Assessment of the Near-Road (monitoring) Network including comparison with nearby monitors within U.S. cities

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Abstract

Emissions from on-road mobile sources have historically been an important anthropogenic contributor to ambient air pollution leading to high levels of air pollution near major roadways. The U.S. EPA recently implemented the Near-Road (monitoring) Network to measure NO$_2$ concentrations by high-traffic roadways in urban centers throughout the U.S., as these locations were believed to characterize worst-case human exposures to traffic-related air pollutants. Many near-road sites also include PM$_{2.5}$ and CO measurements, which along with the NO$_2$ observations, were compared in a pairwise manner against non-near-road monitors located within the city-scale boundary. After controlling for primary emissions from the target highways, we found the PM$_{2.5}$ concentration difference (i.e. near-road concentration minus non-near-road site concentration) between the near-road and non-near-road urban sites to be $\delta = 0.42 \mu g m^{-3}$ ($H_0$: $\mu_{diff} = 0$; $H_a$: $\mu_{diff} > 0$ ($\mu_{non-near-road} > \mu_{near-road}$); $p = 0.051$; $\alpha = 0.05$, 95% CI: $-0.08$–$0.90 \mu g m^{-3}$, $n = 35$ comparisons). NO$_2$ and CO levels were on average higher at the near-road sites compared to the non-near-road urban sites by 5.0 (95% CI: $3.4$–$6.5$) ppb ($n = 44$ comparisons) and $9.2 \times 10^{-2}$ (95% CI: $0.04$–$0.14$) ppm ($n = 42$ comparisons), respectively. The average PM$_{2.5}$ difference found here is 5%, and at 14 of the 35 (~40%) urban monitor comparisons and 28 of the 72 (~39%) overall comparisons, PM$_{2.5}$ is actually higher at the non-near-road site relative to its near-road pair. Cleaner vehicle fleets, formation of secondary PM from on-road emissions occurring downwind (i.e. away from the road), decreased secondary organic aerosol (SOA) formation rates in the near-road environment, the prevalence of other low-volume vehicular and local, non-vehicular sources of emissions at the non-near-road sites (e.g. railyards, truck yards, ports, biomass-fueled heating, backyard barbecuing, and commercial cooking, etc) and local meteorology (e.g. wind speed and wind direction) explain this finding. The wintertime PM$_{2.5}$ concentration difference was higher than the other seasons, likely a result of higher primary PM$_{2.5}$ tailpipe emissions and lower temperatures that both reduced near-road PM volatility and decreased photochemical activity resulting in lower SOA production at the urban scale. Further, all near-road NO$_2$ and CO concentrations were below the annual and hourly NAAQS, while eight (most of which were in wildfire-prone locations) of the 94 PM$_{2.5}$ sites used in this study were above the annual National Ambient Air Quality Standards. In addition, strong agreement with both annual average daily traffic and fleet-equivalent AADT were found for near-road NO$_2$ and CO concentrations, while weaker, but still positive relationships were found for near-road PM$_{2.5}$ levels. Lastly, same observational data was used to assess on-road mobile source emission estimates from the EPA National Emission Inventory, and analysis of the observations are in rough agreement with the current ratio of NO$_x$ to CO emissions from on-road mobile sources.
1. Introduction

High levels of air pollution in U.S. cities have often been observed near major roadways, largely due to the proximity of on-road mobile sources (herewith referred to as mobile sources) [1]. Despite large reductions in mobile source emissions in the U.S., they are expected to continue to be a large fraction of the total anthropogenic emissions as other sources, e.g. power plants, industrial emissions, etc, have also become increasingly well controlled over time [2]. Since 1970 emissions from mobile sources in the US have dropped 82% for CO, 81% for VOCs, 58% for NOx, and 58% for fine particulate matter (PM2.5, particulate matter, PM, whose aerodynamic diameter is less than 2.5 microns) [3, 4], but mobile sources still contribute 59% of all anthropogenic NOx, 54% of all anthropogenic CO, and 18% of all anthropogenic primary PM2.5 [4].

Exposure to air pollution is associated with adverse health outcomes that result in premature mortality and reduced life expectancy [5–8], with the vast majority (~95%) of air pollution-related mortality linked to PM2.5 [6, 9]. Past exposure assessments have estimated that roadway-associated exposures account for elevated PM2.5 and NO2 exposures [10, 11], raising environmental justice concerns for residents near roadways. Mobile source impacts have been found to disproportionately affect poorer, racial minority, and vulnerable populations whose residents live near major highways [12, 13]. As a result, there have been more recent efforts to characterize pollution exposures targeting the near-road environment [14–17], with a trend to finding lower near-road PM2.5 increments [18–21].

The U.S. EPA initiated the Near-Road (monitoring) Network as part of the 2010 NO2 National Ambient Air Quality Standards (NAAQS) review because these sites, which are located at high-traffic locations in cities where peak ambient NO2 concentrations are expected to occur as a result of mobile source emissions, would represent the worst-case population exposures in the near-road environment [22]. All near-road sites are required to measure NO2, PM2.5 and CO are required in every core-based statistical area (CBSA) with 1 million or more persons [22]. Some sites include black carbon (BC), NO, NOx, and VOC monitoring as well.

