 to facilitate conversion of NO to NO_2 . As a result, the sum of NO_2 and NO (total NO_x) is conserved, so that changes in NO_x concentrations can be assumed to occur only by mixing and dilution with the background air. Thus, it is convenient to examine the NO_2/NO_x ratio, as it serves as a metric of the chemistry affecting NO_2 concentrations. While a number of studies have examined changes in the NO_2/NO_x ratio in the on-road environment (e.g., (Chaney et al. 2011)), the NO_2/NO_x ratio has received less attention than trends of absolute concentrations in the context of the near-road environment (e.g., (Baldauf et al. 2013)).

A few studies have developed mathematical functions to describe near-road gradients of air pollutant concentrations. Gilbert et al. (2003), Pleijel et al. (2004), and Roorda-Knappe et al. (1998) used logarithmic regression to estimate the NO_2 gradient, and Cape et al. (2004) applied an exponential decay function. More recently, Zou et al. (2006) employed a shifted power law model, for this purpose; mathematical adjustments were similarly employed in the RLINE (Heist et al. 2013; Snyder et al. 2013) and AERMOD (Heist et al. 2013) model designs to avoid the singularity at the road. These models have some limitations that complicate estimation of on-road NO_2 concentrations. For example, the logarithmic model produces an infinite solution for on-road NO_2 (with a distance of zero from the road). The shifted power law avoids producing an infinite solution on the road by adding one to the distance from road, but this is an artificial method to stabilize the model that also requires the on-road concentration to be known. Likewise, the exponential decay model requires the on-road concentration to be known to produce a solution along a gradient. Therefore, none of these mathematical models are well-suited for estimating on-road concentrations using

data from near-road monitors sited within some distance (typically 10-50 m) from the roadside (EPA 2010).

The objective of this work is to develop a mathematical model for the near-road spatial concentration gradient. We describe a new regression modeling approach to estimate near-road concentrations of NO_2 , NO_x , and the NO_2/NO_x ratio based on distance from the road. We also estimate on-road concentrations of NO_2 , NO_x , and NO_2/NO_x using this model. We take advantage of field data obtained at multiple near-road sites in Las Vegas, NV for fitting and testing our model (ES Kimbrough et al. 2013; Su Kimbrough et al. 2013). The model is then used to evaluate characteristics of the near-road gradients under different meteorological conditions. The goal of this work is to understand what conditions influence on-road NO_2 concentrations and the spatial extent of the near-road concentration gradient.

2. Methods

2.1 Study Area

In this investigation, we use the near-road measurements of air quality, traffic, and meteorology obtained at a study area in Las Vegas, NV (shown in Figure 1). The near road monitoring study area was chosen where 1) the Annual Average Daily Traffic (AADT) exceeded 150,000 vehicles per day, 2) airflow downwind of the highway was not blocked by natural or human-made structures, and 3) state and local governments permitted sampling sites within 300 meters (m) of the road to be established. The Las Vegas study area was located adjacent to Interstate-15 (I-15). Along this segment of the road, AADT is approximately 206,000 vehicles per day, with 10% of those characterized as heavy-duty diesel trucks (Nevada 2008). At this location, I-15 runs in the north-south direction, and the highway sits below grade with walls sloping upwards from the road at a 20° embankment. The terrain above the embankment is flat within a 10 km radius of the road. Near road sampling sites were located approximately 20 m, 100 m, and 300 m east of the highway. A railroad was located alongside the monitors. The 2011 National Emissions Inventory (EPA 2013) shows that the only large source of NO_x in the surrounding area besides I-15 was McCarran International Airport (LAS), and the closest point to the sampling site on the runway was approximately 800 m east of the road, typically downwind of the road. Trains also passed less than once per day, and the duration of their passage was less than five minutes. Baldauf et al. (2013) performed analyses with 5-minute data from this dataset and found that the train only affected the concentration data during calm-to-light winds. Meteorology at this study area is generally characterized as arid, with hot summers and abundant sunshine throughout the year. With mountains surrounding the Las Vegas metropolitan area, atmospherically stable conditions occur frequently during the evening and nighttime. A detailed description of this study area is provided in (ES Kimbrough et al. 2013).

