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Atmospheric Measurement Techniques
Discussions

Mobile Air Quality Monitoring and Comparison to Fixed Monitoring Sites for Quality Assurance

Andrew R. Whitehill[1], Melissa Lunden[2], Brian LaFranchi[2], Surender Kaushik[1], Paul A. Solomon[2,a]

[1]Center for Environmental Measurement and Modeling, Office of Research and Development, United States Environmental Protection Agency, Research Triangle Park, North Carolina, 27711, United States of America
[2]Aclima, Inc., San Francisco, California, 94111, United States of America
[a]formerly at: Office of Research and Development, United States Environmental Protection Agency, Las Vegas, Nevada, 89119, United States of America

Correspondence to: Andrew R. Whitehill (whitehill.andrew@epa.gov)

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Abstract. Air pollution monitoring using mobile ground-based measurement platforms can provide high quality spatiotemporal air pollution information. As mobile air quality monitoring campaigns extend to entire fleets of vehicles and integrate smaller scale air quality sensors, it is important to address the quality assurance needs for validating these measurements. We explore collocation-based evaluation of air quality instruments in a mobile platform against fixed regulatory sites, both when the mobile platform is parked at the fixed regulatory site and when moving at distances of meters to kilometers from the site. We demonstrate agreement within 4 ppbv (for NO_2 and O_3+NO_2) to 10 ppbv (for NO) when using a running median of 40 hourly differences between the moving mobile platform measurements and stationary site measurements. The comparability is strong when only measurements from residential roads are used but are only slightly diminished when all roads except highways are included in the analysis. We present a method for assessing mobile measurements of ozone (O_3), nitrogen dioxide (NO_2), and odd oxygen ($O_X = O_3 + NO_2$) on an ongoing basis through comparisons with fixed regulatory sites.

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1 Introduction

Mobile air pollution monitoring can resolve fine-scale spatial variability in air pollutant concentrations, allowing communities to map air quality down to the scale of tens of meters in a reproducible manner (Apte et al., 2017;Van Poppel et al., 2013). Expanding fleet-based mobile monitoring will allow the mapping of much larger spatial regions over longer periods and with more repetition. This will improve land use regression models (Messier et al., 2018;Weissert et al., 2020) and supplement our understanding of air quality issues in environmental justice regions (Chambliss et al., 2021).

One concern with fleet-based mobile monitoring, especially as it expands to lower-cost and lower-power instrumentation, is instrument performance (e.g., accuracy, precision, and bias) in a mobile environment. Instrumentation can behave differently in a field environment than during laboratory testing and calibration (Collier-Oxandale et al., 2020), making field calibrations or collocations essential for quantitative measurement applications. Field validation usually involves the collocation of one or more test instruments with reference-grade instruments at a fixed monitoring site, such as a regulatory site (Li et al., 2022). Frequent collocations with fixed reference sites have been identified as an important quality assurance for mobile monitoring campaigns (Alas et al., 2019;Solomon et al., 2020;Whitehill et al., 2020). Collocated measurements are also important for validation of other emerging measurement technologies, such as lower-cost sensors (Bauerová et al., 2020;Castell et al., 2017;Masey et al., 2018).

Parking a mobile platform next to a fixed reference site ensures comparability only at that specific location and only under the specific atmospheric conditions over which the collocation occurred. In some cases, strong agreement when collocated at a fixed reference site may not translate into accuracy and precision in other environments (Castell et al., 2017;Clements et al., 2017). Therefore, it is advantageous to both the scalability of mobile monitoring and confidence in the precision and accuracy of mobile measurements to be able to reference mobile measurements to stationary, reference-grade measurements on an ongoing basis during measurement campaigns.

We explore the comparability of fixed reference sites to mobile measurements as a function of distance between the fixed reference sites and vehicle. The objective is to determine whether measurements from air pollution instrumentation on a mobile platform can be anchored to fixed reference site measurements when the moving platform is various distances from the stationary reference site instruments. As mobile platforms drive near regulatory sites (or other fixed reference sites), comparisons during those "rendezvous" periods can be used, over time, to assess mobile instrument performance (Xiang et al., 2020). Through ongoing comparisons of fixed reference site and mobile measurements, it may be possible to identify instrument drift over time or changes in instrument performance that could indicate a malfunction.

Ongoing mobile-versus-fixed-site comparisons are more scalable than frequent site-by-side parked collocations and could provide an important tool for ongoing quality assurance during mobile measurement campaigns. If "collocation" comparisons can be extended out to kilometer scales and spread across multiple fixed reference sites over the course of a single campaign, the amount of data used to validate the mobile measurements will be increased and maximize mobile monitoring for other purposes. This allows a different set of statistical techniques to be used to improve confidence in the ongoing comparability of different mobile platforms. The use of non-stationary mobile platforms also reduces the impact of any spatial or spatiotemporal biases that might impact parked, side-by-side comparisons (Whitehill et al., 2020). These extended analyses that include moving comparisons with fixed reference sites provide the advantage of scalability versus stationary, side-by-side comparisons, which is particularly important during sustained, multi-vehicle (and fleet-based) mobile monitoring campaigns.

Here, we compare air pollution monitoring instruments in a vehicle-based mobile platform to regulatory air quality monitoring site reference measurements. The use of fast response (1-Hz) laboratory-grade equipment in the mobile platforms will aid in unravelling the impact of operational and environmental differences in the measurements versus measurement accuracy and uncertainty arising from different measurement technologies between mobile and stationary. We aim to develop a method to look at the mobile-versus-fixed-site

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data on a dataset of laboratory-grade measurements. This will help us to understand the strengths and drawbacks of these methods. This work can be expanded in future work to mid-range instruments and smaller scale sensors, and eventually developed into a scalable approach for ongoing quality assurance during fleet-based mobile monitoring campaigns.

2. Overview of Methods

We analyze measurements from several different vehicles equipped with the Aclima, Inc. mobile laboratory measurement and acquisition platform (Aclima Inc., San Francisco, CA). These measurements come from two separate deployments in different locations and time periods. For the first deployment, we outfit three Google Street View cars (gasoline powered Subaru Imprezas) in Denver, CO in partnership with the Google Earth team (Whitehill et al., 2020). The second set of data comes from Aclima's Mobile Calibration Laboratory (AMCL), a gasoline powered Ford Transit van that we deployed in the San Francisco Bay Area. Aclima designed the AMCL to support the field calibration of Aclima's sensor-based Mobile Node devices (AMN) for deployment in the Aclima mobile monitoring fleet. In both studies, each mobile platform was equipped with high resolution (0.5 hz or 1 hz data reporting rate) reference-grade air pollution monitors to measure ozone (O_3), nitrogen dioxide (NO_2), and nitric oxide (NO), among other species.

O_3 was measured using ultraviolet (UV) absorption with a gas phase (nitric oxide) O_3 scrubber for the ozone-free channel (2B Technologies Model 211). This technology reduces some of the volatile organic compound (VOC) interferences observed in other UV photometric ozone monitors (Long et al., 2021), and has been designated a Federal Equivalent Method (FEM) by the USEPA (Designation EQOA–0514–215, 40 FR 79, June 18, 2014, p. 34734 - 34735). NO_2 was measured using cavity attenuated phase shift (Teledyne Model T500U) and represents a "true NO_2" measurement (Kebabian et al., 2005) and has also been designated as a Federal Equivalent Method (FEM) by the USEPA (Designation EQNA-0514-212, 40 FR 79, June 18, 2014, p. 34734 - 34735). NO is measured using O_3 chemiluminescence (Ecophysics CLD64), which is a recognized international standard reference method for measuring NO (e.g., EN 14211:2012). Although these instruments have all been evaluated by strict test criteria and are recognized as reference methods by various regulatory agencies, the instruments were selected for the demonstrated excellent data quality and performance and to serve as reference for calibration purposes recognizing that the reference method designations do not apply to the application (mobile monitoring) and timescales (1 second to 1 hour) assessed here. There is value in validating that these measurements in a mobile environment can be used for assessing existing mobile monitoring data. We are also excited about developing techniques to evaluate the performance of next-generation air quality instruments, which might not meet the same strict regulatory standards at present but still provide valuable data in smaller, lower-power, and lower-cost form factors (Castell et al., 2017;Clements et al., 2017;Wang et al., 2021). Using reference-grade instrumentation in this work provides a baseline indication of how different data analysis techniques may work when comparing mobile monitoring platforms to fixed reference sites. This baseline can then be used in future work to assess the performance of lower-cost monitors that may not meet strict "reference" criteria in terms of precision, accuracy, and stability.

We describe the calibration and quality assurance for the Denver mapping in Section 3.1, although a more thorough discussion was presented in a previous work (Whitehill et al., 2020). Calibration and quality assurance of the San Francisco Bay Area driving is discussed in Section 5.1.