Previous assessments of city-scale (e.g. Detroit, MI [12,], Houston, TX [23,] and Boise, ID, Des Moines, IA, St. Louis, MO, Detroit, MI, New York City, NY, and Los Angeles, CA [24,]) and regional-scale (e.g. DeWinter et al [25] and Seagram et al [26]) comparisons of near-road observations to very nearby non-near-road monitors have been infrequent and limited. The NO2 city-scale studies [12, 24] found that the near-road environment had higher NO2 concentrations for each hour of the day than a background, non-near-road site measurement, while the PM2.5 city-scale study showed 24 h PM2.5 concentrations in Houston were 23% higher than background observations [23]. DeWinter et al [25] and Seagram et al [26] compared near-road PM2.5 concentrations with all EPA Air Quality System (AQS) sites within 25 km, 50 km, and 100 km using datasets from 2015 and 2016, respectively, that included Phase 1 (of 2) near-road sites and found a near-road PM2.5 concentration increment of 15%. However, there was no focus on the comparison with other surrounding non-near-road urban sites <25 km, where the majority of the population would be subject to similar urban emissions.

Other approaches to measuring on-road pollution, including via bicycles [27] and Google Street View Cars [18] regularly find elevated levels of PM2.5 and BC on high-traffic roads or roads near highly localized mobile-source emissions (intersections along a major truck route, vehicle repair facilities, etc). Such studies have found that some pollutant hot spots may be caused by non-mobile sources within cities [18].

The two primary objectives of the work presented in this paper are to characterize the differences in PM2.5, NO2, and CO concentrations between near-road and non-near-road environments in U.S. cities nationwide using recent, long-term monitoring and to evaluate potential biases in the emission estimates of traffic-related air pollutants (TRAPs) using the same long-term monitoring data. Here, we: (1) present a 2 year average, nationwide, pairwise comparison of near-road PM2.5, NO2, and CO concentrations against non-near-road PM2.5, NO2, and CO concentrations within the city/urban-area scale to characterize pollutant concentration differences between the near-road and non-near-road environments, (2) compare the seasonal trends of the PM2.5, NO2, and CO average concentration differences, (3) evaluate observed PM2.5, NO2, and CO levels at each site with the corresponding NAAQS level, (4) compare near-road PM2.5, NO2, and CO concentrations with annual average daily traffic (AADT) and fleet-equivalent AADT (FE-AADT) counts to gauge mobile source impacts on near-road air quality, (5) assess local wind speed and wind direction impacts on observed PM2.5, NO2, and CO concentrations at the near-road sites to assess how local meteorology is influencing observed near-road pollution levels, and (6) utilize the NOx and CO concentration difference data to evaluate mobile-source emission estimates in the National Emission Inventory (NEI).

2. Materials and methods

The Near-Road (monitoring) Network is a regulatory network that began monitoring in 2014 to characterize NO2 in the near-road environment. The network contained 70 active, operational sites (referred to herewith as near-road sites) located...
in 62 continental U.S. cities and one site situated in San Juan, PR as of May 2017 (SI figure 1 (stacks.iop.org/ERL/15/114026/mmedia)). Near-road sites are intended to measure at locations that would result in peak NO₂ concentrations related to mobile source emissions, presumably associated with high-traffic spots in cities. Near-road sites (which can also be influenced from other non-mobile source emissions) are within 50 m of the intended high-traffic roadway, and 56% (39 out of 70) of these monitors are within 20 m of the major roadway (SI figure 2). The location of near-road sites are not necessarily representative of other locations on the same road and caution has been recommended from previous studies on using near-road sites for exposure studies [28, 29]. NO₂, PM₂.₅, and CO are monitored at 69 (no data at the Des Moines, IA. site), 43, and 56 of the near-road sites, respectively.

The PM₂.₅ instruments used at the near-road sites are either a Met One Beta Attenuation Monitor (BAM) 1020, a TEI 5014i BAM, a Grimm 180, or a TEI 1405-DF Tapered Element Oscillating Microbalance (TEOM). Previous assessments of co-located Federal-Equivalent Method (FEM) monitors have shown variability [30, 31], including differences approaching 10% on 24-hour averaged PM₂.₅ levels [32]. The biases between the measurement approaches are attributed to local ambient conditions (e.g. aerosol composition, temperature, and relative humidity). In this work, we do not perform any additional quality assurance or quality control to the EPA reported concentrations.

Observations used in this study were from the continental U.S. near-road monitors along with regulatory monitors within the same city or within 10 miles of a near-road site (referred to herewith as non-near-road sites; see SI table 1 for a complete list of sites in the study). The 10 mile condition is applied as some non-near-road monitors are across city lines (e.g. state borders) but still likely subject to the same urban emission profile. The average size of the top-50 most populous U.S. cities is ~300 sq. miles [33], and considering that near-road sites are often centrally located, the 10 mile radius is appropriate to capture observations that may be affected by the same urban emission profile. The near-road sites used in this study are regulatory monitors that can represent a general population or capture areas that may be sited in response to a specific nearby-source and may be a part of other monitoring networks.

We compared the hourly concentrations for PM₂.₅, NO₂, and CO for hours where data existed at both the near-road site and paired non-near-road site to characterize pollutant concentration differences in the near-road and non-near-road environments in U.S. cities; the non-near-road sites were not necessarily sited for the purpose of pairing with a near-road site, but we use those data to do the comparisons. The instruments used to measure the pollutant concentrations at the non-near-road sites are not necessarily the same instruments used at the paired near-road site. The comparisons presented in this study do not characterize the incremental concentration attributable only to traffic on a target highway as the near-road monitoring network was designed to assess potential elevated exposures on or near major roadways, not to explicitly capture the incremental pollutant impacts from the traffic on that particular road [22]. For many near-road sites, there were multiple non-near-road sites within the city boundary/10 mile radius (SI figure 3). Each non-near-road site was compared against the near-road site within the city boundary/10-mile radius. The non-near-road sites were characterized as urban, suburban, or rural as given in the Air Quality System (AQS) dataset [34]. The focus of this assessment is on urban sites; there are no urban-designated sites identified in the same city as the near-road site that are also outside the 10-mile radius. However, there are suburban- and rural-designated sites that could be impacted by the same urban emissions profile but are not within the city boundary or 10 miles of the near-road site.

