Mobile Air Quality Monitoringair quality monitoring and Comparisoncomparison to Fixed Monitoring Sitesfixed monitoring sites for Quality Assurance instrument performance assessment

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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

- 15 integrate smaller scale air quality sensors, it is important to address the quality assurance needsneed for validatingassessing these measurements, in a scalable manner. We explore collocation-based evaluation of air quality instrumentsmeasurements in a mobile platform againstusing fixed regulatory sites, both when the as a reference. We compare two approaches a standard collocation assessment technique where the mobile platform is parked at near the fixed regulatory site for a period of time and when moving at distances of meters to kilometers from the site. We demonstrate agreement within 4 ppby (for NO2)
- 20 and O₃+NO₂) to 10 ppbv (for NO) when using a running median of 40 hourly differences between the moving an expanded approach using measurements while the 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 in motion in the general vicinity of the analysis. We present a method for assessing mobile measurements fixed regulatory site. Based on the availability of fixed reference site data, we focus on three pollutants (ozone
- 25 $(O_3)_{52}$ nitrogen dioxide $(NO_2)_{53}$ and nitric oxide) with distinct atmospheric lifetimes and behaviors. We compare measurements from a mobile laboratory with regulatory site measurements in Denver, Colorado, USA and odd oxygen $(O_x = O_3 + NO_2)$ on an ongoing basis through comparisons with in the San Francisco Bay Area, California, USA. Our one-month Denver dataset includes both parked collocation periods near the fixed regulatory sites as well as general driving patterns around the sites, allowing a direct comparison of the parked and mobile collocation techniques on the same dataset. We show that the mobile
- 30 collocation approach produces similar performance statistics, including coefficients of determination and mean bias errors, as the standard parked collocation technique. This is particularly true when the comparisons are restricted to specific road types, with residential streets showing the closest agreement and highways showing the most variance. We extend our analysis to a larger (year-long) dataset in California, where we explore the relationships between the mobile measurements and the fixed reference sites on a larger scale. We show that using a 40-hour running median converges to within ±4 ppbv of the fixed
- 35 reference site for nitrogen dioxide and ozone and up to about 8 ppbv for nitric oxide. We propose that this agreement can be leveraged to assess instrument performance over time during large-scale mobile monitoring campaigns. We demonstrate an example of how such relationships can be used during large-scale monitoring campaigns using small sensors to identify potential measurement biases.

40 1 Introduction—

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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). (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).the accuracy of 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-powerpowered instrumentation, is <u>maintaining</u> instrument performance (e.g., accuracy, precision, and bias) <u>over time</u> in a mobile environment. Instrumentation can behave differently in a field environment than during laboratory testing and calibration (Collier Oxandale et al., 2020),(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).(Li et al., 2022). Frequent collocations with fixed reference sites have been identified as an important component of quality assurance for mobile monitoring campaigns (Alas et al.,

2019;Solomon et al., 2020;Whitehill et al., 2020).(Alas et al., 2019; Solomon et al., 2020). Collocated measurements within ongoing campaigns can also be used to identify potential measurement issues. For example, Alas et al. (2019) provide an example of how the collocation of two AE51 black carbon monitors was used to identify and correct unit-to-unit discrepancies during an ongoing mobile monitoring campaign. 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).
 sensors (Bauerová et al., 2020; Castell et al., 2017; Masey et al., 2018).

Parking a mobile platform next to For mobile monitoring applications, parking a mobile platform next to a fixed reference site approximates the collocation technique for assessing instrument performance. Collocated parking can be incorporated into driving patterns for large-scale mobile monitoring campaigns. However, parking near a fixed reference site ensures comparability only at that specific location and only under the specific atmospheric conditions over which the

- 65 collocation occurred. As a result, many repeat collocations must be performed, leading to inefficiencies in data collection and an approach that is not easily scalable to larger mobile monitoring campaigns. In some cases addition, natural variability in pollutant concentrations makes the selection of collocation site important (Alas et al., 2019; Solomon et al., 2020), which can often prove restrictive for extended monitoring campaigns. In addition, strong agreement when collocated at a fixed reference site may not translate directly into accuracy and precision in other environments (Castell et al., 2017;Clements et al., 2017).
- 70 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. (e.g., Castell et al., 2017; Clements et al., 2017).

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.

- 80 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 85 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
- Here, we compare air pollution monitoring instruments in a vehicle based mobile platform to regulatory air quality
 monitoring site reference measurements. The use of Ongoing mobile ("in motion") comparisons with fixed reference sites are more scalable than frequent side-by-side parked collocations and could provide an important tool for ongoing instrument performance assessments during extended mobile monitoring 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 evaluate the mobile measurements will be increased, the dynamic range of pollutant concentrations being measured will be
 larger, and more time can be dedicated to meeting the mobile monitoring data objectives. In this study, we compare mobile air

comparisons, which is particularly important during sustained, multi vehicle (and fleet based) mobile monitoring campaigns.

- pollution measurements to fixed reference site measurements from both parked and mobile collocations during the same campaign. The objective is to determine if changes in instrument performance, such as bias, can be identified in mobile collocations to a similar degree as with stationary collocations. We will look at using mobile-versus-fixed-site comparisons as a function of road type and distance between the vehicle and the fixed reference site and compare them to the parked
- 100 comparisons.