3 Mobile platforms parked at fixed reference sites in Denver (2014)

3.1 Methods

We begin the analysis with the comparison of measurements from a parked mobile platform to those at a fixed reference site. In previous work for a similar mobile platform, Solomon et al. (2020) showed an agreement of about 10% on average for NO, NO_2, O_3,

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and O_3+NO_2 (O_X) during two parked collocations between two equivalent mobile platforms and a fixed reference site in Los Angeles, CA. Measurements of O_X are valuable because it is more likely to be conserved in fresh NO_X emission plumes (if most of the NO_X emissions occur as NO) than O_3 and NO_2. In addition, there are emerging technologies (such as some types of electrochemical sensors) that measure O_X directly rather than O_3 and NO_2 separately.

We use data from a mobile measurement campaign that occurred in the Denver, Colorado, USA region during the summer of 2014. Professional drivers drove three identical mobile air pollution monitoring platforms, consisting of specially equipped Google Street View cars, through the Denver, Colorado greater metropolitan region between July 25th, 2014 and August 14th, 2014. The project goals were to evaluate the performance of the mobile monitoring platforms and to develop methods for assessing data quality and platform comparability. The three cars drove coordinated routes in a 5 km area around several regulatory monitoring sites in the Denver, Colorado area, as well as driving around larger (10 km) areas to understand the variability of air pollutants at different spatial scales. At several planned periods during the study, we instructed the drivers to park one or more of the cars near one of the fixed regulatory monitoring sites in the region. We integrated these parked collocations into the experimental plan to assess the data quality of the mobile platform measurements by comparison to the fixed reference site measurements. The parked collocations lasted about 20 minutes and included comparisons at the Colorado Department of Public Health and Environment (CDPHE) CAMP (39.751184° N, 104.987625° W) and La Casa (39.779460° N, 195.005124° W) sites. Additional details about the operations, driving routes, and stationary sites can be found in the Supplemental Information (SI) and in Whitehill et al. (2020). The CDPHE reported hourly Federal Reference Method (FRM) and Federal Equivalent Method (FEM) regulatory measurements and provided 1-minute time resolution data for our study period as part of the 2014 DISCOVER-AQ experiment (https://www-air.larc.nasa.gov/missions/discover-aq/discover-aq.html).

Aclima staff performed quality assurance evaluations on the instruments daily in the field and after the study in the Aclima laboratory. Flow rates remained within instrument specifications throughout the study. We used instrument responses to zero air (from a zero air cylinder) to apply a study-wide zero offset for each instrument on each car. Results from daily span checks for NO (360 ppbv) and O_3 (80 ppbv) did not drift beyond the instruments' specifications during the study, so we did not perform any adjustment to instrument span values during or after the study. All calibrations were performed "through the probe" by connecting a dilution gas calibrator to the sample inlet with a vented tee configuration. A certified NO gas cylinder was diluted to provide a 360 ppbv NO span gas for the NO instrument, and a certified O_3 generator produced 80 ppbv levels of O_3 for the O_3 span checks. We calibrated the NO_2 instruments before and after the study in a laboratory but only performed zero checks on the NO_2 monitors in the field.

The bias of the O_3 instruments varied between 3% and 6% with a standard deviation of 5%. The bias of the NO instrument varied between 3% and 8% with a standard deviation of 6%. The NO gas standard had a concentration uncertainty of ±2% (EPA certified grade). Mass flow controllers in the dilution calibrator were within their certification period and had a specified accuracy of ±3.6% at the conditions we used to generate the span gases. The accuracy of the O_3 generator was of ±1% and was certified less than three months before the study. We attempted to perform gas-phase titration to produce NO_2 span gases in the field, but technical issues prevented us from performing accurate daily span checks on NO_2. We did not observe drift between the pre-study calibration and post-study calibration of the NO_2 instruments, so we used those calibrations for the entire period.

At least one of the cars parked at the CAMP site (within 85 meters) for 15 time periods during the study (Table S1) and at the La Casa site (within 150 meters) for 16 time periods (Table S2). We previously compared measurements among the three equivalently equipped cars (Whitehill et al., 2020) and determined that NO_2 and O_3 measurements for the three platforms agreed within 20% for 73% (NO_2) and 61% (O_3) of 1-second measurements taken when driving together. NO showed higher variability, but still agreed within 20% about 34% of the time. A similar comparison of two similarly Aclima-equipped Google Street View cars in San Francisco

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and Los Angeles also showed excellent car versus car comparability (Solomon et al., 2020). For the purposes of the present analysis, we assume the three cars to be equivalent and interchangeable.

We aggregated the 1-hz mobile measurements to 1 minute (using a mean aggregating function) to put the car measurements on the same timescale as the 1-minute DISCOVER-AQ measurements reported by CDPHE for the CAMP and La Casa sites.

3.2 Results and Discussion

Figure 1 shows scatterplots comparing 1-minute O_3, NO_2, NO, and O_X for the parked cars versus the fixed reference sites (black circles), as well as the period-specific mean data (yellow circles) and the one-to-one (1:1) line (red dashed line). We calculated the period-specific means by averaging the discrete 1-minute measurements over the continuous measurement periods that the car was parked at the fixed reference site (Tables S1 and S2). We also calculated period-specific medians (i.e., median of discrete 1-minute measurements aggregated over the continuous measurement period) and computed ordinary least squares (OLS) regression statistics for the 1-minute data, the period-specific means, and the period-specific medians (Table 1).

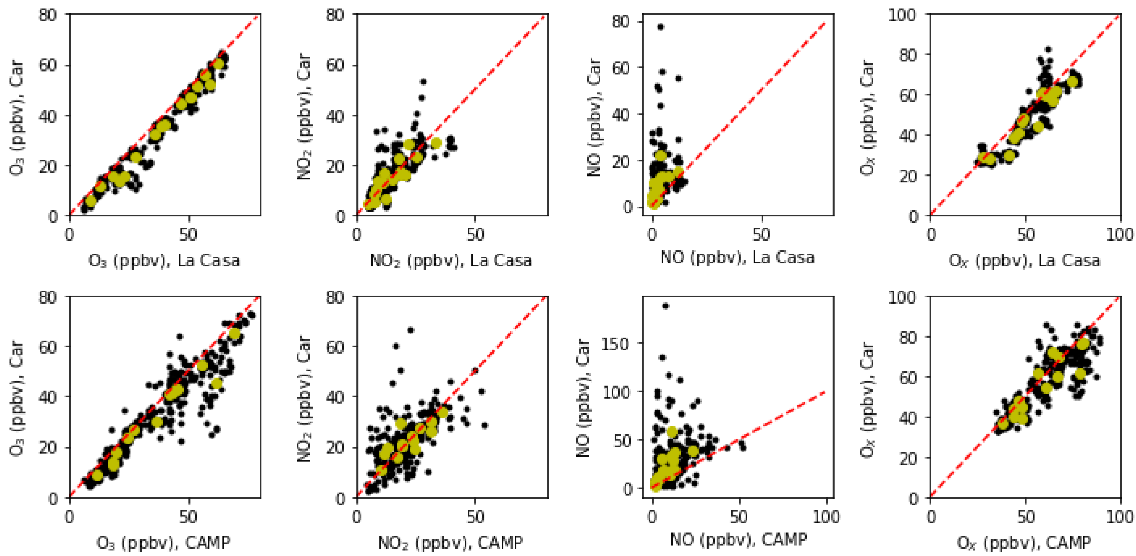

Figure 1: Scatterplots of 1-minute (black dots) and period mean (yellow dots) car measurements (O_3, NO_2, NO, and O_X) versus fixed reference site (La Casa or CAMP) measurements during stationary collocation periods. A red dashed 1:1 line is provided for reference.

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Table 1: Regression statistics for O_3 (ppbv), NO_2 (ppbv), NO (ppbv), and O_X (ppbv) for the parked mobile platform at a fixed reference site (either La Casa or CAMP) during the 2014 Denver study.

	La Casa			CAMP		
	Slope	Intercept	r^2	Slope	Intercept	r^2
O_3						
1 Minute	1.013	-4.12	0.967	0.873	0.51	0.858
Mean	1.020	-4.47	0.988	0.921	-0.91	0.950
Median	1.023	-4.50	0.991	0.960	-2.29	0.960
NO_2						
1 Minute	0.838	2.49	0.596	0.652	8.42	0.399
Mean	0.867	2.12	0.775	0.672	7.78	0.674
Median	0.814	2.78	0.779	0.749	5.82	0.767
NO						
1 Minute	1.164	4.61	0.149	1.099	13.29	0.149
Mean	1.252	4.46	0.442	1.746	6.72	0.445
Median	0.993	3.59	0.405	1.482	7.15	0.341
O_X						
1 Minute	0.932	-0.03	0.828	0.816	7.81	0.708
Mean	0.958	-1.39	0.885	0.893	3.26	0.824
Median	0.932	-0.82	0.918	0.872	4.25	0.839

Of the four pollutants analyzed, O_3 produced the highest coefficients of determination (r^2), with 1-minute mean r^2 = 0.97 for theLa Casa site and r^2 = 0.86 for the CAMP site. The coefficients of determination for 1-minute mean NO_2 and NO were significantly lower, with r^2 = 0.60 for NO_2 and r^2 = 0.15 for NO at the La Casa site. The CAMP site had a lower r^2= 0.40 for NO_2 and similar values for NO. The r^2 values for O_X regressions fall between those for O_3 and NO_2 with r^2 = 0.83 for La Casa and r^2 = 0.71 for CAMP. In the case of all four species, the aggregated (period mean and period median) regression analysis produced higher r^2 values than the 1-minute data with NO_2 and NO showing significantly greater values of r^2 for the period aggregated data than for the 1-minute data. This illustrates how data smoothing can reduce the influence of extreme outlier points on the regression statistics (Brantley et al., 2014).