Non-near-road sites may be affected by on-road emissions (e.g. from the roads next to which near-road monitors are located), but past studies have found that mobile-source emitted pollutants decay within a few 100 meters of the road for all pollutants [18, 35]. To control for such target highway emissions, we use previous findings that estimate BC (surrogate to PM₂.₅) and NO₂ decay to background at 350 m and 970 m [18]. There are no recent studies that estimate CO decay to background from mobile source emissions, so we treat the CO decay to background distance to be the same as the NO₂ decay to background distance. We exclude any comparison pairs where the non-near-road site is within these distances of the target near-road roadway from the analysis (these sites are indicated in SI table 1; we exclude 11 PM₂.₅ comparisons, 37 NO₂ comparisons, and 35 CO comparisons).

There were 72 comparisons at 30 near-road sites for PM₂.₅, 91 comparisons at 55 near-road sites for NO₂ (NO₂ measurements are at each near-road site, but for some near-road sites the non-near-road site pair was within the NO₂ decay distance, while other near-road sites do not have a corresponding comparison site within the city boundary/10 mile radius), and 70 comparisons at 36 near-road sites that measure CO (SI table 2). Because of measurement limitations (e.g. no monitor or recorded data at the near-road site, no non-near-road site within the city boundary/10 mile radius, and/or no non-near-road site outside the pollutant decay distance to background zone), some of the most populous cities in the U.S. were not included in this study. This includes Chicago, IL and Salt Lake City, UT for NO₂, CO and PM₂.₅; Atlanta, GA for NO₂ and PM₂.₅, San Antonio,
TX for PM$_{2.5}$ and CO, and San Diego, CA, Dallas, TX, and Houston, TX for PM$_{2.5}$ comparisons.

The assessment period covers two full years (Jan. 2017–Dec. 2018). The start date of this analysis was 2017 as this was the latest start year of the most recent list of active, near-road monitors at the time of this analysis. Hourly PM$_{2.5}$, NO$_x$, and CO concentrations were retrieved from the EPA AQS database pre-generated data files for 2017 and 2018 and were downloaded following the May 1st, 2019 EPA data certification deadline. In addition, hourly NO$_x$ concentrations were retrieved from the same sites that also had an NO$_2$ measurement, where available.

Here, we examined the hypothesis that two-year average PM$_{2.5}$, NO$_x$, and CO concentrations are elevated at the near-road sites when compared to nearby (~city scale) non-near-road sites. A one-tailed t-test ($H_0$: $\mu_{\text{diff}} = 0$; $H_a$: $\mu_{\text{diff}} > 0$ ($\mu_{\text{non-near-road}} > \mu_{\text{near-road}}$); $\alpha = 0.05$) was performed to assess the statistical significance of the concentration differences at a 95% confidence level for each pollutant. A one-tailed t-test was selected so that the alternative hypothesis, $H_a$, could be defined as the unexpected situation that the non-near-road concentration is greater than the near-road concentration. We present the results of our statistical significance tests but note there are limitations when interpreting results from statistical tests for significance [36, 37], and recent literature suggests comparison tests that are significant at 95% but not at 99.5% should be called suggestive and not significant [38], and we follow this nomenclature in this paper. Seasonal and monthly trends of the concentration differences were also assessed. The measurements were evaluated against the U.S. NAAQS levels and near-road concentrations were compared against annual average daily traffic (AADT) and fleet-equivalent annual average daily traffic (FE-AADT). FE-AADT gives heavy-duty vehicles 10 times the vehicle count of a standard vehicle as defined by:

$$FE \cdot AADT = (AADT - HD_c) + (10 \times HD_c)$$

where $HD_c$ is the total number of heavy-duty vehicles. The same analysis was performed for BC, which would be a more direct combustion emission surrogate, but there were only four cities where the appropriate data existed for comparison (see SI section 1 for BC findings). In addition, coefficients of divergence (COD), a measure of spatial heterogeneity [39], were computed to gauge the spatial variability among the near-road and non-near-road monitors for each pollutant (SI section 2). Inverse-distance weighting of the pollutant concentration differences between the near-road and non-near-road sites was assessed (SI section 3). Further, NO$_2$ to NO$_x$ concentration ratios, which is useful to estimate the fraction of ambient NO$_x$ that has undergone oxidation from the primarily-emitted NO [40], were assessed at the near-road and non-near-road sites (SI section 4).

The local meteorology (e.g. wind speed (WS) and wind direction (WD)) and local configuration (vehicle-induced turbulence, physical barriers, monitor distance to highway, depressed roadways, etc) of near-road sites influence how traffic emissions from the target roadways can effect observed concentrations [25, 26, 41]. In this work, we relate the observed near-road PM$_{2.5}$, NO$_x$, and CO concentrations with near-road site monitored wind direction and wind speed ($n = 41$). We do not assess configuration affects in this analysis. Downwind was when the angle between the target highway and near-road site was in-line with the observed wind direction (+ a 90° buffer; we choose a wide angle window under the rationale that at non-perpendicular angles an air parcel travels over more roadway length and thus receives more emissions [42]). The remaining conditions were treated as upwind conditions. Wind speed conditions were split as either less than or equal to 1 m s$^{-1}$ or greater than 1 m s$^{-1}$, consistent with wind speed bins used in DeWinter et al [25]. Here, we compare the following four combinations of hourly wind conditions on hourly near-road PM$_{2.5}$, NO$_x$, and CO observations: (1) WS < 1 m s$^{-1}$ and near-road site upwind, (2) WS < 1 m s$^{-1}$ and near-road site downwind (expected to give the highest average concentration at each site), (3) WS > 1 m s$^{-1}$ and near-road site upwind (expected to give the lowest average concentration at each site), and (4) WS > 1 m s$^{-1}$ and near-road site downwind. The results presented are two-year averages for each of the four combinations.