If mobile collocation is able to quantitatively assess bias in mobile instrumentation, it would allow easier detection of instrument drift over time or sudden changes in instrument performance that could indicate a malfunction. This methodology will not serve as a calibration of the mobile platform or replace traditional calibration and quality assurance techniques; rather, it is meant to supplement traditional techniques to allow earlier identification of measurement issues during ongoing mobile monitoring campaigns. We explore the concept of mobile collocations using fast response (1-Hz or 0.5-Hz) laboratory-grade

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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 aimair pollution monitoring instrumentation which is independently calibrated and subject to develop a method to look at the mobile versus fixed site data on a dataset of laboratory grade measurements.strict quality assurance.

- 110 This allows us to explore the impact of spatial variability in pollutant concentrations and operational differences in the mobileversus-fixed-site comparisons without being limited by instrument accuracy or precision concerns. This will help us to understand the strengths and drawbackslimitations of these methods- and to quantify the magnitude of biases that could be detected using these methods. This work eancould be expanded in the future work-to mid-range instruments instrumentation and smaller scale sensors, of various pollutants and eventually further developed into a scalable approach for ongoing quality
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calibrations of sensors using traditional methods is not feasible.

2. Overview of Methods

For this study, we focus on the pollutants ozone (O_3) , nitrogen dioxide (NO_2) , and nitric oxide (NO). This decision is largely based on the availability of both mobile and fixed reference site data for the two studies we analyze. O_3 and NO_2 are

assurance instrument performance assessment during fleet-based mobile monitoring campaigns where frequent in-situ

- 120 Criteria Pollutants with adverse health effects that are measured and regulated by the United States Environmental Protection Agency (USEPA), and all three species are commonly measured and air quality monitoring sites in the United States of America (USA) and other countries. The data collected in this study comes from two mobile monitoring deployments – one in Denver, Colorado, USA in 2014 and one in the San Francisco Bay Area, California, USA in 2019 – 2020. The first study included both parked and mobile collocations and allows a direct comparison of the two techniques, whereas the second study
- 125 contains a larger dataset that allows for a deeper exploration of the relationship between distance and temporal aggregation scales for optimizing the comparisons. We also demonstrate a real world application of the method to detect drift in a NO₂ sensor deployed as part of Aclima's collection fleet. Finally, we discuss the results within the context of spatial heterogeneity of the observed pollutants and the implications for extending the approach to additional pollutants not included in this study.

2 Instrumentation used in this study

- We analyze measurements from several different vehicles equipped with the Aclima, Inc. mobile laboratory measurement and acquisition platform (Aclima Inc., San Francisco, CACalifornia, USA). 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) were outfitted with the Aclima platform in Denver, CO in partnership with the Google Earth teamColorado, USA during the summer of 2014 (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- of California in 2019 2020. Aclima designed the AMCL to support the field calibration of Aclima's sensor-based
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Mobile Node devices (AMN) for deployment in the Aclima mobile monitoring fleet. In both studies, <u>eachthe</u> mobile <u>platform</u> <u>wasplatforms were</u> equipped with high resolution (0.5 hz or 1 hz data reporting rate) reference-grade air pollution <u>monitorsinstrumentation</u> to measure <u>ozone (O₃), nitrogen dioxide (, NO₂), and nitric oxide (NO), among. Additional</u>

- 140 measurements, including black carbon (BC), size-fractionated particle number counts (PN), and other species were also measured during these campaigns but are not discussed in this manuscript. For this manuscript, we focus on measurements that had equivalent mobile and fixed reference site measurements for both studies. A critical part of this work is our comparison of parked and mobile collocations during the 2014 Denver study, for which we had one-minute averaged reference site data for O₃, NO₂, and NO but not the other measured species.
- O₃ was measured using ultraviolet (UV) absorption with a gas phase (nitric oxide) O₃O₃ 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), (Long et al., 2021), and has been designated <u>as</u> a Federal Equivalent Method (FEM) by the USEPA (Designation -EQOA-_0514-_215, 40 FR 79, June 18, 2014, p. 34734 -_ 34735). The 2B Model 211 only reports ozone at 2-second intervals, so ozone was measured and reported as 2-second averages for the
- 150 Denver study. For the California study, all ozone data was averaged up to 10-second averages during initial data collection, so 10-second averaged data is used for the analysis. NO₂ was measured using cavity attenuated phase shift (Teledyne <u>API</u> Model T500U) and represents a "true NO₂" measurement (Kebabian et al., 2005) and (Kebabian et al., 2005). The Teledyne <u>API</u> Model T500U 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 iswas measured using O₃ chemiluminescence (Ecophysics CLD64), which
- 155 is a recognized international standard reference method for measuring NO (e.g., EN 14211:2012). <u>Although theseNO and NO₂ are both measured as 1-second averages in both studies. These</u> instruments have all been evaluated by strict test criteria and are recognized as reference methods by various regulatory agencies, the <u>showever</u>, the reference method designations do not apply to the applications (mobile monitoring) and timescales (1 second to 1 hour) assessed here. These instruments were selected chosen for thetheir demonstrated excellent data quality and performance and to serve as a mobile reference for
- 160 calibration purposes recognizing. It is important 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 environmentwe assess these methods using reference-grade equipment so we can be used for assessing existing mobile monitoring data. We are also excited about developing techniquesfocus on variability due to spatial heterogeneity instead of instrument issues. If the methods we develop here are successful with reference-grade equipment, they can be used to evaluate
- 165 the performance of next-generation air quality instruments, which might not meet the same strict regulatory standards at present but <u>can</u> 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
- 170 terms of precision, accuracy, and stability.(Castell et al., 2017; Clements et al., 2017; Wang et al., 2021).
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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)

175 3.1 Methods

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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, NO₂, O₃, and O₃+NO₂ (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 O₃ and NO₂. In addition, there are emerging technologies (such as some types of electrochemical sensors) that measure O_x directly rather than O₃ and NO₂ separately.