We anticipated agreement between the parked car and the fixed regulatory site to be strongest for O_X, since direct NO emission from mobile sources cause hyperlocal variations in O_3, NO, and NO_2 (due to rapid O_3 – NO_X titration), but O_X remains constant in the absence of photochemical O_3 production or direct emissions of NO_2 or O_3. Photochemical O_3 production is unlikely to be a major factor for the spatial scales (10 – 100 m) and time scales (1 minute) of this comparison. We do not anticipate direct O_3 emissions in an on-road environment. Although direct NO_2 emissions are possible, especially from heavy duty diesel traffic, previous work has constrained them to be about 5.3% of total NO_X emissions in the Denver, CO region during the time period of this study (Wild et al., 2017).

The La Casa site is located over 80 m from the nearest street in a predominantly residential neighborhood (Figure 2). The CAMP site, in contrast, is located within meters of the intersection of two major roads (Broadway and Champa St.) and is surrounded by commercial properties (Figure 2). We anticipate a heavier influence from direct traffic emissions on the vehicle-based measurements at the CAMP site than at the La Casa site. The major sources of discrepancies between the mobile platform and the fixed reference site are the relative biases in instrument calibration, relative biases in the locations of the instruments being compared, and higher influence of concentrated direct emission plumes at the mobile platform. The dominant source of transient direct emission plumes in this study were on-road vehicle exhaust emissions. The comparisons at the busier intersections (CAMP) had a greater influence from traffic emissions than at the lower traffic (La Casa) site. Agreement between the car and the fixed site was stronger for the La Casa site than at the CAMP site, despite the larger car-to-site distances (Tables S1 and S2).

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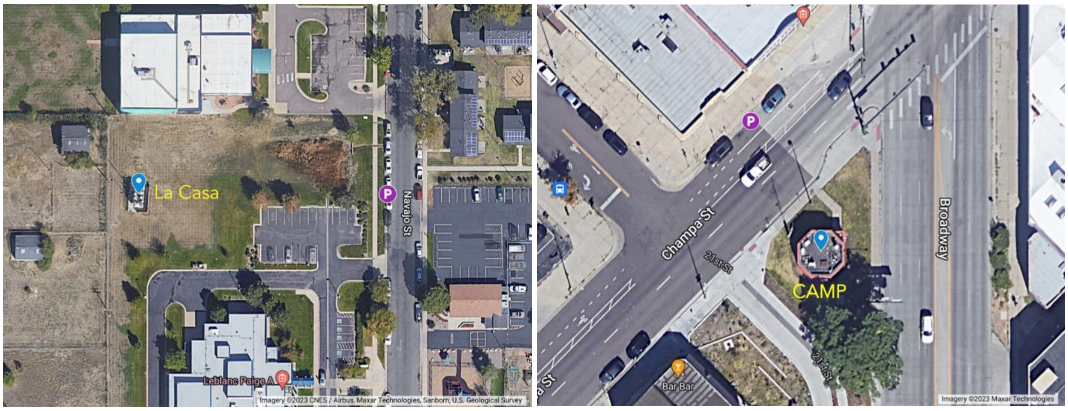

Figure 2: Satellite view of the area in the immediate vicinity of the La Casa (left) and CAMP (right) regulatory sites. The blue markers denote the regulatory sites, whereas the purple markers note the approximate parking location of the mobile platform for the parked collocations. Car-to-site distances varied between 80 to 145 meters for the La Casa site and between 10 and 85 meters for the CAMP site.

In addition to looking at regression statistics, we also looked at car-versus-fixed site differences, ΔX (parked mobile monitor - fixed reference site). We took the median 1-minute averaged ΔX for each individual collocation period, resulting in a ΔO_3, ΔNO_2, ΔNO, and ΔO_X for each of the 16 La Casa collocation periods and the 15 CAMP collocation periods. The distribution of these (Figure S1) shows a typical median difference of about $\Delta O_3 \approx -3$ ppbv and $\Delta NO_2 \approx 1$ ppbv, but also variability between different periods. The period-median ΔO_3, ΔNO_2, and ΔO_X distributions were similar between the two fixed reference sites (Figure S1), indicating more stable regional pollution between the two sites with variability impacted by local sources.

4. Mobile platforms driving around a fixed reference sites in Denver (2014)

4.1 Methods

Coordinating direct side-by-side comparisons between two or more parked vehicles or between a parked vehicle and a fixed reference site can be logistically challenging and time consuming, especially when many cars are involved. Aside from the logistical challenges, parked collocation in urban environments is often impacted by emission plumes from mobile sources, adding additional biases. Comparing to fixed monitoring sites during routine driving operations beyond parked collocations could improve long-term performance assessments of mobile measurements and reduce the logistical burden for large-scale mobile monitoring campaigns. "Rendezvous" collocations provide scalability advantages compared with parked collocations and allow for more campaign time to be dedicated to collecting hyperlocal mobile monitoring data versus dedicated solely to quality assurance.

To explore the feasibility of mobile platform versus fixed reference site comparisons during routine mobile monitoring operations, we analyzed the mobile car measurements during the entire 2014 Denver study (while stationary and moving) in reference to the fixed reference site measurements at the CDPHE La Casa and CAMP sites. Raw 1-Hz car data were associated with the nearest road using a modified "snapping" procedure (Apte et al., 2017), where each mobile datapoint was associated with the nearest 1-meter road segment whose direction was approximately parallel (i.e., within 45°) to the car's heading.

We assigned each 1-hz datapoint one of four different road types ("Residential", "Major", "Highway", or "Other") based on the OpenStreetMap (OSM) road classifications (Table 2) of the nearest road segment identified during the snapping procedure. We also

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created an aggregate road class, "Non-Highway", which consisted of the union of the Residential, Major, and Other road classes (e.g., everything that was not in the "Highway" road class). Based on our results from Section 3, we predicted that measurements made on lower traffic roads (such as Residential roads) would have stronger correlations to measurements at fixed reference sites, especially those located away from major roads or intersections, than higher traffic roads (such as Highway roads). We also anticipated that the distance between the fixed reference site and the mobile platform would affect correlations, with the highest correlations when the distances were small (e.g., less than 1 km) and larger differences as the distance increased. We focused on assigned road type and car-versus-site distance because of their expected importance in performing these regression analyses. These two parameters can also be determined algorithmically using global positioning system (GPS) and OSM data, facilitating data analysis.

We subdivided the dataset into the 4 different road classes (plus the aggregate non-highway class). For each road class, we took subsets of the data within 100 m, 300 m, 1000 m, 3000 m, and 10000 m of the La Casa site and performed an OLS regression analysis of the 1-minute averaged data from that subset. The distance classes were chosen to be 5 approximately equally spaced values on a logarithmic scale between 100 m and 10000 m, which we determined to be an appropriate range for the given datasets. The La Casa site was chosen for this analysis because it provided better absolute agreement for the side-by-side parked collocations (Figure 1, Table 1).

4.2 Results and Discussion

Regression results for different road type and distance class combinations are shown in Figure 3 for O_3 and Figure 4 for NO_2. Similar figures for NO (Figure S2) and O_X (Figure S3) are provided in the supplemental information. The La Casa site is in a residential area, so most of the road types within 300m of the site are Residential or Major. The designed drive patterns within the 5 km radius around reference sites resulted in most of the mapped roads designated as Residential. Further away from the La Casa site, the roads consisted of a larger fraction of major roads and highways that were used to commute to the different areas where the denser mapping occurred.

Table 2: Relationship between OSM road classifications and the road type designations we assigned the segment for the purposes of the current analysis.