2.2 Data Collection

Nitric oxide (NO) and total oxides of nitrogen (NO_x) were monitored continuously, logged on a five-minute averaging basis, and averaged on an hourly basis. NO and NO_x were monitored by chemiluminescence with a trace oxides of nitrogen analyzer (Ecotech, Model

EC 9841 B, Knoxfield, VIC, Australia), and nitrogen dioxide (NO₂) was estimated via differencing. Multipoint calibration was performed at the beginning of the study, and zero and span checks were performed nightly for each of the gaseous monitors. Inlets for each of these monitors were placed approximately 3 m above ground, and air pollutant concentrations were measured at the 20 m, 100 m, and 300 m sites.

Surface meteorological parameters monitored included wind speed and direction, along with air temperature, relative humidity, precipitation, and solar radiation. All meteorological parameters were measured at LAS as part of the standard urban scale meteorological measurements made by the National Weather Service (NWS) at most major airports. The LAS meteorological station, which is approximately 1.5 km from the Las Vegas near-road site, is part of the Automated Surface Observing Systems (ASOS) and has a One-minute temporal resolution for wind speed and direction. Upper air data for elevation, temperature, moisture, ozone column, and wind components were obtained from the Universal Rawinsonde Observation (RAOB) station in Mercury/Desert Rock, NV (KDRA, elevation 1006 m). One-Minute ASOS wind data were processed for input to AERMET using AERMINUTE for the period 12/1/2008 through 2/28/2010 (Cimorelli et al. 2005). Upper air and surface data were processed through AERMET to obtain hourly averages of surface and upper air meteorological parameters. Wind speed and direction were the only parameters used in their raw form. All other measured surface parameters used in this analysis were calculated by AERMET. A wind rose is presented in Figure 2 to summarize the wind data.

Ambient concentration data were collected at the Las Vegas near-road site between December 12, 2008 and January 21, 2010. There were 8,466 complete hours (81.8%) of data (based on the presence of NO₂ data at all three downwind sites) available for that time period and used in the analyses presented here. Data were further selected such that NO_x concentration decreased with distance from the road to increase the likelihood that sources of NO_x other than the road influenced the study area. This further selection reduced the dataset to 6,361 hours of data.

2.3 On-Road Concentration and Ratio Estimation

A regression model was fit to the concentrations measured at the three downwind monitoring locations to estimate the on-road concentration of NO₂ and NO_x at each hour where measurement data were complete. All models were of the basic form:

$$f(C) = m \cdot g(x) + b \quad (\text{equation 1})$$

where $f(C)$ = a statistical distribution fit to the concentration (C) data across the three monitoring sites, $g(x)$ = a statistical distribution fit to the location of the monitors across the three sites, m = the model estimated slope, and b = the model estimated intercept. The C and x data were transformed according to the functions, $f(C)$ and $g(x)$ shown in Table 1 to create linear, ln-ln, ln-linear, and logit-ln models. A lognormal distribution was fit to the distance data in some of the models to linearize that term, since the monitor sites were not evenly spaced relative to one another. Likewise, a lognormal distribution was fit to the concentrations in some of the models to linearize that term for the same reason. If a ln-ln

model were used, then the model could not be solved for on-road concentrations (at a distance of $x = 0$), because the declining on-road concentration would produce a negative slope, leading to a solution of infinite concentration when integrating the derivative to solve for $f(C)$ at $x = 0$. A logit function was fit to the concentrations for the logit-ln model to test if the concentration distribution approximated an S-shaped curve. Use of the logit-ln model was most physically sensible, because turbulent mixing related to traffic in some instances could cause concentration levels to plateau on and near the road and then gradually drop off in a manner similar to a Gaussian distribution (centerline to lateral extrema) or an exponential decay model. Note that the reference value occurred at $x = 300$, because the reference point had to be along the data distribution for the functional fit to apply. The reference was considered a point downstream where concentration returned to background levels.

The median, average, and percentile statistics were calculated for R^2 across the data, since there were 6,361 curve fits corresponding to each complete time period. The logit-ln formulation generally had the highest median R^2 of all model types ($R^2 = 97.1\%$ compared with 88.4%, 96.4%, and 92.1% for the linear, ln-ln, and ln-linear models, respectively for NO_2 ; $R^2 = 97.1\%$ compared with 78.9%, 98.0%, and 86.3% for NO_x ; and $R^2 = 93.1\%$ compared with 71.7%, 92.4%, and 69.5% for the linear, ln-ln, and ln-linear models for the ratios). Median model error for the logit-ln was highest at the 100 m site at 3.9% for NO_2 ; it was less than 1% for NO_2 at the 20 m and 300 m sites. These results, in conjunction with the physical rationale described above, support use of the logit-ln model to estimate on-road concentrations.