We use data from a mobile measurement campaign that occurred in the Denver, Colorado, USA region during the summer of 2014.nearby fixed reference site. For this analysis, we use data from a mobile measurement campaign that occurred in Denver, Colorado, USA 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, wethe drivers were 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 These parked collocations were integrated into the experimental plan to assessing data quality of the mobile platform measurements by comparison to the fixed reference site measurements. The parked collocations lasted about 20 minutes each 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°

- 195 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.lare.nasa.gov/missions/discoveraq.html). °N, 104.987625 °W) and La Casa (39.779460 °N, 105.005124 °W) sites. The drivers were instructed to park as close
- 200 as possible to the site but were required to park in an available public parking space on a public street. These restrictions limited how close each car could get to the reference site for each collocation and resulted in individual parked collocation locations ranging in distance from the regulatory stations, as shown in Figure 1. In addition, the drivers were instructed to park facing
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into the wind when possible to minimize the influence of self-sampling, which posed additional restrictions Additional details about the monitoring campaign can be found in the Supplemental Information and in Whitehill et al. (2020). The CDPHE reported hourly Federal Reference Method (FRM) and FEM measurements of O₃, NO₂, and NO, and also reported one-minute time resolution data for our study period as part of the 2014 DISCOVER-AQ field campaign (https://wwwair.larc.nasa.gov/missions/discover-aq/discover-aq.html).

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- 210 Figure 1: 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 and the red shaded areas indicate the range of areas where the vehicles parked during the parked collocation periods. Car-to-site distances varied from 80 to 145 m for the La Casa site and from 10 to 85 m for the CAMP site.
- 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 O₃ (80 ppbv) did not drift beyond the instruments' specifications during the study, so we did not perform any adjustmentno adjustments were made 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 within a vented tee configuration. A certified NO gas cylinder was diluted by zero air to provide a 360 ppbv NO span gas for the NO instrument, and a certified O₃ generator producedwas used to produce 80 ppbv levels of O₃ for the O₃ span checks. We calibrated the NO₂ instrumentsinstrument before and after the study in a laboratory but only performed zero checks on the NO₂ monitorsinstruments in the field.

_____The bias of the $\Theta_3 O_3$ instruments varied between 3% and 6% with a standard deviation of 5%. The bias of the NO instrumentinstruments varied between 3% and 8% with a standard deviation of 6%. The NO gas_standard gas_had a

concentration uncertainty of $\pm 2\%$ (EPA certified gradeCertified Grade). Mass flow controllers in the dilution calibrator were within their certification period and had a specified accuracy of $\pm 3.6\%$ at the conditions we used to generate the span gases. The accuracy of the O₃ generator was $-of \pm 1\%$ and was certified less than three months before the study. We attempted to perform gas-phase titration to produce NO₂ span gases in the field, but technical issues prevented us from performing accurate

230 daily span checks on NO₂. We did not observe any drift in the NO₂ instruments between the pre-studycampaign calibration and the post-studycampaign calibration of the NO₂ instruments, so we used those calibrations for the entireassumed the calibration of the NO₂ instrumentation was constant throughout the campaign period.

At least one of the cars was parked at the CAMP site (within 85 metersm) for 15 time periods during the study (Table S1) and at the La Casa site (within 150 metersm) for 16 time periods (Table S2). We previously compared measurements among the three equivalently equipped cars (Whitehill et al., 2020)(Whitehill et al., 2020) and determined that one-second 235 NO₂ and O₃ measurements for the three platforms agreed to within 20% for 73% (NO₂) and 61% (O₃) of 1 second measurements taken whenduring a day of collocated driving together. NO showed higher variability, likely reflecting hyperlocal differences in NO concentrations from exhaust plumes, but were still agreed within 20% about 34% one third of the time. A similar comparison of two similarly Aclima-equipped Google Street View cars in San Francisco and Los Angeles also showed excellent car versus car comparability (Solomon et al., 2020). For the purposes of the present analysis, we assume the data from the three cars to beis equivalent and interchangeable.

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We aggregated all the 1-hz mobile measurements data up to 1--minute averages (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**discussion 245

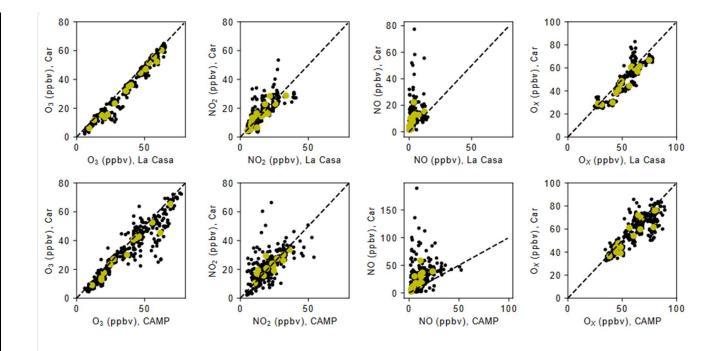


Figure 42: Scatterplots of 1-minute (black dots) and period mean (vellow dots) car measurements (O₃, NO₂, NO, and O_X) versus fixed reference site (La Casa or CAMP) measurements during the stationary collocation periods. A black dashed 1:1 line is provided for reference.