Study Road Type	OSM Highway Designation
Highway	motorway motorway_link trunk trunk_link
[1]**Major**	primary primary_link secondary secondary_link tertiary tertiary_link
[1]**Residential**	living_street unclassified
[1]**Other**	service unclassified

[1]Major, Residential, and Other roads are also included in the aggregate road class "Non-Highway"

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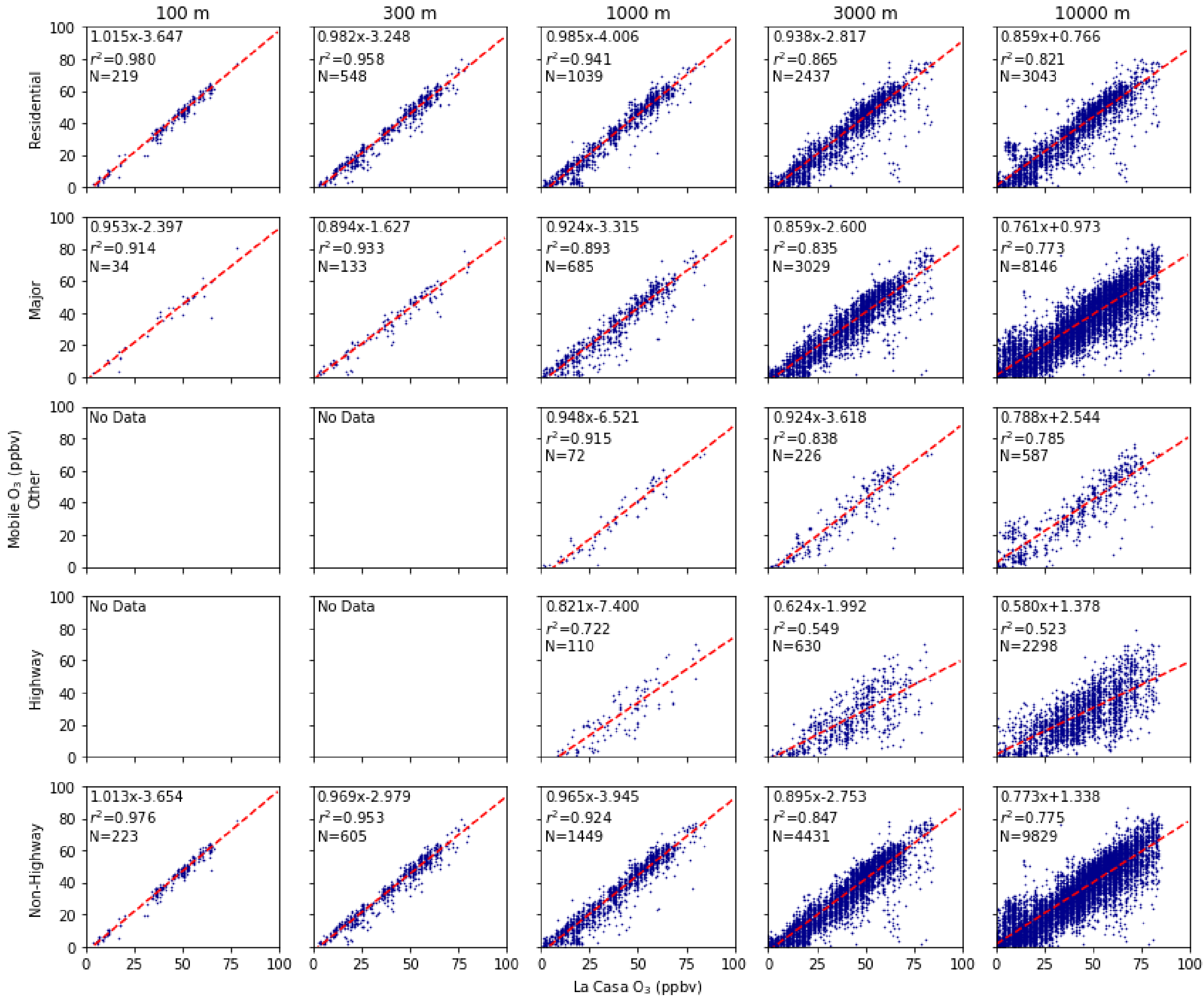

Figure 3: Scatterplots of 1-minute average mobile platform versus 1-minute fixed reference site O_3 for different road classes within five distance buffers of the fixed reference sites in the Denver study. The red dashed lines show the result of the ordinary least squares regression.

The concentrations of NO_2 (Figure 4) and NO (Figure S2) measured by the mobile platform while driving on Highways were higher and O_3 concentrations lower (Figure 3) than the fixed reference site relative to other road types due to the pervasiveness of mobile sources on highways. The influence of near-source vehicle emissions on the concentrations measured complicates stationary-versus-mobile comparisons on highways, as typical urban regulatory monitoring sites (excluding near roadway sites) are intentionally located away from significant major sources.

The results show good correlations for O_3 and O_X, modest to poor correlations for NO_2, and poor to no correlation for NO. Overall, the correlation between the mobile measurements and the stationary site data was highest for the data collected closer to the site, decreasing as data from increasingly farther away from the site is included. For all but NO, the correlations started to show marked decrease in correlation between 1000 m and 3000 m buffer radius distance. Measurements collected on Highways start to be included in the comparison at the 3000m distance, which is likely an additional factor contributing to the decrease in correlation at that distance.

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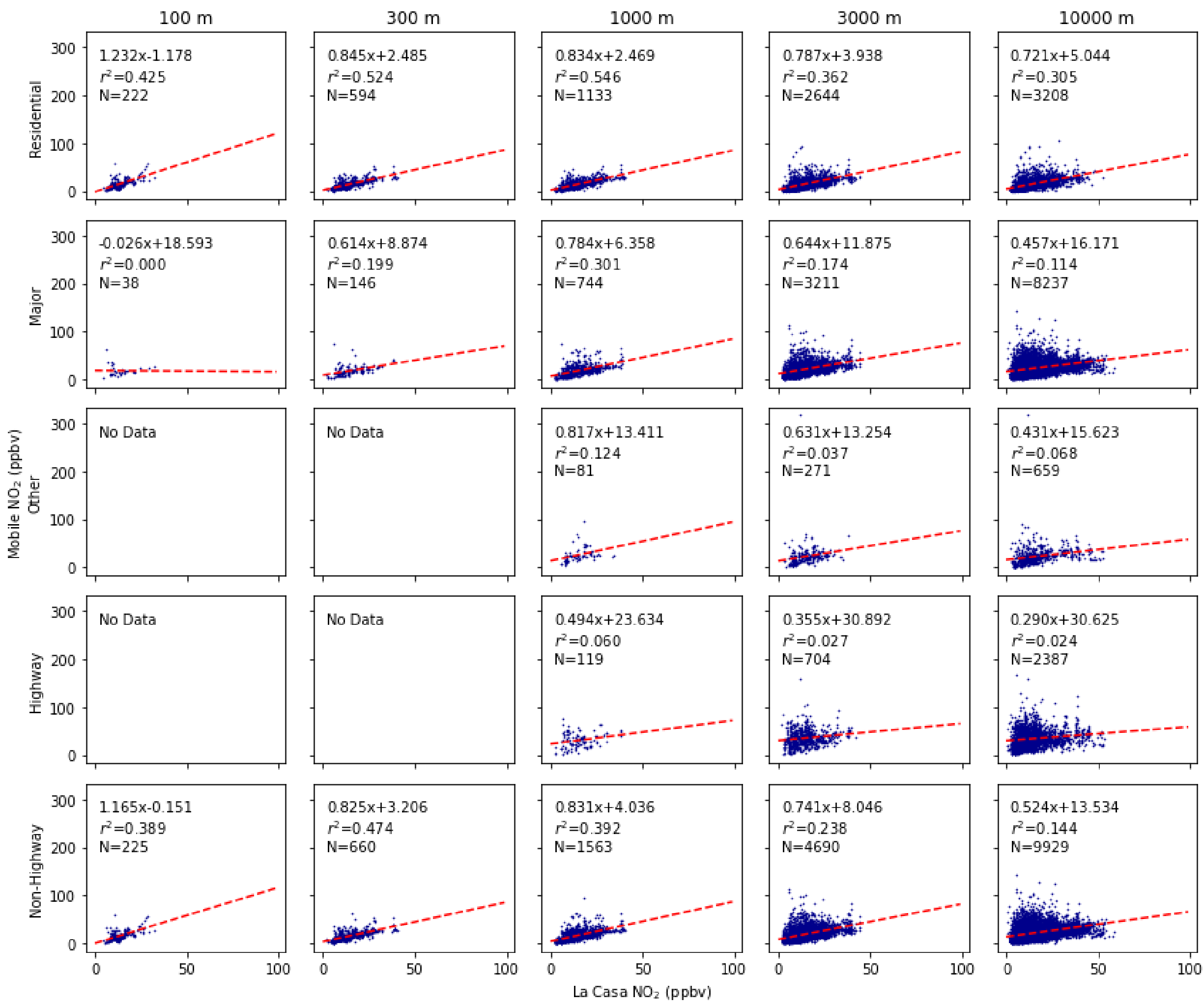

Figure 4: Scatterplots of 1-minute average mobile platform versus 1-minute fixed reference site NO_2 for different road classes within five distance buffers of the fixed reference sites in the Denver study. The red dashed lines show the result of the ordinary least squares regression.

We found that Residential roads provide the best comparability with the La Casa site at smaller car-versus-site distance classes. For O_3 at buffer radii of 1000 m or less, the mobile measurements had excellent correlation with the stationary data, with r^2 values between 0.98 and 0.94, as well as slopes close to 1 and an intercept of around -4 ppb. The correlation between mobile and stationary for NO_2 was modest, with r^2 values between 0.42 to 0.55 for radii of 1000 m or less, with slopes ranging from 1.23 to 0.83 and intercepts between 1.2 and 2.4 ppb. Results for O_X fell between those of O_3 and NO_2. Correlations for NO were generally poor, with a clear influence of emission plumes measured by the mobile platform over a floor of low atmospheric background concentrations. This is consistent with O_3 (and to some extent NO_2) being predominantly regional pollutants, which should vary on the scale of kilometers when not influenced by hyperlocal sources. In contrast, NO is a localized pollutant (especially in the presence of O_3 and other oxidants) and high NO concentrations are mostly driven by source emission plumes.