Recent studies have questioned the accuracy of mobile source emission estimates from the EPA National Emissions Inventory (NEI) [43, 44]; we use two approaches from the available data to evaluate the average NO$_x$ to CO emission ratio given in the 2017 and 2018 inventories [4]. The NEI estimates mobile-source NO$_x$ emissions and not NO$_2$ emissions. We cannot explicitly evaluate emission estimates from the dataset used here, but we can estimate ratios of the emissions from the concentration differences. The first approach was to estimate a NO$_x$ emission ratio from the slope of the orthogonal regression curve fitting differences in the CO concentrations averaged over 2 years to the difference in the NO$_x$ concentrations averaged over 2 years for each near-road and non-near-road site combination where both CO and NO$_x$ concentration differences existed. The second approach took the ratio of the average enhancement (i.e. the average concentration difference) of NO$_x$ between all near-road and non-near-road observations to the average enhancement of CO between all near-road and non-near-road observations (SI section 5). These approaches both assume the enhancement of NO$_x$ and CO at the near-road sites is driven by the difference in mobile source emissions affecting the near-road site minus the mobile source emissions affecting a paired non-near-road site, only. We consider both cases where negative
concentration differences (i.e. the near-road annual average observation is less than the non-road near-road annual average) are and are not included (SI section 5). Removing negative concentration differences was done because negative concentration differences should not exist for NO2 and CO as mobile sources are the largest contributors to each pollutant’s emissions, and the near-road sites were sited at highest-concentration locations in the near-road environments in cities.

3. Results and discussion

3.1. Comparisons of near-road and non-near-road PM2.5, NO2, and CO concentrations

Average concentration levels of PM2.5, NO2, and CO as well as the differences between the Near-Road (monitoring) Network (near-road sites) and non-near-road sites, were calculated using observational data from 2017 and 2018, further considering if the non-near-road sites were designated as urban, suburban, or rural (table 1). The PM2.5, NO2, and CO concentration differences between near-road and non-near-road sites were 0.50 µg m⁻³ (95% CI: 0.17–0.84 µg m⁻³), 6.2 ppb (95% CI: 5.2–7.1 ppb), and 9.2 × 10⁻² (95% CI: 5.8 × 10⁻²–0.13 ppm), respectively. The PM2.5 concentration difference between the urban non-near-road sites and the near-road sites was 0.42 µg m⁻³ (H0: μdiff = 0; Hα: μdiff > 0 (μnon-near-road > μnear-road); p = 0.051, α = 0.05, 95% CI: 0.08–0.90 µg m⁻³) (table 1 and SI section 6 and SI table 3). A ramifications of this finding is that policy makers should not immediately view that the near-road environment has significantly higher PM2.5 levels than the non-road environment, particularly as one moves away from the highway given the rapid dispersion of PM [45]. The PM2.5 comparison between the near-road and rural designated non-road-pair also did not show a suggestive difference (α = 0.05); however, there was a limited number of sample comparisons for these comparisons, and we do not include all rural monitors that could be affected by the urban emission profile as they fell outside the city/10-mile criteria radius.

Thirty-nine percent (28 out of 72) of the non-road sites have a higher 2-year average PM2.5 concentration than the corresponding near-road site, with the highest difference being 3.2 µg m⁻³ between Oakland West (AQS Site ID# 06-001-0011) and the Berkeley near-road site (AQS Site ID# 06-001-0013) (figure 1 and 2, SI figures 4 & 5, and SI table 4). The Oakland West monitoring site began operation in 2009 to capture downwind emissions from the nearby Port of Oakland, a major source of diesel PM [46]. A review of city and state air monitoring plan handbooks found 10 of the 72 non-road sites from the paired PM2.5 comparisons were sited at source-oriented or highest concentration sites, and four of them actually had lower concentrations than the corresponding near-road site (SI table 5).

The average difference between near-road NO2 measurements and corresponding non-road near-road is 6.2 ppb (95% CI: 5.2–7.1 ppb, p = 1.9 × 10⁻²). Five (out of 45) cities (St. Louis, MO., Buffalo, NY, Queens, NY, Philadelphia, PA, and Detroit, MI) had one or more non-road monitors with higher average NO2 than the companion near-road site (figure 1, SI figure 5, and SI table 4). The seven (out of 91) non-road sites that exceed the corresponding near-road NO2 level are all either in industrial areas or closely located to major waterways, which could be subject to local boating and shipping emissions, a source whose emissions are growing [47, 48] and are not well controlled [48]. As expected, average levels decreased (in order) from the near-road, to urban, suburban, and rural environments (table 1 and figure 1).

CO was 9.7 × 10⁻² ppm higher (95% CI: 6.0 × 10⁻²–0.13 ppm, p = 6.1 × 10⁻⁷) on average at the near-road sites than the non-road sites (table 1). The spatial pattern of the non-road vs. near-road difference for CO was similar to NO2 (figure 1 and SI figures 4 & 5), which was expected considering that mobile sources account for the majority of anthropogenic CO and NO2 emissions [3]. There are 15 (out of 70) comparisons where the average CO concentration is higher in the near-road environment than at the near-road monitor (SI table 4). The highest difference is 0.40 ppm between the Indianapolis—Illinois St. monitor (AQS Site ID# 18-097-0072) and the Indianapolis—I-70E near-road monitor (AQS Site ID# 18-097-0087). The Illinois St. monitor is located downtown in a commercial area and is identified as a highest concentration monitor (SI table 5). This site does not measure NO2 or PM2.5, so it is difficult to assess what else may be causing the relatively elevated CO levels. Like NO2, average CO levels decreased from the near-road, to urban, suburban, and rural environments (table 1 and figure 1).