Figure 2 shows scatterplots comparing 1-minute O₃, NO₂, NO, and O_X (O₃ + NO₂) 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). O_X was included in the analysis because it is more likely to be conserved in fresh NO_X emission plumes
 (assuming most of the NO_X is emitted as NO) than O₃ or NO₂ separately. We calculated the period-specific means by averaging the discrete 1-minute measurements over the continuous measurement periods that the car wascars were 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)in a similar way and computed ordinary least squares (OLS) regression statistics for the 1-minute data, the period-specific means, and the period-specific medians (Table 4<u>S3</u>).

Figure 2 shows relatively good agreement in the 1-minute observations between parked mobile and stationary reference measurements of O₃ and NO₂ (and O_x), despite some scatter in the relationship. The agreement for NO is poor relative to that for the other pollutants. The coefficients of determination (r²) are highest for O₃, then NO₂, followed by NO (see Table S3). With the exception of O₃, the linear regression statistics such as slope and intercept did not provide an accurate assessment of bias due to the influence of outlier points on the OLS regression statistics, as evidenced by the relatively low r²
 (for NO₂ and NO in particular). This relationship is consistent with the expected trends in spatial heterogeneity between the 3

pollutants (Section 7) and illustrates how there can be significant variability in the 1-minute differences between parked mobile

and stationary measurements. The r^2 does improve somewhat when using the period specific aggregates (mean and median, Table S3), suggesting that temporal aggregation can reduce some of the variability in the difference and improve the comparisons.

- 270 In order to minimize the impact of scatter in the measurements further, we also looked at the statistics of 1-minute mobile versus fixed site differences, here denoted as $\Delta X = X_{mobile platform} - X_{reference site}$. We looked at the mean (i.e., mean bias error), median, standard deviation, and 25th and 75th percentiles of the ΔX distributions (Table 1). The mean and median of the ΔX values effectively aggregates the observations across all of the parked collocation periods and is a more direct assessment of systematic bias than the slope and intercept of the OLS regressions (especially for NO₂ and NO). This is particularly true
- 275 given that the measurements are made under non-ideal conditions, such as these on-the-road, parked (nearby but not spatially coincident) collocations where spatial variability in pollutant concentrations at fine spatial scales appears to be significant. For the ease of discussion, in this and subsequent sections we refer to these ΔX values generally as bias, which includes both spatial and measurement bias.

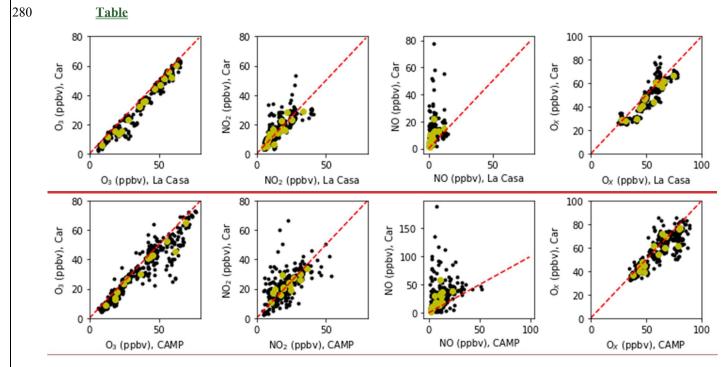


Figure 1: Seatterplots of 1-minute (black dots) and period mean (yellow dots) car measurements (O₃, NO₂, NO, and O_x) versus fixed reference site (La Casa or CAMP) measurements during stationary collocation periods. A red dashed 1:1 line is provided for reference.

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Table 1: Regression statistics for O_3 (ppbv), NO_2 (ppbv), and O_X (ppbv) for the parked mobile platform at a fixed reference site (either La Casa or CAMP) during the 2014 Denver study.

290 <u>1: Statistics of 1-minute ΔX comparisons for the stationary collocated periods. Units are ppbv. sd is standard deviation, P₂₅ is the 25th percentile, and P₇₅ is the 75th percentile.</u>