Depending upon the study objectives and driving patterns, limiting comparisons to just Residential roads can restrict the amount of data. We found that "Non-Highway roads" (including all Residential, Major, and Other road types) also provide useful comparisons

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between mobile and fixed site measurements and significantly expand the amount of available data in the present dataset. For La Casa at smaller car-versus-site distance classes, the regressions statistics were not substantially worse than for Residential roads only, particularly for distance classes of 1000 m or less. It is important to exclude Highways (and potentially other high-traffic roads) when doing this analysis to avoid the potential significant bias resulting from emissions from heavy duty vehicles and a high portion of sampling emission plumes versus background air sampling. We believe that while excluding highways may limit the dynamic range of pollution concentrations for direct emissions (e.g., NO and NO_2) in the analysis, it provides a more robust mobile-to-fixed reference site comparison for the purpose of verifying mobile platform sensor performance during deployment. This approach facilitates the automation of quality assurance check procedures by allowing algorithms to decide (based on OSM street classification and mobile platform GPS coordinates) which data to use in a comparability analysis.

To assess the suitability of mobile-to-fixed "collocation" for use in assessing instrument performance as part of regular mobile monitoring activities, we compared the results with those from the stationary collocations presented in Section 3. For this comparison, we used the results from the Residential and "Non-Highway" correlations for distance classes of 1000 m or less. As discussed previously, the driving routes designed for this study included time parked near the La Casa stationary site as part of the coordinated mobile mapping around the site. As a result, most of the mobile-to-fixed data within 1000 m of the site was measured on the same day and generally within a 1-to-2-hour window of the stationary collocation data. Table 3 lists the statistics for the 1 minute parked "stationary" comparison as well as the 1-minute mobile-to-fixed comparisons for the Residential and Non-Highway at distance classes of 300 m and 1000 m. The stationary and mobile-to-fixed results for the slope, intercept, and r^2 using the Residential data only are very similar within each pollutant. The regression statistics that include the Non-Highway data are also similar. The relatively small change in the intercepts for the Non-Highway may reflect the influence of traffic emissions, likely from Major roads. For instance, the intercept for NO_2 increases from 2.5 ppb to 4 ppb.

Table 3: Regression statistics for the mobile platform compared with the La Casa reference site for both parked, stationary collocations and when driving within a 300 m and 1000 m radius away from the reference site. The stationary statistics are shown for 1-minute averaged mobile data compared with 1-minute regulatory data and mean value for each collocation event. Regressions statistics for the mobile-stationary comparison are shown for mobile and stationary data averaged for 1-minute and 1-hour.

	O_3 (ppbv)			NO_2 (ppbv)			NO (ppbv)			O_X (ppbv)		
	Slope	Intercept	r^2	Slope	Intercept	r^2	Slope	Intercept	r^2	Slope	Intercept	r^2
1 min stationary	1.013	-4.12	0.967	0.838	2.49	0.596	1.164	4.61	0.149	0.932	-0.03	0.828
1 min Res 300m	0.928	-3.248	0.958	0.845	2.49	0.524	1.209	5.943	0.115	0.925	0.524	0.863
1 min Res 1000m	0.985	-4.01	0.941	0.834	2.47	0.546	1.156	6.809	0.086	0.965	-2.626	0.883
1 min Non-Hwy 300m	0.969	-2.979	0.953	0.825	3.21	0.474	1.257	8.799	0.055	0.927	0.045	0.851
1 min Non-Hwy 1000m	0.965	-3.945	0.924	0.831	4.04	0.392	1.583	12.06	0.048	0.945	-0.715	0.846
Mean Stationary	1.02	-4.47	0.988	0.867	2.12	0.775	1.252	4.46	0.442	0.958	-1.39	0.885
1 Hour Res 300m	0.985	-3.7	0.981	0.953	1.15	0.724	1.792	7.596	0.204	0.96	-1.485	0.932
1 Hour Res 1000m	0.983	-4.1	0.967	0.902	2.05	0.609	1.287	7.908	0.223	0.978	-3.133	0.934
1 Hour Non-Hwy 300m	0.945	-2.96	0.968	0.88	3.531	0.573	1.555	13.6	0.159	0.963	-1.23	0.921
1 Hour Non-Hwy 1000m	0.959	-4.152	0.956	0.972	3.134	0.559	1.756	15.5	0.13	0.943	-0.018	0.918

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Figure 3 indicates that O_3 tends to be lower at the mobile monitoring platform than the fixed reference site (for La Casa), whereas NO and NO_2 are higher at the mobile platform. If we consider the fixed reference site to be representative of "urban background" pollutant levels, direct combustion emission sources (including mobile sources) are likely to increase NO and NO_2 on a local scale near the mobile platforms, while titrating O_3. The directionality of the mobile versus fixed site discrepancies from this comparison is the same as that when the mobile platform was parked at the fixed site (Figure 1). Comparison of the results of parked to mobile comparison with stationary should enable us to assess the feasibility of using mobile platform versus fixed reference site comparisons in the assessment of mobile measurement performance. For distances less than ~2 km and on residential roads, the bias from the mobile-stationary comparison for O_3 is between -3 and -4 ppbv, similar to the intercept of -4.1 ppbv from stationary collocation (Table 1). The results for NO_2 and NO for residential roads are also similar, with mobile-stationary bias of ~2 ppbv and ~5 ppbv, respectively, compared to intercepts of 3.2 ppbv and 4.7 ppbv. These results suggest that mobile-stationary comparisons may be suitable for assessing bias in sensor performance, although additional verification of this is necessary.

The availability of data from a fixed reference site at a 1-minute time resolution was unique to the experimental study in Denver, with additional instrumentation added to support the research objectives of the 2014 DISCOVER-AQ experiment (https://www-air.larc.nasa.gov/missions/discover-aq/discover-aq.html). Data from regulatory monitoring stations in the US will typically only be available at 1-hour time resolutions. For mobile-to-fixed "collocation" to be broadly applicable to large-scale mobile monitoring campaigns, it is important to assess how the correlation results change when the mobile data is compared with 1-hour stationary data. We averaged the mobile data by taking the mean of 1-second measurements within each hour period that fit the appropriate criteria (e.g., road type, distance, etc.). The results are compared to the correlations from the period mean of 1-minute stationary collocation data (Table 1). These period means are averages of approximately 10 to 20 minutes of data, which is the closest comparison of averaging time possible from the stationary data. The results are also shown in Table 3 for the same Residential and Non-Highway correlations for distance classes of 1000 m or less. As with the 1-minute data, the regression statistics from the 1-hour data are similar within each pollutant for the 300 m and 1000 m compared to stationary. The results for O_3 are more-or-less identical. The correlation for NO_2 improved with time averaging, with the 1-hour r^2 results for the mobile-to-fixed data increasing to a range of 0.72 to 0.56 for the 1-hour averaged data from a range of 0.55 to 0.39 for the 1-minute data for different road type and distance classes. The results imply that averaging the data over longer time periods reduces the influence of high concentrations from individual plumes on the comparison, improving the correlation.

These results indicate that mobile-to-fixed comparisons are similar to those resulting from stationary parked collocation and that the use of mobile-to-fixed comparisons may be suitable for assessing sensor performance. One key question in operationalizing such a comparison is the best distance buffer and the number of measurements sufficient for performance assessment of mobile monitoring measurements. Increasing the distance buffer increases the number of measurements available for the analysis, but the correlations and thus comparability are stronger for the smaller buffer distances. When checking sensor performance (e.g., stability, potential sensor issues) the buffer distance may not be as important as when wanting to quantify sensor bias. The data set from Denver was collected during summer and over a limited number of days, and thus will not reflect the influence of seasonal variability in concentrations. To further assess the suitability of mobile-to-fixed method for sensor assessment, we extended our analysis to a second dataset based on one full year of driving in the San Francisco Bay Area between 2019 and 2020.

5. Mobile platforms driving around fixed reference sites in California (2019 – 2020)

5.1 Methods

Aclima-operated fleet vehicles are equipped with a mobile sensing device, the Aclima Mobile Node, which measures CO, CO_2, O_3, NO, NO_2, $PM_{2.5}$, and TVOC. As part of the AMN calibration procedure, up to 16 AMNs at a time are deployed in the Aclima Mobile Calibration Laboratory (AMCL), a gasoline-powered Ford Transit van equipped with laboratory-grade air pollution measurement instrumentation. The AMCL is driven around the San Francisco Bay Area of California to calibrate the sensors within the AMNs through comparison of the AMN response measurements with the laboratory-grade equipment collocated in the same van. As part of the validation process, the AMCL regularly drives around several Bay Area Air Quality Management District (BAAQMD) regulatory monitoring sites (Figure 5). These BAAQMD sites are equipped with EPA-approved reference measurements of NO, NO_2, and O_3 (or a subset of these species), along with other regulatory measurements. We present an analysis comparing the AMCL reference measurements with nearby BAAQMD reference measurements between November 2019 and October 2020.

Calibrations using reference gases were performed throughout the collection period to assess bias and precision. The calibration procedures have been described in Solomon et al., (2020). Bias and precision results across approximately 25 zero and span checks are shown in Table 4.