3. Results and Discussion

We used near-road measurements of NO_2 and NO_x in Las Vegas, NV during a variety of meteorological conditions to predict on-road concentrations of NO_2 and NO_x , on-road ratio of NO_2/NO_x , and near-road gradients of all three metrics. The logit-ln regression model was used to estimate these metrics for any downwind distance from the roadway based on measured near-road concentrations and ratios. Here we present estimates for on-road, 20 m, 100 m, and 300 m sites corresponding directly to measurement locations. The concentration models generally had a negative slope, where concentration decayed with distance from the road. The models of the NO_2/NO_x ratio typically had a positive slope, where the ratio increased as NO was converted to NO_2 as the air moved away from the road. Any deviations from this pattern indicate the influence of sources other than the roadway at intervals between the monitors (e.g., airport emissions, the emissions from a truck parked near the 300 m site would decrease the NO_2/NO_x ratios observed at the monitor). Since the regression assumed a consistent trend between the roadway and the monitors, these data have been excluded from the regressions for all meteorological condition subsets. Limitations of this approach are discussed later in this paper.

The on-road concentrations and ratios were estimated for several scenarios: A) all wind and stability conditions combined, B) time of day (0900–1200, 1700–2000, 0000–0300), C) wind direction and stability combinations (winds parallel or perpendicular to the road, stable or unstable atmospheric mixing conditions), D) season (winter: December, January,

February, summer: June, July, August), and E) weekday/weekend. Table 2 presents the model parameters for the logit NO₂ concentration gradient model for each set of conditions. Unstable conditions tended to occur during the daytime, while stable conditions typically took place during the evening and overnight hours. The 0900–1200 time period was generally characterized by convective mixing with moderate traffic. The 1700–2000 time period coincides with late afternoon/early evening rush hour, when atmospheric conditions were transitioning to stable and traffic was at rush hour levels. The 0000–0300 period had stable atmospheric conditions and low traffic. Because Interstate-15 runs north-south in this portion of Las Vegas, predominant winds from the southwest made the monitors downwind of the highway most of the time. The measured NO₂ concentration distributions at each measurement site are shown for all conditions combined in Figure 4. Summary statistics for the observed concentrations and model fit based on all input data combined are provided in Table 3. Overall, we found good agreement between measurements and predictions when examining the figures for each scenario, because the median and range of the observations (given by the second to ninety-eighth percentile of the data) typically coincided with the median and range of the model predictions.

For the analysis based on all data, the average on-road concentrations were estimated to be 33 ppb for NO₂ and 73 ppb for NO_x (see Table 3 and Figure 3 for summary data). The maximum estimated on-road concentration was 105 ppb for NO₂ and 264 ppb for NO_x. Despite the use of the logit function to represent the concentration variable, Figure 4 illustrates that the concentration trend with increasing distance from the roads was consistent with exponential/logarithmic decay functions described in other similar measurement studies (Beckerman et al. 2008; Bignal et al. 2007; Cape et al. 2004; Durant et al. 2010; Gilbert et al. 2003; Gonzales et al. 2005; ES Kimbrough et al. 2013; Maruo et al. 2003; Massoli et al. 2012; McAdam et al. 2011; Monn 2001; Pleijel et al. 2004; Polidori and Fine 2012; Singer et al. 2004; Smargiassi et al. 2005; Uemura et al. 2008; Zou et al. 2006). This suggests that human exposure to ambient NO₂ will substantially increase closer to the roadway at this study site.

Concentrations declined from the estimated on-road to 10 m from the road by a median of 16% and from on-road to the 20 m site by a median of 21% for NO₂ (Table 4) for all data combined. To evaluate whether these concentration decreases varied across different concentration levels, the 10 m and 20 m modeled data were stratified by quintile, and then the median relative difference between on-road and the 10 m or 20 m site was computed for each quintile. This is informative for estimating on-road NO₂ exposures based on measured near-road concentrations. When stratifying the percent change in concentration by quintile of concentration at the 10 m or 20 m location, the highest median difference between on-road and the 10 m or 20 m site was 26% and 34%, respectively (Table 4) and occurred in the second quintile (i.e., at comparably lower NO₂ concentrations). At higher NO₂ concentrations, the median percent change decreased to 10% for within 10 m of the roadway and 13% within 20 m of the roadway (Table 4).