			La Casa							САМР				
			meanSlop medianInt			$r^2 \underline{sd}$ Slope <u>P</u>		Interco	ept	<u>r²me</u>	medi	<u>sd</u>	<u>P₂₅</u>	<u>P₇₅</u>
			e	rcept			<u>25</u>	<u>P₇₅</u>		<u>an</u>	<u>an</u>			
Q 3			I				1	1		1	1	I		
1 Minute	<u>40</u> ₃		<u>-3.6</u>	<u>-2.8</u>		<u>3.3</u>	<u>-5.3</u>	<u>-</u> 1. 013	<u>2</u>	-	0.96	0.873	0.5	=
										4. 12	7 <u>-3.4</u>	<u>6.9</u>	4_	0.858
										<u>3</u>			<u>6.5</u>	<u>5</u>
<u>Mean∆</u>	1.0	-	0. 988<u>0</u>	-0. 921 9		<u>5.5</u>	- <u>3.</u> 0 .91	2.6		<u>1.0</u>	0. <mark>95</mark>	7.6	-2.8	<u>3.8</u>
<u>NO₂</u>	20	4.4 7									θ <u>5</u>			
Median		1		4.50	0.9	91 0.	960 -2	.29	0.9	960	1	1	1	1
NO ₂			3											
<u>∆NO</u>			<u>5.</u> 1	0.8381.9		<u>2.498.</u>	0.5969	0.652	5.9	<u>14.2</u>	<u>6.</u> 8.4	<u>22.</u> 0 .3	<u>1.6</u>	<u>19.7</u>
						<u>8</u>					2	99		
Mean				2.12	0.7	75 0 .	672 7.	78	0.6	574				
Median			7 0.81	2.78	0.7	79 0.	749 5.	<u>82</u>	0.7	767				
Wieuldiff			4	2.10	0.7		, 17 .	02	0.					
NO														
1 Minute			1.16 4	4. 61	0.1	49 1.	099 13	<u>3.29</u>	0.	149				
<u>Mean∆O</u> >	K		1.252 -3.6	4.46-3.8		0.442 5	<u>-6.8</u>	-1.746	51	-3.2	-2.7	8.2	=	0.44
	_					<u>.6</u>							6.7	1.4
						<u></u>							27	
M 1			0.00	2.50	0.1	105 1	192 7	1.5	0.2	2.4.1			<u> </u>	
Median		0.99 3	<u>3.59</u>	0.4	1 05 1.	4 82 7.	15	0	341					
Ox														
1 Minute			-	-0.03	0.8	3 <u>28</u> 0.	816 7 .	81	0.	7 08				
Mean			2 0.95	1.39	0.8	3 85 0.	893 3.	26	0.9	324				
			8											
Median			0.93 2	0.82	0.9	018 0.	872 4.	25	0.8	839				

Of the four pollutants analyzed, O₃-produced the highest coefficients of determination (r^2), with 1-minute mean r^2 =

295 0.97 for theLa Casa site and $r^2 = 0.86$ for the CAMP site. The coefficients of determination for 1-minute mean NO₂ and NO were significantly lower, with $r^2 = 0.60$ for NO₂ and $r^2 = 0.15$ for NO at the La Casa site. The CAMP site had a lower $r^2 = 0.40$ for NO₂ and similar values for NO. The r^2 values for O_X regressions fall between those for O₃ and NO₂ 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 NO₂ and NO showing significantly greater values of r^2 for the 300 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).

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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 O₃, NO, and NO₂ (due to rapid O₃ - NO_X titration), but O_X remains constant in the absence of photochemical O₃ production or direct emissions of NO₂ or O₃. Photochemical O₃ 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 O3 emissions in an on-road environment. Although direct NO2 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).

- From Table 1, NO₂ shows excellent agreement between the mobile platforms and the fixed reference sites. Mean and 310 median bias values for NO₂ were within 1.0 ppbv of 0 for both the CAMP and La Casa sites. O₃ shows a minor (but persistent) offset of around 3 ppbv for both sites, which is also apparent from the scatterplots (Figure 2). Both the 25th and 75th percentiles for ΔO_3 were negative as well, suggesting a real differences in the ozone measurements between the mobile platform and the fixed reference site. The O_X biases were similar to the sum of those for O_3 and NO_2 , as anticipated from our definition of O_X . Although the median NO bias for the La Casa site was small (1.9 ppbv), the mean bias for the La Casa site and the mean and
- 315 median biases from the CAMP site were significantly larger, with values between 5.1 ppbv (for the La Casa mean) and 14.2 ppbv (for the CAMP mean).

The La Casa site is located over 80 m from the nearest street in a predominantly residential neighborhood (Figure 21). 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 . The influence of concentrated direct emission plumes 320 at the mobile platform are 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 primary source of discrepancies between the mobile platform and the fixed reference site are the. The relative biases in instrument calibration, relative biases and differences in the concentrations due to the distance between 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 325 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), may also contribute to the discrepancy. From the

combined timeseries of all collocations (Figures S4 and S5), short-term peaks in NO and NO_2 are present in the mobile platform measurements but not the fixed reference site measurements. This reflects the impact of emission plumes from local traffic.

330 The traffic influences are particularly noticeable at the CAMP site, reflecting its location at a major intersection. While we cannot rule out self sampling of exhaust from the mobile platform, the higher frequency of plume events at the CAMP site versus the La Casa site suggests that local traffic emissions are the primary source of the observed pollution plumes.



- 335 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 ΔO₃, ΔNO₂, 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—______The parked collocation results support the assessment of instrument bias; however, the influence of local traffic emissions on the collocation does result in non-optimal conditions. This can be alleviated somewhat through temporal aggregation of the differences, but still reduces the applicability of a linear regression approach to determining slopes and intercepts for comparisons. Although it is possible to impose strict collocation criteria for parked collocations that would limit the influence of local emissions, the operational constraints during large-scale mobile monitoring campaigns often necessitate the use of publicly-accessible sites for frequent collocations. Since most scalable parked collocation solutions are likely to be affected by traffic emissions, expanding to allow the use of additional data while added advantages that spatial biases are averaged out by the motion of the mobile platform through space, effectively allowing

each mobile datapoint to sample a larger (and, by extension, more representative) amount of air in the same sampling duration (e.g., Whitehill et al., 2020)

median difference of about ΔO₃ ≈ -3 ppbv and ΔNO₂ ≈ 1 ppbv, but also variability between different periods. The period 355 median ΔO₃, ΔNO₂, 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 sitessite in Denver (2014)