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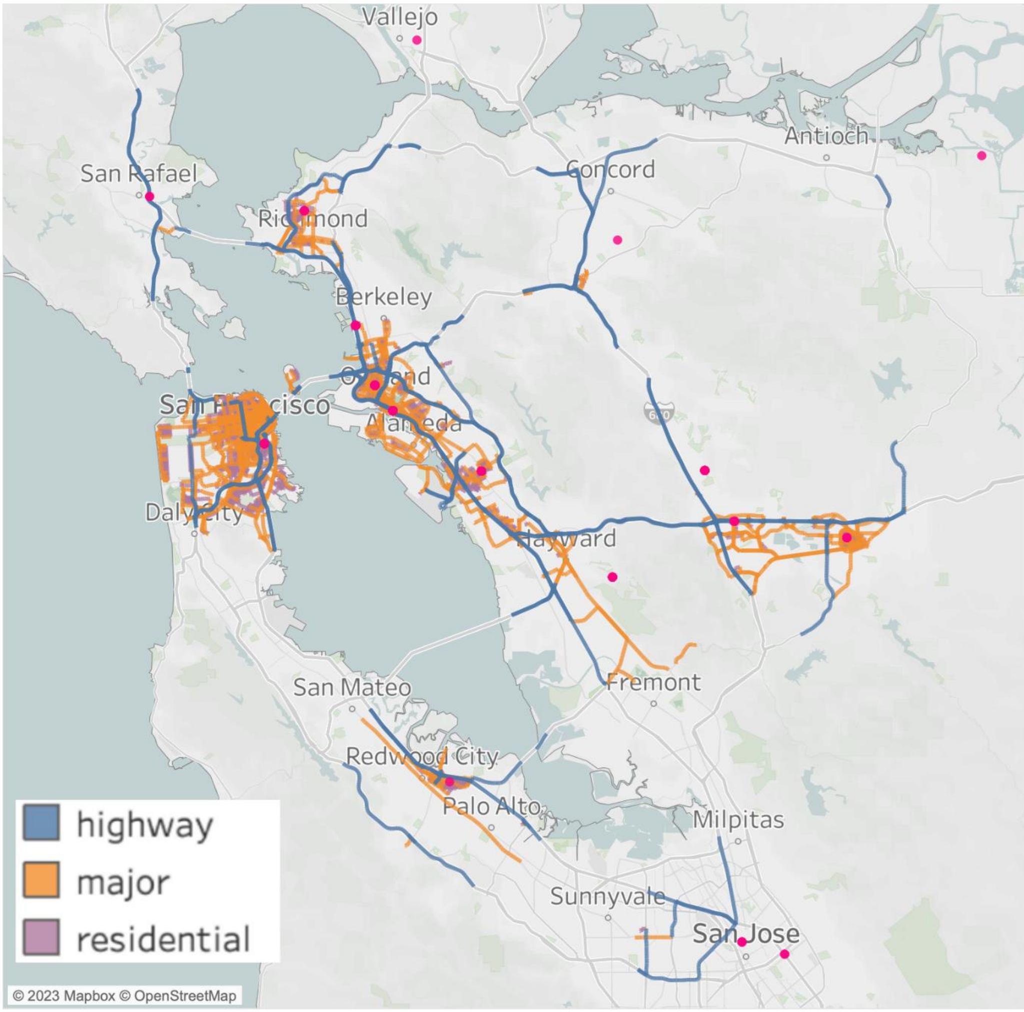

Figure 5: Driving patterns around regulatory sites (red dots) in the San Francisco Bay Area. Different road types driven are illustrated by different colors.

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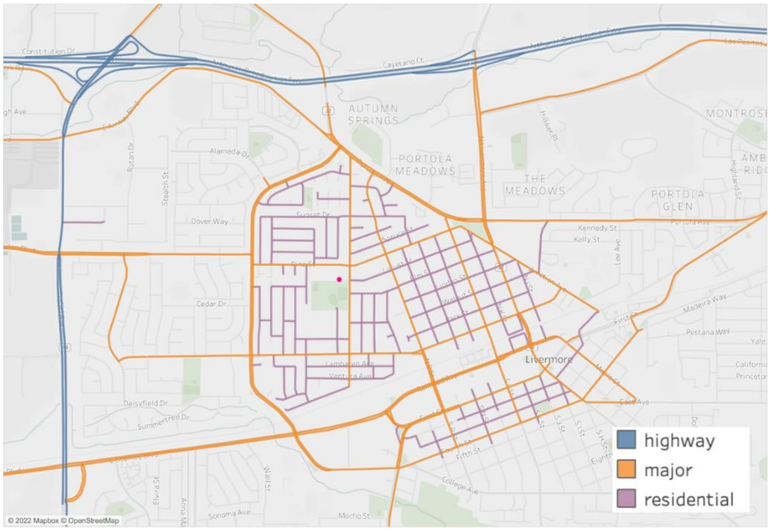

Figure 6: Zoomed in view of the street-by-street Aclima Mobile Calibration Laboratory driving pattern around the Livermore site in the San Francisco Bay Area.

Table 4: Precision and bias of measurements in the Aclima Mobile Calibration Laboratory from approximately 25 QA checks during the San Francisco Bay Area study period. Each QA check consisted of a zero and span point for each pollutant.

Pollutant	Bias		Precision	
	Zero	Span	Zero	Span
NO_2 (ppbv)	0.3	5.6%	0.2	4.3%
NO (ppbv)	0.7	4.4%	0.3	4.4%
O_3 (ppbv)	0.5	2.5%	0.4	2.0%

The dataset included comparisons to 19 different BAAQMD sites that represent a number of different spatial representativeness scales (40 CFR 58 Appendix D), but most of the data was collected near the Livermore, San Francisco, and West Oakland sites (Figure 5). These three stations were selected to represent different climatological and land use regimes in the San Francisco Bay Area – San Francisco (warm summer Mediterranean climate with marine influence, cool winds and fog in summer, overall little seasonal temperature variation, mixed residential with light industrial), Oakland (warm summer Mediterranean climate with marine influence, overnight fog in summer, mixed industrial and residential), and Livermore (hot summer Mediterranean climate, inland with some marine influence, residential, upwind of a large fraction of the urban Bay Area emissions). In contrast to the mapping performed in Denver, measurements near these monitoring sites generally involved mapping essentially all publicly available streets within a few kilometers of the site (Figure 6). Measurements near other regulatory sites are also included and were generally chance encounters due to the AMCL driving past sites on its way to its daily mapping assignments. As a result, this data tends to be from highways or major roads and not mapped on a street-by-street basis.

Based on results from the 2014 Denver dataset (Section 4), we focused our analysis on two specific road type scenarios – Residential roads and Non-Highway roads. Because the BAAQMD measurements, like most regulatory gas-phase measurements in the US, are reported at 1-hour time resolution, we focused on 1-hour aggregated comparisons. We aggregated each subset of AMCL measurements up to one hour using the median as an aggregating function. We chose to use the median (versus the mean) to minimize the influence of outliers caused by unusual emissions or environmental conditions. The fraction of each 1-hour collocation period that included measurements fitting the defined criteria varied depending upon the buffer distance and road type subset (Figures S4 and S5).

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For smaller distance buffers (e.g., 100 m or 300 m) or more restrictive road subsets (e.g., the Residential subset), the distribution was skewed towards a smaller fraction of measurements within each hour fitting the criteria for the comparison. For distance buffers 1 km and higher on Non-Highway roads, the distribution was approximately uniformly distributed in the 0 – 100% range. No minimum number of data points were required in each hourly average, such that anywhere between a few seconds and a full hour of mobile platform data may have been considered in each hourly average.

5.2 Results and Discussion

Regression analysis of 1-hour aggregated mobile platform versus fixed reference site measurements for the Non-Highway roads is shown in Figure 7 for O_3, NO_2, NO, and O_X, and ordinary least squares regression statistics (for both the Residential and Non-Highway subsets) are provided in Table 5. O_3 and O_X have strong correlations ($r^2 > 0.90$) for both Residential and Non-Highway roads within all buffer distances up to 3 km, and the 10 km buffer distance has r^2 values that are only slightly lower (above 0.88, Table 5). For all buffer distances, O_3 and O_X produced slopes within the range of (0.95, 1.05) and intercepts in the range of (-2.1, 1.6). NO_2 regressions produced lower r^2 values than O_3 and O_X, ranging between 0.62 and 0.85. Ordinary least squares regressions from NO_2 produced slopes within 5% of 1.0 for Residential roads within 100 m, 300 m, and 1 km buffer distances and for Non-Highway roads with a 100 m buffer distance.

These results suggest that comparisons of O_3, NO_2, and O_X provide the potential for ongoing calibration or performance assessment of instruments within on-road mobile platforms, especially if the data are pre-filtered based on certain criteria (e.g., road type and maximum distance from the fixed reference site). While NO may also be suitable, the current data set is limited by the range of measured NO concentration and the correlations are likely dominated by higher concentrations from mobile sources, so a reliable assessment cannot be made.

Table 5: Regression statistics, median ΔX values (mΔX) and number of measurements in each dataset for 1-hour median mobile measurements versus 1-hour stationary measurements for data collected in the San Francisco Bay Area.