The gradients in NO₂ concentration and NO₂/NO_x ratio for different conditions were thought to be influenced by convective mixing, prevailing winds, emissions levels, and atmospheric chemistry. The estimated on-road concentration was thought to be impacted by

emissions levels, while mixing, prevailing wind, and atmospheric chemistry all were thought to affect the steepness of the NO concentration gradient. The NO₂/NO_X ratio was thought to be influenced heavily by atmospheric chemistry. Diurnal O₃ concentration patterns on weekdays and weekends at background ambient monitoring sites is shown in Figure 6 to provide additional context for discussion of reaction between NO and O₃ to produce NO₂.

As shown in Figure 7, the concentration gradients were steeper in the morning and late afternoon compared with overnight when stable conditions reduce mixing. Estimated on-road concentrations were highest in the late afternoon. In general, on-road concentrations were expected to be higher during stable conditions, when the warm air mass aloft hinder transport and dispersion of air pollutants (Seinfeld and Pandis 2006). Moreover, in the absence of large-scale atmospheric mixing, traffic-induced turbulence theoretically became much more important for local dispersion of oxides of nitrogen emitted from mobile sources (e.g., He and Dhaniyala 2011; Rao et al. 2002; Sedefian et al. 1981; Wang and Zhang 2009). This would be most true during the early morning, before the inversion layer lifts and coinciding with an increased number of vehicles during rush hour.

The median estimated NO_X gradient over 20 m was -43% for both winds from the west and parallel winds during neutral-to-unstable conditions. The similarity between these concentration gradients could imply that atmospheric mixing was more important than wind direction during neutral-to-unstable conditions. Likewise, differences in photochemistry with respect to wind direction were unlikely during neutral-to-unstable conditions, which mostly occurred during the day. The estimated near-road gradient was sharper for parallel winds during stable conditions (Figure 9) compared with the gradient when winds were from the west (Figure 8). The median estimated NO_X gradient over 20 m was -29% for winds from the west compared with -38% for parallel winds during stable conditions. This was reasonable, given that perpendicular winds directed air pollution towards the monitors, even as some of it dispersed. In general, the NO concentration gradient with increasing distance from the road tended to be sharper than the NO₂ gradient (Karner et al. 2010).

Photochemical conversion of NO to NO₂ is known to occur over very short time scales. Thoma et al. (2008) measured the NO gradient downwind of a highway in Raleigh, NC under varying wind conditions and observed a linear relationship between percent decrease in concentration between two downwind sites and deviation of wind direction from perpendicular to the road. In other words, the smallest magnitude gradient occurred when winds were perpendicular to the road and directed towards the monitor, while the gradient increased in magnitude as wind direction shifted towards parallel. Because many studies accounted for wind direction in the study design a priori, where sampling was only performed under downwind conditions, the number of studies that examined effects of wind direction on the near-road concentration gradient for other study areas is small.

When analyzing the influence wind direction has on the on-road concentrations (Figures 8 and 9), estimated concentrations were generally higher for winds from the west during stable conditions compared with concentrations when winds were parallel to the road during stable conditions. Similarly, on-road concentration estimates were higher for winds from the west during neutral-to-unstable conditions compared with on-road concentration estimates when winds were parallel to the road during neutral-to-unstable conditions. Hence, on-road

concentration estimates were higher when winds were perpendicular to the road compared with when winds were parallel to the road. When considering the influence of wind direction on the on-road concentration estimates, it is notable that the below-grade shallow roadway embankment has been shown in wind tunnel studies to create elevated concentrations at the roadway within the embankment and lower concentrations at the ground level on par with the sampler locations (i.e., at the edge of the embankment) (Baldauf et al. 2013; Heist et al. 2009). This occurs because air movement slows down as it expands to fill the embankment space. Hence, the on-road concentrations estimated here likely underestimated those on the actual section of Interstate-15 in Las Vegas. At the same time, a street canyon dynamic did not occur, because there is no airflow separation at the edge of the embankment (Heist et al. 2009). The plume of traffic-related emissions therefore maintained spatial dependence downwind of the road, so it was valid to develop an empirical estimate of concentration at any point along the near-road gradient. The embankment was also likely to have curtailed lateral dispersion from the road for the parallel winds scenarios, so it was possible that the embankment suppressed the gradient for this wind scenario. Using data from flat terrain experiments by Cadle (1976) testing dispersion of automobile emissions using SF₆ as a tracer gas, Rao et al. (2002) observed that on-road concentrations were slightly higher for parallel winds scenario compared with perpendicular winds. Perpendicular winds may act to move NO₂ and NO away from the road more effectively than parallel winds, which would likely push the traffic-related pollution along the road but not ventilate it as well (e.g., Venkatram et al. 2013)).