4.1 Methods

360 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

- 370 were associated___To assess the performance of a mobile collocation approach, we used all the measurements (stationary and moving) collected during the 2014 Denver study, using the parked collocation results as a point of reference. We associated the raw 1-Hz car data with the nearest road using a modified "snapping" procedure (Apte et al., 2017), (Apte et al., 2017), where each mobile datapoint was we associated each mobile datapoint with the nearest 1-meter road segment whose direction was approximately parallel (i.e., within 45°) to° of the car's heading.
- 375 We assigned each 1-hz datapoint to 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. (Table S4). We also created an aggregate "Non-Highway" road class, "Non-Highway", which consisted of the union ofroads in the Residential, Major, and Other road classes (i.e.g., everything that was not in the "classified as a Highway" road class). Based on our results from Section 3, we predicted believe that the measurements made on lower traffic roads (such
- 380 as-Residential roads) would will generally have lower traffic, and thus stronger correlations to agreement with measurements at most fixed reference sites, especially those located away from major roads or intersections, than higher. While travelling on high traffic roads (such as Highway roads). We also anticipated highways), the cars are more likely to be impacted by direct emission plumes. In effect, we are assuming that the OSM road classifications is a general proxy for on-road traffic volume.

In addition to road type, we anticipate that the distance between the fixed reference site and the mobile platform wouldwill

- 385 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 analysisthe comparisons, with the closest agreement when the distances are small.
- 390 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).

We looked at the mean and median biases and coefficients of determination for measurements made by the cars versus those at the La Casa site. We broke down the analysis by road class and distance buffer, starting at a distance buffer of 500 m and expanding out in 250 m increments. Our goal was to explore how the central tendency of the bias distribution was affected by the cars' distance from the La Casa site, as well as the impact of different road types. We also present (in the Supplemental Information) scatterplots and linear regression statistics for five discrete buffer distances (100 m, 300 m, 1000 m, 3000 m, and

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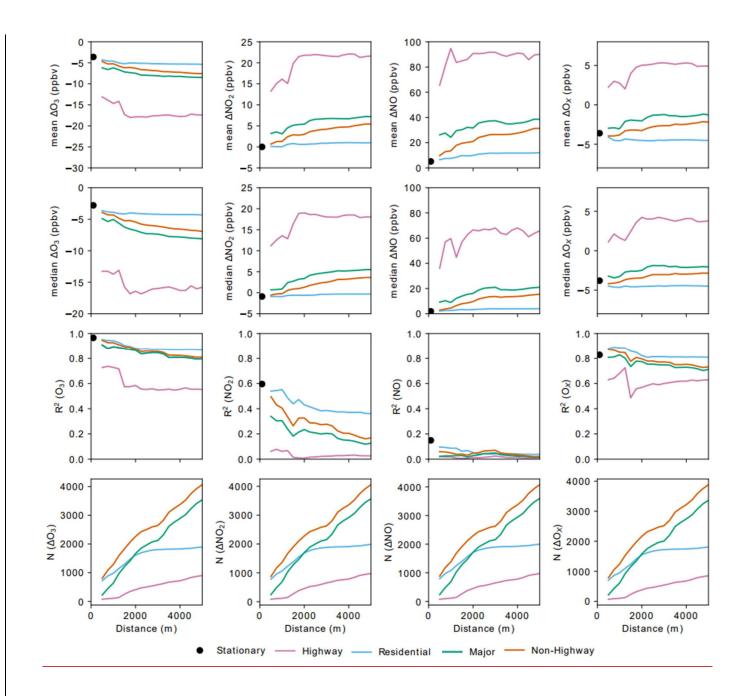
10000 m) and the five different road classes.

4.2 Results and Discussiondiscussion

Regression results for different road type and distance class combinations are shown in Figure 3 for O₃ and Figure 4 for NO₂. 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 405 the 5 km radius around reference sites resulted in most of the mapped roads designated as Residential._____ There_____ fundamental differences between the in-motion collocations discussed in this section and the parked collocations discussed in Section 3. These differences have implications for the interpretations of central tendency (mean or median) bias values and r^2 values. In general, the mean and median bias values can reflect both measurement bias as well as persistent spatial differences. The r^2 values generally reflect the random variability between the mobile and stationary measurements that results from a 410 combination of measurement precision as well as true spatial variability. Both the bias values and the r^2 values for the inmotion observations reflect additional sources of spatial variability that need to be considered as compared to the parked collocations. This is due to the wider range of distances (up to 5 km), varying road types and associated traffic patterns, and potentially different spatial distributions of non-mobile sources in the wider areas covered. As discussed in Section 3, r^2 values 415 vary depending on the pollutant measured, and can be quite low even for parked collocations. As a result, we conclude that r^2 would not be a good indicator of instrument performance (i.e., precision error) and that using a parametric linear model to

attribute gain and offset instrument biases separately is not possible, particularly for NO and NO₂. Therefore, the inclusion of r^2 in this section is primarily as context for understanding random spatial variability in the comparisons. The focus of this section is to characterize the dimensions over which these spatial differences, both random and systematic, manifest in the