3.2. Seasonal variation of the PM2.5, NO2, and CO concentration differences

PM2.5 experienced more seasonal variation in the near-road and non-near-road site concentration differences as compared to NO2 or CO seasonal concentration differences (table 2 and SI figure 6). The wintertime (Dec.–Feb.) urban PM2.5 concentration difference is higher than the other season average PM2.5 concentration differences which may be explained by higher wintertime PM2.5 tailpipe emissions [49] and lower temperatures, the latter reducing volatilization of semi- and intermediate volatility organic aerosols [50, 51]. Further, secondary organic and inorganic aerosol formation is depressed in the wintertime due to decreased photochemical activity, decreasing the impact of on-road emissions of PM precursors on secondary PM levels on urban scales.
Figure 1. Two-year average concentration differences between near-road sites and companion non-near-road sites for (a) PM$_{2.5}$ ($\mu$g m$^{-3}$) ($n = 72$) (b) NO$_2$ (ppb) ($n = 91$), and (c) CO (ppm) ($n = 70$). Positive values indicate near-road sites have higher two-year average concentrations than a companion non-near-road site. Only paired comparisons where the non-near-road monitor fell outside a ‘decay to background’ distance of 350 m for PM$_{2.5}$ or 970 m for NO$_2$ and CO from the target highway were included in the analysis. Some near-road sites have multiple non-near-road pair comparisons. The stars indicate a near-road site location, and arrows from the circles (two-year average concentration differences) indicate the comparison is made at that near-road site. See SI figure 4 for city-averaged comparisons.
The observed data at the near-road and non-near-road sites were compared against the NAAQS levels for each pollutant. Three (of the 29) near-road sites (San Francisco—Laney College, Los Angeles—Long Beach, and Los Angeles—Ontario) and five (of the 65) non-near-road sites (Oakland West, Pittsburgh—Liberty, Los Angeles—Long Beach-South, Los Angeles—Upland, and Los Angeles Mira Loma Van Buren) used in this study have higher two-year average PM$_{2.5}$ levels than the primary standard of 12 µg m$^{-3}$ (NAAQS standard is annual mean averaged over 3 years; here, we average over 2 years; SI table 8). The Pittsburgh monitor that is above the 35 m or 970 m for NOX and CO from the target highway were included in the analysis. The asterisk (*) indicates the concentration difference is not suggestive ($H_0$: $\mu_{diff} = 0$; $H_1$: $\mu_{diff} > 0$; $\mu_{near-road} > \mu_{non-near-road}$; $\alpha = 0.05$). Pollutant concentration comparisons where non-near-road sites that are source-oriented or highest concentration are excluded from the comparisons can be found in SI table 6.

### Table 1

| All sites       | Near-road sites | Non-near-road sites | Urban non-near-road sites | Suburban non-near-road sites | Rural non-near-road sites | All sites range |
|-----------------|-----------------|----------------------|---------------------------|-----------------------------|---------------------------|----------------|
| Two-year average (± 1σ) concentrations | Two-year average (95% CI) concentration differences |
| PM$_{2.5}$ (µg m$^{-3}$) | NO$_X$ (ppb) | CO (ppm) | PM$_{2.5}$ (µg m$^{-3}$) | NO$_X$ (ppb) | CO (ppm) |
| 9.5 (±1.7); n = 95 | 12.8 (±5.9); n = 121 | 0.37 (±0.13); n = 92 | 0.50 (0.17–0.84); n = 72 | 6.2 (5.2–7.1); n = 91 | 0.09 (0.06–0.13); n = 70 |
| 9.8 (±1.4); n = 30 | 15.9 (±5.8); n = 55 | 0.42 (±0.13); n = 36 | N/A | N/A | N/A |
| 9.2 (±2.0); n = 65 | 9.7 (±4.1); n = 66 | 0.32 (±0.10); n = 56 | N/A | N/A | N/A |
| 9.4 (±1.4); n = 29 | 11.4 (±3.6); n = 31 | 0.33 (±0.11); n = 30 | 0.42* (−0.08 to 0.90); n = 35 | 5.0 (3.4–6.5); n = 43 | 0.09 (0.04–0.13); n = 42 |
| 9.0 (±1.9); n = 32 | 8.4 (±3.8); n = 30 | 0.30 (±0.10); n = 24 | 0.52 (−0.02 to 1.1); n = 33 | 6.4 (5.3–7.4); n = 40 | 0.10 (0.04–0.17); n = 26 |
| 8.1 (±1.2); n = 4 | 5.3 (±1.7); n = 5 | 0.19 (±0.04); n = 2 | 0.92* (−0.63 to 2.5); n = 4 | 10.9 (6.2–15.6); n = 8 | 0.10* (−0.26 to 0.45); n = 2 |
| N/A | N/A | N/A | N/A | N/A | N/A |

| Winter | Spring | Summer | Fall |
|--------|--------|--------|------|
| 0.65 (0.15–1.15) | 0.83 (0.16–1.49) | 5.5 (4.5–6.5) | 6.4 (5.4–7.4) |
| 0.32 (−0.08 to 0.74) | 0.32 (−0.08 to 0.74) | 4.0 (2.6–5.5) | 4.9 (3.4–6.5) |
| 0.34 (8.1 × 10$^{-3}$–0.67) | 0.20* (−0.26 to 0.66) | 6.4 (5.4–7.3) | 6.4 (5.4–7.3) |
| 0.39 (0.11–0.84) | 0.28* (−0.19 to 0.75) | 6.2 (5.2–7.2) | 5.2 (3.6–6.8) |