When analyzing the influence of atmospheric stability on the on-road concentrations, estimated concentrations were higher for stable conditions compared to periods of atmospheric instability. For example, on-road concentrations were higher for stable conditions for winds from the west compared with neutral-to-unstable conditions for winds from the west. Likewise, on-road concentration estimates were higher for stable conditions for parallel winds compared with neutral-to-unstable conditions for parallel winds. Hence, on-road concentration estimates were higher during stable conditions compared with neutral-to-unstable conditions, such that atmospheric mixing diluted on-road concentrations. In general, on-road concentrations are expected to be higher during stable conditions, when the warm air mass aloft hinders transport and dispersion of air pollutants (Seinfeld and Pandis 2006). Moreover, in the absence of large-scale atmospheric mixing, traffic-induced turbulence theoretically becomes much more important for local dispersion of NO_x emitted from mobile sources (e.g., He and Dhaniyala 2011; Rao et al. 2002; Sedefian et al. 1981; Wang and Zhang 2009). This would be most important during the early morning, before the inversion layer lifts and this time period coincides with an increased number of vehicles during rush hour.

Median estimated concentrations on-road and east of the road were lower during summer compared with winter, with a steeper gradient during the summer (Figure 10). The steeper gradient is consistent with greater convective mixing during the summer, when solar radiation is higher for a longer portion of the day (Thornton and Running 1999). The magnitudes of estimated concentrations were also higher for weekdays compared with weekends (Figure 11), which reflected lower NO_x emissions related to lower traffic. However, no discernible difference in the slope of the NO₂ gradient was observed for

weekends compared with weekdays, given that no difference in photochemistry would be related to day of week.

The average on-road NO_2/NO_x ratio for the analysis of all data combined was 0.39 (Figure 5, Table 3, and Supplemental Figure S1). This is consistent with NO_2/NO_x ratios observed during the Advanced Collaborative Emissions Study, which ranged from 0.33 to 0.69 over 16-hour tests (Southwest Research 2013). Other studies reported on-road NO_2/NO_x ratios to be relatively low. Yasuyuki et al. (2014) estimated on-road ratios ranging from 0.1–0.15 in Osaka, Japan, while Kota et al. (2013) estimated on-road ratios of 0.29 were needed to match modeling concentrations with those observed in Austin, TX, much higher than 0.05 on-road ratio indicated from the emissions modeling alone. Jimenez et al. (2000) measured concentrations of on-road NO and NO_2 and observed that NO_2 concentrations were roughly 8% of NO concentrations, which would produce NO_2/NO_x ratios of approximately 0.07. Such diversity in on-road emissions, combined with on-road conversion of NO to NO_x could result in a wide variety of on-road NO_2/NO_x ratios. Indeed, the on-road estimates of the NO_2/NO_x ratios found span a wide range values, from 0.0 to 1.0, with the average on-road ratios estimated to range from 0.25 to 0.35 (Table 3 and Figures 7–9). Wang et al. (2011) emphasized the need to use on-road NO_2/NO_x ratios that vary, based on the hourly traffic conditions, in order to appropriately replicate near-road observations.