- 420 data. By doing so, we hypothesize that we will be able to isolate the conditions under which the variability in bias values can be expected to reflect variability in measurement bias between the mobile and stationary monitors. In particular, we are looking for an optimal operational method for mobile collocation that provides comparable results to that determined from the parked collocations.
- The mean and median bias values and r^2 values for the car versus the La Casa comparisons are shown as a function of buffer distance and road type in Figure 3. We also display the results from the stationary collocation analysis (Section 3) to demonstrate the relationship between the mean and median biases and r^2 values from the stationary collocations alongside those from our expanded analysis in this section. Because measurement bias is not expected to correlate with the spatial dimensions featured in these analyses, the difference between road type and with varying distances from the site can be interpreted purely as persistent spatial differences. As shown in Figure 3, measurements from the Highway road class resulted
- 430 in a significantly higher magnitude of bias compared to other road types, indicating larger spatial variability in concentrations measured on Highways (with respect to the stationary site measurements) than for other road types. The r^2 values on Highways are generally lower than on other road types, indicating that there is higher random variability on Highways compared to the stationary site measurements. Differences in bias and r^2 between Major and Residential road types are significant in some cases and less so in others, depending upon the pollutant and the distance. However, even in cases where the differences are
- 435 significant (e.g., NO_2 at distances greater than 1000 m), the magnitude of the bias for Major roads is only a fraction of that from Highways. As discussed above, the degree of bias and random spatial variability between the mobile measurements and the fixed reference sites are primarily impacted by the direct emission plumes on the roadway. We anticipate that Highways have higher traffic and a higher fraction of more heavily polluting vehicles (e.g., heavy duty trucks), which explains the larger magnitude of bias and lower r^2 values. Roads with lower expected traffic volumes and less heavy duty vehicles, such as
- 440 <u>Residential roads, have a lower magnitude of bias.</u>



445 Figure 3: Mean and median ΔX, coefficient of determination (R²), and number of datapoints (N) for 1-minute car versus
 445 La Casa comparisons as a function of the maximum distance between the car and the La Casa site. Results from stationary collocations (Section 3) are shown as black dots, whereas different road classes are shown in different colors.

For all road types, the bias between the mobile measurements and the reference site is lowest (and r^2 is highest) for distance buffers closest to the site. For the Residential road class, the bias between mobile collocation and parked collocation changes very little as the distance buffer increases for all species. The bias for observations collected on Major and Highway road types generally increases as additional samples are included at greater distances from the site.

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When considering the results of bias by road type and distance class, it is important to note that the distribution of road type varies by distance. This is indicated by the number of data points (N) by road type as a function of distance in Figure 3. The La Casa site is in a residential area, so most of the roads within 500 m of the site are Residential. The designated drive

455 patterns near the La Casa site resulted in most of the mapped roads within 2 km of the site being a combination of Residential and Major road types. Further away from the La Casa site, the roads consisted of a larger fraction of majorMajor roads and highwaysHighways that were used to commute tobetween the different areas where the denser mapping occurred.

For both Major and Residential road types, the bias and r² values tend towards the values from the parked collocations as the buffer distance decreases in most cases. Note that most of the mobile-to-stationary data within 1 km of the La Casa site
 was measured on the same days and generally within a 2 hour window of the stationary collocation data. For NO and NO₂, there are some slight differences between the parked collocation results and the 500 m buffer distance, with slightly higher bias on Major roads compared to the parked collocations. For NO and NO₂, this indicates an increase in the systematic spatial bias when in motion on nearby Major roads compared to when parked along with minimal differences in random variability. These results highlight the potential of in-motion mobile collocation compared to parked collocations in reducing the random

- 465 variability, especially on Residential roads in the immediate vicinity of the stationary site. In general, however, there is remarkable consistency between the in-motion and parked collocation results for all pollutants when Highways are removed from the data set. Depending upon the target quality assurance guidelines of the study, there might be significant advantages to using in-motion mobile collocations instead of parked collocations to determine changes in mobile versus stationary measurement biases.
- 470 Scatterplots of the one minute mobile platform measurements versus the one minute La Casa measurements are shown in the Supplemental Information for O_3 (Figure S6), NO_2 (Figure S7), NO (Figure S8), and O_X (Figure S9). These are shown, along with ordinary least squares linear regression statistics, for each road class individual and for distance buffers of 100 m, 300 m, 1000 m, 3000 m, and 10000 m. As with the stationary collocation scatterplots (Figure 2), these show the best agreement (i.e., closest to the 1:1 line) for O_3 and O_X , with moderate agreement for NO_2 and the worst agreement for NO. As suggested
- 475 <u>above, the regression statistics (slope and intercept) appear to be a poor indicator of agreement, especially for directly emitted</u> species like NO (and, to a lesser degree, NO₂). Therefore, we focus our analysis on the central tendency metrics of the bias and the r^2 values as shown in Figure 3.

One final consideration for interpreting the impact of spatial variability in the Denver dataset is the impact of temporal aggregation.