### Table 2

Wintertime (Dec.–Feb.) NO$_X$ concentration differences are slightly lower, on average, compared to the other seasons, which is likely due to lower average mobile-source NO$_X$ emissions in the winter and a longer NO$_X$ lifetime, providing for further downwind NO$_X$ transport and city-wide buildup [52]. There is little observed seasonal difference of CO concentration differences, on average, between the near-road and non-near-road sites (table 2 and SI figure 6). Surprisingly, both NO$_X$ and CO concentration differences have an opposite trend to PM$_{2.5}$ in the winter (i.e. it is the season where the smallest differences between the near-road and non-near-road sites are observed), so decreased wintertime mixing would not appear to be a major factor for the increased wintertime PM$_{2.5}$ concentration differences.

### 3.3. National Ambient Air Quality Standards (NAAQS) evaluations

The observed data at the near-road and non-near-road sites were compared against the NAAQS levels for each pollutant. Three (of the 29) near-road sites (San Francisco—Laney College, Los Angeles—Long Beach, and Los Angeles—Ontario) and five (of the 65) non-near-road sites (Oakland West, Pittsburgh—Liberty, Los Angeles—Long Beach-South, Los Angeles—Upland, and Los Angeles Mira Loma Van Buren) used in this study have higher two-year average PM$_{2.5}$ levels than the primary standard of 12 µg m$^{-3}$ (NAAQS standard is annual mean averaged over 3 years; here, we average over 2 years; SI table 8). The Pittsburgh monitor that is above the standard is sited as a highest-concentration site, is downwind of a steel plant, and is located on top of a school building adjacent to the bus loop—subject to local emissions from both sources. Each of the other non-near-road and near-road sites where observed concentrations are higher than the standard are in areas that have been prone to wildfires in recent years [53]. There were seven (of the 29) near-road sites and 11 (of the 65) non-near-road sites that were above the 35 µg m$^{-3}$ 24 h standard at least once (98th...
Figure 2. Box-plot 2 year average concentration differences between (a) PM$_{2.5}$, (b) NO$_2$, and (c) CO near-road sites and all non-near-road sites, urban non-near-road sites, and non-urban (suburban and rural) non-near-road sites. Positive values indicate near-road sites have higher two-year average concentrations than the non-near-road sites. The boxes, red line, and red diamond represent the interquartile range (25%–75%), the median, and the 2 year average concentration difference, respectively.

percentile, NAAQS standard is averaged over 3 years, here we average over 2 years; SI table 8), each of which are also locations prone to wildfire plumes.

All NO$_2$ and CO concentrations (averaged over 2 years) at near-road and non-near-road sites used in this study are below the hourly and annual NO$_2$ and hourly and annual CO NAAQS. There were no NO$_2$ or CO concentrations that were above the hourly standards in 2017 or 2018, an improvement compared to the first 2 years of the network’s inception when exceedances occurred [25], and the maximum levels observed at the near-road sites over these 2 years were lower than those from 2016 at the near-road sites [26]. The highest observed NO$_2$ annual mean concentration amongst the sites used in this study was 32 ppb at the Ontario, CA. Route 60 near-road site (AQS Site ID# 06-071-0027) in 2017, below the 53 ppb NAAQS standard. The highest 98th percentile of 1-hour daily maximum concentrations averaged over the 2 year study period was 89 ppb at the Long Beach, CA. near-road site (AQS Site ID# 06-37-4008), below the 100 ppb NAAQS standard (which is averaged over 3 years). The highest CO 8 h average concentration is 4.5 ppm at the Minneapolis Near-Road I-35/I-94 site (AQS Site ID# 27-053-0962), below the NAAQS of 9 ppm that is not to be exceeded more than once per year. The highest 1-hour reporting from observations used in this study is 9.0 ppm at the Pittsburgh Parkway East Near-Road site (AQS Site ID# 42-003-1376), well below the 35 ppm hourly NAAQS that is not to be exceeded more than once per year.

3.4. Annual Average Daily Traffic (AADT) and Fleet-Equivalent AADT (FE-AADT) against near-road PM$_{2.5}$, NO$_2$, and CO concentrations

AADT had a positive and statistically significant ($\alpha = 0.05$) correlation with two-year average NO$_2$ and CO concentrations at the near-road sites, and a weaker and not statistically significant relationship with PM$_{2.5}$ concentrations. All three of the pollutant concentrations had a positive, statistically significant ($\alpha = 0.05$) correlation with FE-AADT at the near-road sites (figure 3). FE-AADT gives heavy-duty diesel vehicles 10 times the vehicle
count of a standard vehicle, and the most recent Vehicle Inventory and Use Surveys (VIUS) reports approximately 87% of heavy-duty vehicles (>26 001 pounds) have diesel engines. Current literature now finds that diesel vehicles equipped with a diesel particulate filter (DPF) actually have lower PM$_{2.5}$ exhaust emissions per amount of fuel burned than gasoline direct injection (GDI) engines [54, 55]; though older non-DPF-equipped diesels still dominate primary carbonaceous emissions from all mobile sources [54, 56]. Data on nationwide DPF penetration is unavailable, but an assessment at an overpass at the Port of Oakland and the nearby Caldecott Tunnel found that 8% of heavy-duty trucks are non-DPF-equipped [56]. Considering that non-DPF-equipped diesels produce more than two orders of magnitude more PM$_{2.5}$ per kg fuel burned than DPF-equipped diesels [55], we expect to see a larger source impact from diesels on ambient PM$_{2.5}$ concentrations.