In general, the NO_2/NO_x ratio was hypothesized to increase with increasing distance from the roadway due to mixing with background air and conversion of NO to NO_2 (Figures 5, 7–11) (Chaney et al. 2011; Kota et al. 2013). Thus, the behavior of the data and the shape of the modeled distribution of the NO_2/NO_x ratios across the near-road zone was expected. While the constituent data should represent downwind impacts only from the roadway and without interference from intervening sources (as indicated by a decrease in the NO_2/NO_x ratio between any two stations), the data at the monitoring locations and the on-road estimates showed several interesting patterns. For example, the late afternoon NO_2/NO_x ratio gradient was much steeper than in the morning. The late afternoon period coincided with a steep drop in O_3 concentration at the background sites that may have corresponded to O_3 consumption by NO emitted by motor vehicles (Figure 6). Additionally, during the parallel winds scenario, the monitors were effectively farther downwind than when winds were from the west, giving more time for entrainment of O_3 and thus resulting in higher NO_2/NO_x ratios along the monitor transect. This is clear in for winds from the west under stable conditions, with a median 300 m ratio of 0.88. Based on data from Las Vegas and two other near-road field studies, (Venkatram et al. 2013) confirmed that periods with winds parallel to the roadway tended to have lower concentrations at nearby monitors than periods with perpendicular winds, partly due to the increased travel time to the receptors during parallel winds and additional mixing with background air that occurs with the longer travel times. Similarly, during the daytime scenarios, there should have been more O_3 available for NO conversion, and there should have been more mixing with background air due to the neutral-to-unstable conditions, resulting in higher NO_2/NO_x ratios than during the stable nighttime scenarios. However, the stable air mass scenarios had higher mean NO_2/NO_x ratios at all monitors. This was most likely related to the NO_2/NO_x ratio of the background air and the reduced total emissions from fewer vehicles on the roadway. The NO_2/NO_x ratio was driven both by the on-going conversion of NO to NO_2 and by mixing with the

background air, which would drive the in-plume NO_2/NO_x ratio towards the NO_2/NO_x ratio in the background air (Carslaw and Beevers 2004; Yasuyuki et al. 2014). During the daytime, the photochemistry associated with NO_x and O_3 has a tendency to force the NO_2/NO_x equilibrium ratio lower, because NO_2 is photolyzed by sunlight, turning back into NO .

This analysis was limited by the assumption that the logit model is appropriate for every atmospheric condition included in the data set. Model performance, as measured by R^2 , is reasonably high for all atmospheric conditions ($R^2 = 95.6\text{--}98.7\%$ across wind directions and stability conditions studied). Furthermore, model evaluations (predicted vs. observed concentrations) did not vary substantially by atmospheric condition. Therefore, the logit model appeared to be a robust choice to fit the data. A second limitation existed for all data combined, which included periods when the wind was blowing from the monitors towards the roadway during neutral-to-unstable conditions (generally daytime with higher background ozone) and during stable conditions (generally during nighttime with lower background O_3). Winds from the east occurred during 1,412 of the 6,361 data records (22%). These factors complicated interpretation of the regression results. On the other hand, there was the potential for downward bias and artificially decreased confidence intervals from selective removal of data that did not meet the criteria of concentration declining with distance from the road. However, this would only introduce bias for cases where elevated concentrations at the 100 m or 300 m site compared with the 20 m site were caused by photochemistry or meteorology rather than by unaccounted sources. Comparison of the two approaches at calculating the on-road NO_2/NO_x ratio (i.e., regression of the measured ratios vs. ratio of the estimated average on-road NO_2 and NO_x) gave some sense of the uncertainty of the three regressions by allowing a comparison of on-road estimates derived from two different sets of reactions. Although the comparison between these two estimates of the on-road NO_2/NO_x ratio are most appropriate on an hour-by-hour basis, rather than the comparison of the averages presented here, this comparison still lends some insight into the range of uncertainty associated with the regressions. Another limitation of this work was that on-road NO_2 and NO_x measurements were not available to validate the on-road estimates that are presented here. However, good validation of the models at the 20 m, 100 m, and 300 m sites lends confidence to the model fit. Finally, this analysis was performed only for the Las Vegas study area. This study area had limited influence from sources other than those originating from the roadway, and the measurements were taken orthogonal to a cut roadway section. This relationship would not necessarily be representative for urban sites with multiple NO_x sources including, for example, emissions from additional arterial roads or combustion-related power plants. However, this work provides important insight about NO_2 concentration changes from a single highway.