		Residential Roads					All Non-Highway Roads				
Species	Distance	Slope[1]	Intercept[1]	r^2	*mΔX[1]	N	Slope[1]	Intercept[1]	r^2	*mΔX[1]	N
O_3	100 m	1.02	0.02	0.943	0.70	206	1.03	-0.23	0.963	0.80	469
O_3	300 m	1.04	-0.98	0.963	0.90	704	1.03	-0.71	0.953	0.57	786
O_3	1 km	1.04	-0.97	0.965	0.80	903	1.03	-1.06	0.957	0.50	1041
O_3	3 km	1.03	-0.58	0.935	0.70	1133	1.03	-2.04	0.910	-0.20	1652
O_3	10 km	1.00	0.61	0.890	0.80	1536	1.01	-0.62	0.889	0.00	1920
NO_2	100 m	0.96	0.66	0.771	0.32	351	0.98	0.72	0.759	0.34	608
NO_2	300 m	1.05	0.21	0.850	0.27	812	0.89	0.81	0.809	0.24	950
NO_2	1 km	0.97	0.39	0.801	0.20	1074	0.95	0.87	0.787	0.37	1285
NO_2	3 km	0.76	1.11	0.725	0.09	1336	0.88	1.76	0.641	0.61	1908
NO_2	10 km	0.81	0.83	0.628	-0.10	1755	0.86	1.33	0.627	0.37	2164
NO	100 m	1.02	2.06	0.284	1.20	312	1.11	1.98	0.293	1.10	545
NO	300 m	1.15	1.77	0.200	0.95	738	0.65	2.39	0.228	0.65	862
NO	1 km	0.76	2.05	0.414	0.60	970	0.65	2.63	0.298	1.00	1150
NO	3 km	0.35	2.52	0.214	0.10	1206	0.47	2.92	0.307	1.20	1703
NO	10 km	0.43	2.50	0.143	-0.10	1587	0.47	2.61	0.270	0.60	1939
O_X	100 m	1.03	0.30	0.944	1.53	199	1.00	1.54	0.958	1.43	461
O_X	300 m	1.02	0.03	0.961	1.27	691	1.00	1.00	0.946	1.09	771
O_X	1 km	1.03	-0.23	0.962	1.16	890	1.01	0.32	0.961	1.06	1024
O_X	3 km	1.03	-0.23	0.944	1.11	1108	1.01	0.49	0.922	1.01	1623
O_X	10 km	1.03	-0.58	0.909	0.91	1503	1.01	-0.10	0.894	0.82	1888

* mΔX is the median ΔX value.
[1] units are in ppbv.

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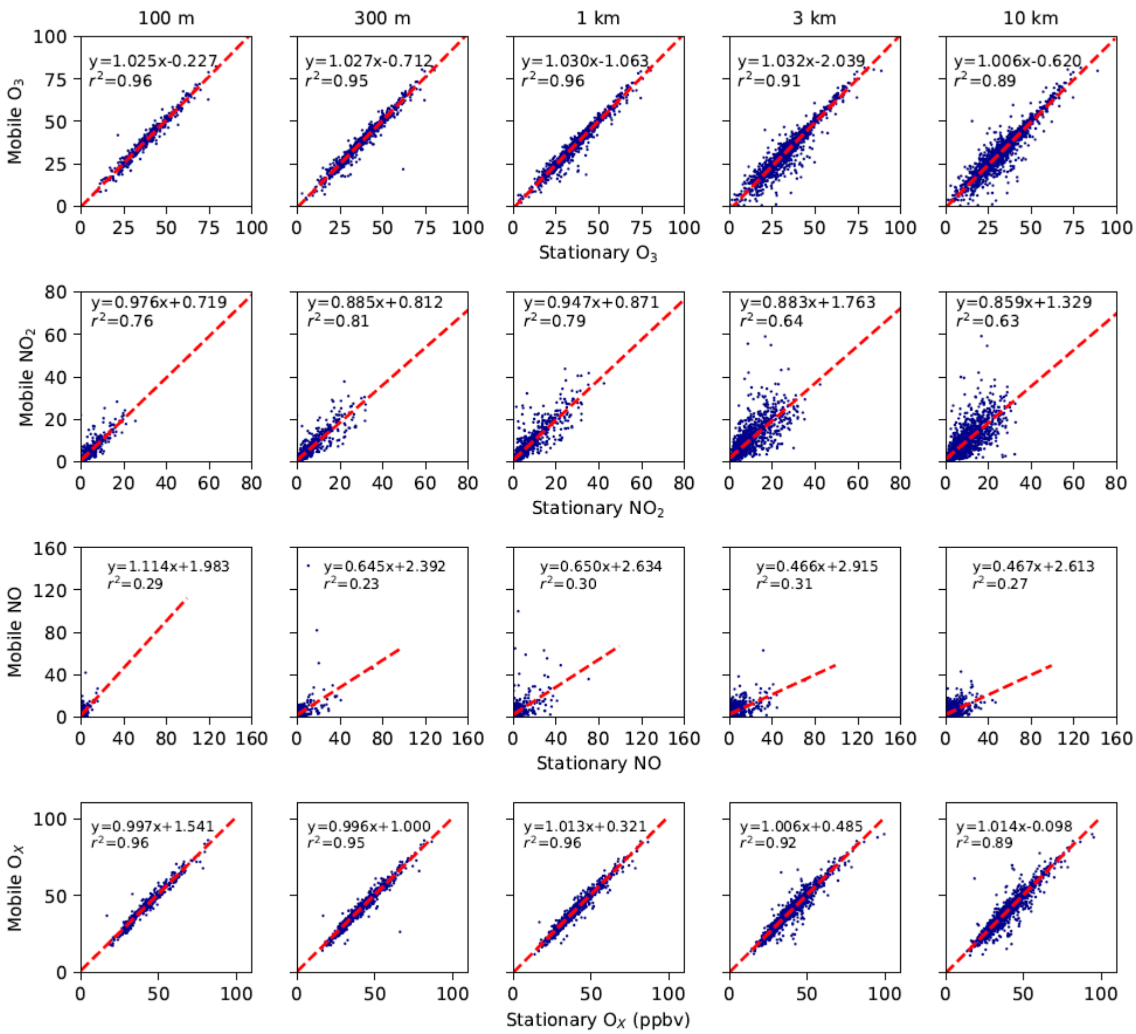

Figure 7: Scatterplots of 1-hour median mobile platform versus 1-hour fixed reference site O_3, NO_2, NO, and O_X for Non-Highway roads within five distance buffers of the fixed reference sites in the California study.

5.3 Using driving data for ongoing performance evaluation

We considered the possibility of using mobile-versus-fixed-site measurements for calibration corrections, using metrics such as running slopes and intercepts of ordinary least squares linear regressions to provide a "correction" model to apply to the instrument data from the mobile monitoring platform. Many parametric linear regression methods (such as ordinary least squares linear regression) can be significantly influenced by outliers, such that any running regression statistics would have the potential to produce anomalous calibrations if influenced by any outliers in the dataset. The statistical distribution of absolute differences (e.g., ΔX values) can also contain outliers due to the influence of spatial variability in pollutant concentrations during certain sampling periods. When we compare the mobile platform to the fixed reference site, we want to allow for a situation where they agree most of the time (to within a certain amount), but also have a method that allows for occasional outliers, some of which may be large. As a first pass at better understanding whether this approach was feasible and how it would apply, we considered looking at running median ΔX values and whether they could be used as an ongoing quality assurance indicator during long measurement campaigns.

Now that we have shown that comparisons between moving mobile platforms and fixed reference sites show excellent agreement for O_3, NO_2, and O_X, we would like to use this knowledge to explore a method for using mobile platforms versus fixed reference site measurement differences as an ongoing quality assurance indicator. We envision a method that can provide ongoing assessment of either instrument malfunctions or drift during deployment. For the larger California data set, the median of the hourly ΔX values (mΔX in Table 5) were within 1.6 ppbv of 0 and tended to be positive (except for three values $-0.2 \leq m\Delta X < 0$). We considered a method whereby the mobile platform versus fixed reference site measurement differences are used for ongoing quality assurance checks. We looked at running medians of hourly ΔX values to assess mobile-versus-fixed site comparison over time. The running window size is a trade-off between minimizing the time it takes to collect enough data to detect instrument drift and minimizing the magnitude of drift that can be detected. As the running median window size is increased, the more data can be aggregated into a single bias estimate, giving higher confidence in the estimate between the mobile platform and stationary reference site. However, it will take longer to collect enough measurements to detect changes in the bias. To determine the optimal window size, we calculated the range of running median ΔX values as a function of window size. Figure 8 shows how the range in running median ΔX observed decreases with running window size. After about a 30-40 hour window size, there is a reduction in improvement in the range of running median ΔX values expected for any single running median calculation with increased window size. The range of ΔX values eventually converges at a value around 2 – 3 times the actual bias determined from ongoing calibration checks over the course of the study (Table 4), which are approximately 1-2 ppbv and depend on the pollutant concentration at any given time. This result is consistent for ΔO_3, ΔNO_2, ΔNO, and ΔO_X. Thus, we choose a 40-hour running median window as optimal for the following analysis, where the range of ΔX values is around 3.5 ppbv for NO_2, 6.4 ppbv for O_3, 5.2 ppbv for O_X, and 7.1 ppbv for NO. Larger running window sizes (N) will decrease the range of ΔX values but has a tradeoff in terms of time response to "detect" a drift or malfunction. The number of hour-averaged datapoints in our 1 year dataset for each pollutant and distance class is given in Table 6. For the 3 km distance class, a sample size of ~1600 (O_3) to ~1900 (for NO_2) gives an "effective response time" of about 7 – 9 days (e.g., one week), where the effective response time is the study period (365 days) divided by the number of discrete hour periods (1600 to 1900) times the rolling window size (40). This provides an order-of-magnitude estimate for the length of time necessary for a significant malfunction or large drift to be visible in the data. Note that the number of 1-second mobile measurements contributing to each 1-hour ΔX value in this data set varies (Figures S4 and S5) and is often much less than 3600, so this analysis does not require a full 40 hours of data collection within 3 km.