We find a lower CO correlation coefficient in the FE-AADT comparison compared to the correlation coefficient for either NO$_2$ or PM$_{2.5}$. FE-AADT is adjusted for the presence of trucks that are primarily diesel-powered, and diesels are not large contributors to CO emissions. Also, photochemical CO production from isoprene and other volatile organic compounds (VOCs) vary regionally and temporally. Global estimates show that isoprene oxidation contributes to about 13% (9%–16%) of the CO budget [57], and CO production from isoprene is more efficient in high NO$_x$ environments (e.g. near roadways) [58]. Although the NO$_x$ and CO relationships with both AADT and FE-AADT are statistically significant, the low correlations suggest near-road monitors are influenced by local characteristics (e.g. wind speed, wind direction, vehicle-induced turbulence, physical barriers, monitor distance to highway, depressed roadways, etc) and emissions other than those from mobile sources.

3.5. Wind speed and wind direction on two-year average near-road PM$_{2.5}$, NO$_2$, and CO concentrations

The near-road site downwind and wind speed (WS) < 1 m s$^{-1}$ criteria often results in the highest average observed PM$_{2.5}$, NO$_2$, and CO concentrations as expected (figure 4). However, at four (of the 21) near-road PM$_{2.5}$ sites, 22 (of the 41) near-road NO$_2$ sites, and 19 (of the 30) near-road CO sites, the downwind and WS < 1 m s$^{-1}$ does not result in the highest two-year average concentrations. At three of the four PM sites where this occurs (Riverside, CA AQS Site ID# 06-071-0027, Las Vegas, NV AQS...
Site ID# 32-003-1501, San Antonio, TX AQS Site ID# 48-029-1069), the downwind and WS < 1 m s$^{-1}$ condition does not give the highest average concentration for NO$_2$ or CO, either. However, at the Indianapolis, IN near-road site (AQS Site ID# 18-097-0087), the downwind and WS < 1 m s$^{-1}$ condition does result in the highest average NO$_2$ and CO concentrations. Brown et al [42] conducted a detailed assessment of meteorological factors influencing the Indianapolis near-road site and found a weak correlation between NO$_2$ and PM$_{2.5}$ concentrations during high PM$_{2.5}$ concentration hours, suggesting mobile sources are not a dominant contributor to high PM$_{2.5}$ concentration hours. They attributed the high PM$_{2.5}$ concentration hours that often occur at night to residential wood burning, low inversion heights, and upwind (wind moving in the direction of the road from the near-road site) conditions during these times of the day in Indianapolis.

52% of the total wind direction observations used in our full network analysis were during hours when the near-road site was upwind of the target highways. Thus, use of the near-road monitors to capture the near-road increment suffers from being upwind about half the time.

The low wind speed (WS < 1 m s$^{-1}$) and downwind condition does not result in the highest average concentration at the majority of the near-road sites for NO$_2$ and CO. For 15 (of the 22) NO$_2$ sites, 13 (of the 19) CO, and zero (of the four) PM$_{2.5}$ sites where this occurs (SI table 9), there is another major highway or interstate nearby (not the target highway) that is within the decay to background distance buffer ($L_{PM2.5} = 350$ m and $L_{NO2} = L_{CO} = 970$ m) whose emissions will influence the monitor at a different wind direction range. The hours of calm wind speeds and the monitor downwind of the target highway do not necessarily occur during peak emission hours. In addition, the actual highway configuration (e.g. vehicle-induced turbulence, physical barriers, depressed roadways, etc) will also impact near-road observations. Assessing these conditions on near-road monitors would require detailed meteorology, temporally-resolved emission inventories, and near-road dispersion modeling that considers the local roadway configuration.

3.6. Using NO$_2$ and CO concentration differences to assess the National Emission Inventory (NEI) mobile-source emission estimates

NO$_2$ and CO concentration differences between near-road and companion non-near-road site pairs that monitor both pollutants are positively correlated with each other (figure 3), which is expected considering mobile sources contribute the majority of NO$_2$ and CO emissions in the U.S [4]. The average of national on-road mobile NO$_2$ emission estimates in 2017 and 2018 according to the NEI is $5.6 \times 10^7$ kg yr$^{-1}$ while the average of CO on-road mobile emission estimates is $2.8 \times 10^{10}$ kg yr$^{-1}$ [4], a NO$_2$ to CO emission ratio of 0.20. The slope of the orthogonal best-fit regression between CO and NO$_2$ concentration differences is 172 (95% CI: 90–310) ppb NO$_2$ (ppm CO)$^{-1}$ (∼0.27 (0.14–0.49) µg m$^{-3}$ NO$_2$ (µg m$^{-3}$ CO)$^{-1}$), about 30% higher than the ratio in the NEI of 0.20 (figure 4, SI section 1 for statistical significance tests, and SI section 4 for unit conversion explanation). The NO$_2$ to CO emission ratio using the average enhancement of NO$_2$ (16 ppb) and CO (0.14 ppm) where both sets of measurements exist between the near-road and non-near-road measurements ($n = 31$) is 0.17 µg m$^{-3}$ NO$_2$ (µg m$^{-3}$ CO)$^{-1}$, about 15% less than the NEI NO$_2$ to CO emission ratio estimate (see SI section 5 for the case where negative concentration differences were removed). Recent studies have questioned the accuracy of the NEI’s mobile source emission estimates [43, 44], but the findings presented here suggest rough agreement between observations and the NEI NO$_2$ to CO emission ratio estimate (other pollutant emissions and ratios might still have high uncertainties). There was little correlation between PM$_{2.5}$ concentration differences to NO$_x$ or CO concentration differences, as expected, further emphasizing that mobile sources do not contribute as much to ambient PM$_{2.5}$ concentrations as they do to NO$_x$ or CO concentrations (SI figure 8). The average NO$_2$ to NO$_x$ at the near-road sites was 0.63 and was 0.81 for the non-near-road sites (SI section 4).