4. Conclusion

From a practical perspective, this analysis can shed light on how useful the existing near-road monitoring network may be for estimating the near-road gradient and on-road NO_2 and NO_x concentrations and NO_2/NO_x ratios. This method of estimation needs to be confirmed with concurrent near-road and on-road measurement studies. Nighttime inversions are a prevalent feature of the meteorology in Las Vegas. Hence, our stable condition estimates of

approximately 10–25% reductions in NO₂ concentration concurrent with an approximate 75% increase in the percent of NO_x that is NO₂ within 20 m of the roadway might be reasonable predictions for nighttime gradients in many cases for other regions where stable conditions tend to occur. When near-road concentrations are relatively low (i.e., lower percentiles of the concentration distribution) during nighttime inversions, reductions around 25–30% within the 20 m of the roadway might be more reasonable. During neutral-to-unstable conditions, which are more prevalent during the daytime hours, we observed reductions in NO₂ concentrations of 35% concurrent with an approximate 85% increase in the proportion of NO_x made up by NO₂ within 20 m of the roadway, although NO₂ concentrations are generally lower for this scenario. The selection of a higher gradient applied equally to all possible atmospheric conditions or concentration levels would tend to produce a lower estimate of on-road NO₂ concentrations. In general, this approach provides exposure scientists a low-cost tool for estimating how near-road exposures vary for people driving on major roadways or living in close vicinity to a busy street.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Figure 1.

The Las Vegas study area. (top) Plane view showing that Interstate-15 runs north-south, and the monitoring sites follow along a northwest-to-southeast transect along a road crossing; (bottom) view from the road looking downwind, showing the embankment and position of the monitor 20 m downwind of the road (Baldauf et al. 2013; ES Kimbrough et al. 2013).

Figure 2.
Wind rose data obtained at LAS airport for the study period.

Figure 3.

Measured NO₂ and NO_x at the three measurement sites during the study period. The box represents the middle 50% of the data, extending for the 25th-75th percentiles. The horizontal line through the center of the box is the median. The whiskers represent 1.5*IQR (the interquartile range, the range from the 25th-75th percentiles). The points are outliers beyond 1.5*IQR.

Figure 4. Predicted and observed NO₂ concentrations for all wind and stability conditions combined. Predicted median (solid), predicted 98th and 2nd percentile (dotted), observed median (circles), and observed 98th and 2nd percentiles (error bars) are shown.

Figure 5. Predicted and observed NO_2/NO_x ratios for all wind and stability conditions combined. Predicted median (solid), predicted 98th and 2nd percentile (dotted), observed median (circles), and observed 98th and 2nd percentiles (error bars) are shown.

Figure 6. Measured O_3 from two ambient monitors sited in the Las Vegas metropolitan area (left) west of the field site, (right) east of the field site. Weekend and weekday patterns are shown.

Figure 7.

(left) Predicted and observed NO_2 concentrations for (top) 0900 – 1200, (middle) 1700 – 2000, (bottom) 0000 – 0300. (right) Predicted and observed NO_2/NO_x ratios for (top) 0900 – 1200, (middle) 1700 – 2000, (bottom) 0000 – 0300. Predicted median (solid), predicted 98th and 2nd percentile (dotted), observed median (circles), and observed 98th and 2nd percentiles (error bars) are shown.

Figure 8. (left) Predicted and observed NO_2 concentrations for winds from the west under (top) stable and (bottom) unstable atmospheric mixing conditions. (right) Predicted and observed NO_2/NO_x ratios for winds from the west under (top) stable and (bottom) unstable atmospheric mixing conditions. Predicted median (solid), predicted 98th and 2nd percentile (dotted), observed median (circles), and observed 98th and 2nd percentiles (error bars) are shown.

Figure 9.

(left) Predicted and observed NO_2 concentrations for winds parallel to the road under (top) unstable and (bottom) stable atmospheric mixing conditions. (right) Predicted and observed NO_2/NO_x ratios for winds from the west under (top) unstable and (bottom) stable atmospheric mixing conditions. Predicted median (solid), predicted 98th and 2nd percentile (dotted), observed median (circles), and observed 98th and 2nd percentiles (error bars) are shown.

Figure 10.

(left) Predicted and observed NO_2 concentrations for (top) winter, (bottom) summer. (right) Predicted and observed NO_2/NO_x ratios for (top) winter, (bottom) summer. Predicted median (solid), predicted 98th and 2nd percentile (dotted), observed median (circles), and observed 98th and 2nd percentiles (error bars) are shown.

Figure 11.
 (left) Predicted and observed NO_2 concentrations for (top) weekday, (bottom) weekend.
 (right) Predicted and observed NO_2/NO_x ratios for (top) weekday, (bottom) weekend.
 Predicted median (solid), predicted 98th and 2nd percentile (dotted), observed median (circles), and observed 98th and 2nd percentiles (error bars) are shown.