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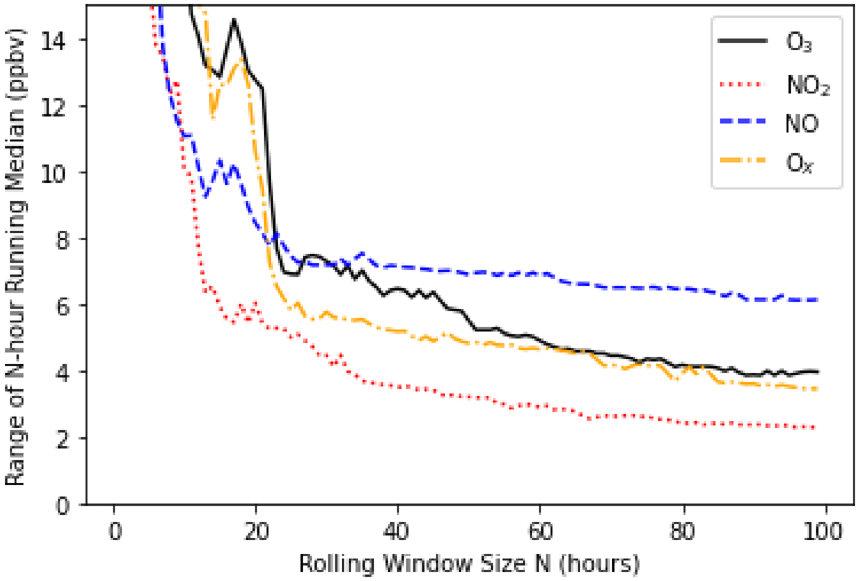

Figure 8: Influence of averaging window size on the range (maximum - minimum) of the running mean ΔO_3, ΔNO_2, ΔNO, and ΔO_X values for 3 km, non-highway roads.

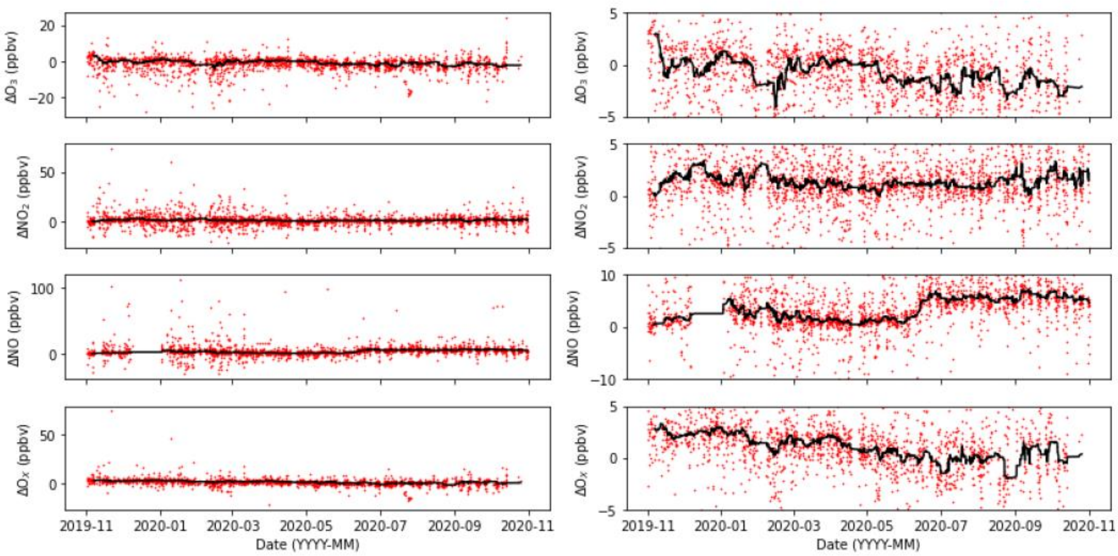

Figure 9: Timeseries of 3 km ΔO_3, ΔNO_2, ΔNO, and ΔO_X, showing individual 1-hour measurements (red points) and 40-sample running median (solid line). The right column truncates many of the 1-hour measurements and scales the axis to highlight the running median.

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Table 6: Number of hour-averaged measurements in the California dataset (non-highway roads).

Distance Class	O_3	NO_2	NO	O_x
100 m	469	608	545	461
300 m	786	950	862	771
1 km	1041	1285	1150	1024
3 km	1652	1908	1703	1623
10 km	1920	2164	1939	1888

We examined the timeseries of ΔO_3, ΔNO_2, ΔNO, and ΔO_X for Non-Highway roads for the 3 km distance class (Figure 8), including both the raw 1-hour data (red points) and a 40-sample (40-hour) running median (black line). Figure 9 shows that, despite the large range of 1-hour ΔX values across the timeseries, the running median tends to converge to a small range around zero, with values within ±4 ppbv for O_3, NO_2, and O_X. For long-term driving campaigns that frequently pass near stationary monitoring sites, this method appears to be a practical way to monitor system performance on an ongoing basis without requiring frequent side-by-side parked collocations.

The method presented here for assessing ongoing performance of air pollution monitors in a moving mobile platform is meant to be a quality assurance indicator to identify potential measurement issues in a scalable way for fleet-based monitoring campaigns. It is not an absolute method for calibration or instrument verification, as a direct collocated comparison with reference monitors is. It provides a useful method for identifying either systematic measurement drift or sudden instrument or sensor malfunction. However, for any chosen threshold value it also has the potential to detect anomalies (e.g., running median values outside the thresholds) even when instruments are operating properly. There are tradeoffs between the running median window size, the threshold values, and sensitivity and specificity of the method for detecting measurement issues. These must be determined empirically for each study based on the study design and the quality assurance requirements.

6. Conclusions

In this paper, we address the issue of ongoing quality assurance during a large-scale mobile monitoring campaign, with a focus on discerning changes in instrument performance over time during mobile platform deployment. To assess instrument drift over time using any sort of collocation or sensor-versus-reference comparison, it is first necessary to constrain the uncertainty inherent in the collocation or comparison process. We used a set of parked and moving mobile monitoring data from a one month study in Denver, CO (2014) and compared reference grade NO, NO_2, and O_3 measurements from a mobile platform to fixed reference site measurements both when the mobile platform was parked side-by-side with the reference site and when driving at distances out to 10 km from the reference site. Using data from a more extensive, 1-year study in California (starting November 2019), we show large-scale comparisons of hourly mean mobile measurements to hourly fixed reference site measurements. We highlight the importance of grouping data based on street type (to remove the influence of highways) and demonstrate a possible data aggregation technique for large-scale, long-term comparisons. Hourly averaged regulatory site data are reported by most state and local air quality monitoring agencies in the US (and several other countries). These hour-aggregated mobile platform measurements show excellent agreement with hour-averaged fixed site measurements using both regression analysis and running medians of moving mobile versus fixed site differences. The work presented here will be extended in the future to examine how these methodologies can be used to assess the ongoing performance of low-cost sensor nodes in mobile monitoring platforms.

Conflict of Interest

The authors declare no conflicts of interest.

Data Availability

Data used for this manuscript is proprietary and owned by Aclima, Inc. (https://aclima.io). Interested researchers are encouraged to contact Aclima, Inc. for data availability and collaboration opportunities. Additional questions about analysis techniques or code can be directed to the corresponding author of this manuscript.

Author Contribution

Conceptualization – ML, SK, and PS
Data Curation – ML, BL
Formal Analysis – AW, ML, BL
Funding Acquisition – ML, BL, PS
Investigation – AW, ML, BL
Methodology – AW, ML, BL, and PS
Project Administration – ML, SK, PS
Resources –ML, BL, PS
Software – AW, ML, BL
Supervision – ML, SK, PS
Validation – ML, BL, and PS
Visualization – AW, ML
Writing – Original Draft – AW, ML, BL
Writing – Review and Editing – AW, ML, BL, PS

Disclaimer

This manuscript has been subjected to internal review at the U.S. Environmental Protection Agency and has been cleared for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use. The views expressed in this article are those of the author(s) and do not necessarily represent the views or policies of the U.S. Environmental Protection Agency.

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