3.7. Explanations for similar PM$_{2.5}$ concentrations at near-road and non-near-road urban observations in U.S. cities

Historically PM$_{2.5}$, NO$_2$, and CO concentrations have been higher near major roads when compared to other, non-near-road locations in U.S. cities [1, 25, 26]. Recent assessments using various approaches, including fine-scale reduced-complexity models [21], emission inventory trends [3], distance decay transect studies [18, 19], and land-use regression predictions [20], have found that mobile sources generally contribute to low amounts of ambient PM$_{2.5}$ but still lead to enhancements near major highways. Here, using nationwide observations from the Near-Road (monitoring) Network and nearby non-near-road monitors, we also find a slight near-road enhancement for PM$_{2.5}$ (∼5% difference).

A number of factors have led to decreasing near-road PM levels over time. First, modern automobiles and trucks have lower primary PM$_{2.5}$ exhaust emissions [59, 60]. Near-road sites are also alongside high-traffic interstates and likely not subject to cold start emissions, which are estimated to contribute the majority of tailpipe PM emissions [61] and select VOC emissions from mobile sources [62]. Further, higher NO$_x$ levels, like those observed in a near-road environment, are associated with lower secondary organic aerosol (SOA) yields [63–65] and reduce the rate of VOC oxidation to SOA precursor species.
Figure 4. Wind speed and wind direction impacts on two-year average near-road site (a) PM$_{2.5}$ (µg m$^{-3}$) (b) NO$_2$ (ppb) and (c) CO (ppm) concentrations. The data presented here, including the average values, is only for hours when both wind data and pollutant concentrations exist in 2017–2018. The city names and the last four digits of the AQS Site ID are given for each near-road site. The asterisk (*) next to the AQS Site ID indicates a site that is within the decay to background distance buffer ($L_{PM2.5} = 350$ m and $L_{NO2} = L_{CO} = 970$ m) of another major highway or interstate (not the target highway).
These reasons would only explain why the near-road levels are not as elevated now as in the past, not why there is very little difference between near-road and nearby non-near-road observations.

Recent lab experiments estimate that SOA formation exceeds primary organic aerosol (POA) emissions after a few hours of atmospheric oxidation for most vehicles [66, 67], which would contribute to downwind PM$_{2.5}$ observations from mobile sources. Although newer vehicles tend to have higher SOA yields (SOA produced per unit mass of reacted precursor) on average, they also have less total SOA formation compared to older vehicles [66, 68]. Gordon et al. concluded that some vehicles generate as much as six times the amount of SOA as primary PM under typical oxidant levels [66]. Further, Zhao et al. showed that changing tailpipe emissions (i.e. the ratio of non-methane organic gases (NMOG) to NO$_x$) and atmospheric chemistry (SOA yields) may result in the same SOA production from cleaner vehicle fleets compared to previous fleets, despite large reductions in direct tailpipe precursor emissions [69]. These emissions would affect downwind, background observations more than the near-road PM observations.

The finding that PM$_{2.5}$ levels are similar near-road and at other urban monitoring sites in U.S. cities suggests that other non-major-road sources play an increasingly important role in air quality. High-emitting local sources (e.g. commercial cooking, barbecuing, biomass-fueled residential heating emissions that escape outdoors, rail yards, truck yards, ports, school busses, etc) and lower-volume vehicular sources can also play a role at non-near-road monitoring sites. Aligned with nationwide observations that organic carbon is becoming a major fraction of PM$_{2.5}$ loadings [70], the low PM$_{2.5}$ difference between near-road and non-near-road observations and low levels, in general, supports findings of the potential importance of biogenic and volatile consumer product emissions on aerosol loadings [71, 72].

The near-road environment having similar levels of PM$_{2.5}$ to adjacent areas has implications for health outcome assessments. PM$_{2.5}$ has been implicated as having the highest disease burden of air pollutants by the Global Burden of Disease (∼95%) [6, 9], and recent air pollution health effect studies in the near-road environment have found excess risk for near-road populations [73–75]. However, the finding that PM$_{2.5}$ is slightly higher in the near-road compared to non-near-road environments, which is consistent with recent near-road studies that used other approaches (e.g. fine-scale reduced-complexity models [21], distance decay transect studies [18, 19], and land-use regression predictions [20]), suggests that PM$_{2.5}$ mass itself should not be the only indicator of potential health impacts of traffic-related air pollutants. Because of their potential significance to environmental health, the results found in this paper should continue to be evaluated through further air quality comparisons in the near road and non-near road environment, as there are significant uncertainties when interpreting the results from near-road monitors. Future work should also include consideration of additional pollutants (e.g. PM$_{2.5}$ and PM$_{1}$ species, PAHs, etc) and characteristics (e.g. oxidative potential) as PM$_{2.5}$ measured at near-road

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**Figure 5.** The difference in 2 year average NO$_x$ and CO concentrations between a near-road site and non-near-road site. The positive values indicate the near-road two-year average concentration is higher than the non-near-road concentration. Three cases exist where the CO concentration difference falls below zero and the regression relationship was found to be statistically significant ($\alpha = 0.05$).
monitors may not be a robust indicator of traffic-related air pollutant emissions on potential health impacts.

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Author contributions

RML designed the research, conducted the research, and wrote the manuscript. AGR and AR advised the research and assisted with manuscript preparation and revisions.

Data availability statement

The data that support the findings of this study are openly available at https://doi.org/10.25412/iop.10028855.x1

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