Table 1.

Concentration vs. distance from road model formulations.

Model Form	f(C)	g(x)	C(x)
linear	C	x	$m \cdot x + b$
ln-linear	$\ln(C)$	x	$\exp(b) \cdot \exp(m \cdot x)$
ln-ln	$\ln(C)$	$\ln(x)$	$\exp(b) \cdot x^m$
logit-ln	$e^{C(x)} / [e^{C(x)} + e^{C(\text{ref})}]$	$\ln(x)$	$C(\text{ref}) + \ln[b + m \cdot \ln(x)] - \ln[1 - [b + m \cdot \ln(x)]]$

C = concentration; x = distance; m = slope; b = intercept; ref = reference point (for the logit model, ref = 300 m)

Table 2.Logit model parameters for different conditions studied, NO₂ gradient model.

	Slope		Intercept		Onroad NO ₂ Concentration		R ²	
	Avg	σ	Avg	σ	Avg	σ	Avg	σ
All winds combined	-4.79E-04	4.30E-04	5.03E-01	2.46E-03	31.08	13.98	0.81	0.28
0900-1200	-4.29E-04	3.65E-04	5.02E-01	2.09E-03	23.65	12.14	0.85	0.25
1700-2000	-5.86E-04	4.60E-04	5.03E-01	2.63E-03	34.67	14.04	0.82	0.28
0000-0300	-3.41E-04	4.51E-04	5.02E-01	2.61E-03	32.51	12.14	0.71	0.32
Winds from west (perpendicular), stable	-3.98E-04	3.63E-04	5.02E-01	2.09E-03	36.79	11.20	0.79	0.29
Winds from west (perpendicular), unstable	-7.68E-04	3.79E-04	5.04E-01	2.16E-03	32.04	13.44	0.91	0.18
Winds parallel to road, stable	-6.45E-04	5.37E-04	5.04E-01	3.08E-03	34.00	14.36	0.78	0.29
Winds parallel to road, unstable	-7.21E-04	4.48E-04	5.04E-01	2.55E-03	28.68	13.48	0.86	0.24
Summer	-5.84E-04	4.91E-04	5.03E-01	2.81E-03	29.03	14.58	0.80	0.29
Winter	-3.90E-04	3.68E-04	5.02E-01	2.11E-03	34.06	13.03	0.81	0.28
Weekday	-4.96E-04	4.47E-04	5.03E-01	2.56E-03	32.68	14.28	0.80	0.28
Weekend	-4.36E-04	3.79E-04	5.02E-01	2.17E-03	26.97	12.28	0.82	0.27

Table 3.

Summary statistics for observed and predicted concentrations for all wind and stability conditions combined.

Location with respect to the road ^a	NO ₂		NO _x		NO ₂ /NO _x	
	Avg	σ	Avg	Avg	σ	Avg
Observations						
20 m	22.01	13.96	32.56	28.64	0.79	0.17
100 m	25.92	12.00	51.02	36.47	0.59	0.15
300 m	22.40	12.10	37.55	32.00	0.72	0.18
100 m upwind	19.47	12.25	31.31	29.70	0.76	0.18
Model Predicted						
On-road	33.03	13.35	73.17	47.35	0.39	0.22
20 m	25.93	12.00	51.05	36.49	0.59	0.15
100 m	22.11	11.97	39.17	31.96	0.69	0.16
300 m	19.50	12.25	31.07	29.69	0.77	0.18

^aDownwind unless otherwise noted.

Table 4.

Summary statistics for percent change in NO₂ concentration from modeled on-road to concentrations at distances of 10 m and 20 m away from roads for scenario A (all atmospheric conditions combined), by quintile of NO₂ predicted concentrations at 10 m and 20 m away from the roads.

	10 m to on-road comparison		20 m to on-road comparison	
Quintile	Conc range (ppb)	Median % change	Conc range (ppb)	Median % change
1	2.25 to 15.66	24%	2.31 to 14.09	32%
2	15.66 to 23.09	26%	14.09 to 21.07	34%
3	23.09 to 31.25	18%	21.07 to 29.43	24%
4	31.26 to 39.13	10%	29.43 to 37.55	13%
5	39.13 to 67.78	10%	37.56 to 66.94	13%
overall	2.25 to 15.66	16%	2.31 to 66.94	21%