480 **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
other	

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

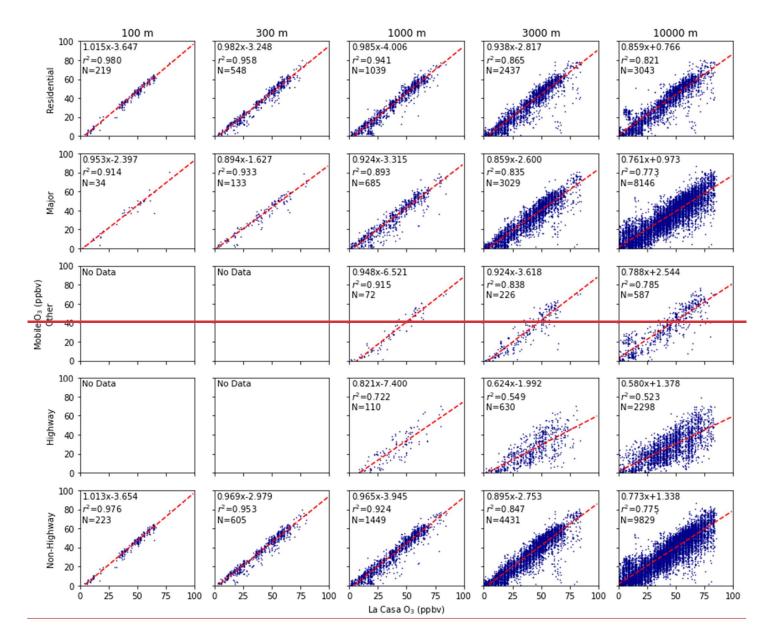


Figure 3: Scatterplots of 1-minute average mobile platform versus 1-minute fixed reference site O_3 for different road classes within five distance buffers of the fixed reference sites in the Denver study. The red dashed lines show the result of the ordinary least squares regression.

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The concentrations of NO₂ (Figure 4) and NO (Figure S2) measured by the mobile platform while driving on Highways were higher and O₃ 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.

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The results show good correlations for O₃ and O_x, modest to poor correlations for NO₂, 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.

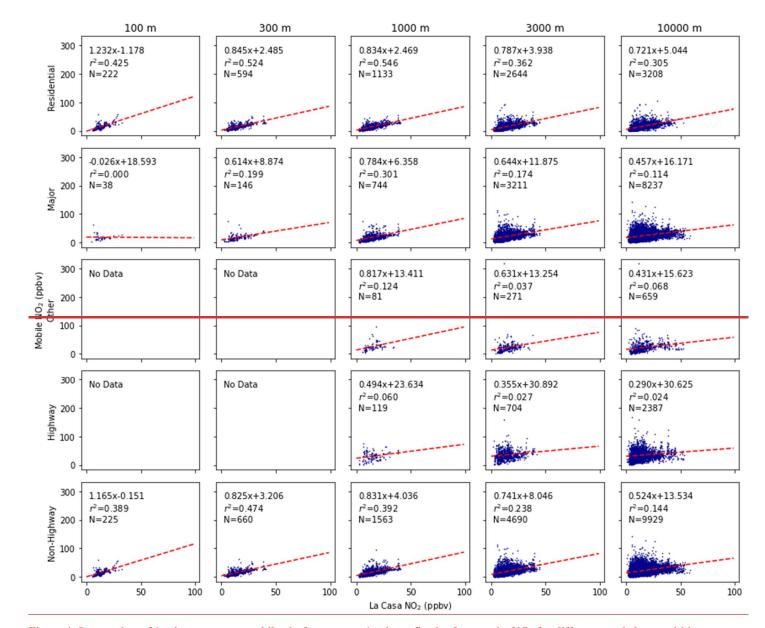


Figure 4: Scatterplots of 1-minute average mobile platform versus 1-minute fixed reference site NO₂ for different road classes within five distance buffers of the fixed reference sites in the Denver study. The red dashed lines show the result of the ordinary least 505 squares regression.

We found that Residential roads provide the best comparability with the La Casa site at smaller car versus site distance classes. For O₃ 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 NO₂ was modest, with r^2 values between 0.42 to 0.55 for radii of 1000 m or less, with slopes

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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 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 NO₂) in 525 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.

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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 intercept for NO₂ 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

545 event. Regressions statistics for the mobile stationary comparison are shown for mobile and stationary data averaged for 1minute and 1-hour.

	C	} ₃ (ppbv)	N	O₂ (ppbv)	Ņ	<mark>O (ppbv)</mark>	ł		O _X (ppbv)	•
	Slope I	ntercept r ²	Slope 1	Intercept	₽ ²	Slope	Intercept	₽ ²	Slope	Intercept	₽ 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	0.52 4	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	0.825	3.21	0.474	1.257	<u>8.799</u>	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	0.867	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	<u>-4.1 0.967</u>	0.902	2.05	0.609	1.287	7.908	0.223	0.978	-3.133	0.93 4
1 Hour Non-Hwy 300m	0.945	-2.96	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	<u>-4.152</u> 0.956	0.972	3.134	0.559	1.756	15.5	0.13	0.943	-0.018	0.918

Figure 3 indicates that O₃ tends to be lower at the mobile monitoring platform than the fixed reference site (for La
Casa), whereas NO and NO₂ 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 NO₂ on a local scale near the mobile platforms, while titrating O₃. 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 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 O₃ is between -3 and -4 ppbv, similar to the intercept of -4.1 ppbv from stationary collocation (Table 1). The results for NO₂ 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, 560 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.lare.nasa.gov/missions/discover-aq/discover-aq.html). ____ Data from regulatory monitoring stations in the USUnited States will typically only be available at 1-hour time resolutions. For mobile-to-fixed "collocation", and this is the

- 565 <u>data we had available for the California data set in Section 5. For mobile collocations</u> to be broadly applicable to large-scale mobile monitoring <u>campaignsapplications</u>, it is important to assess how the <u>correlation</u> 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 <u>criteria (e.g., road type, and buffer</u> distance, <u>etc.)</u>. The results are compared to the <u>correlations from the period mean of 1 minute stationary collocation data (Table 1)</u>. These period means are averages of
- 570 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 criteria. Figure 4 compared the results of the 1-hour aggregated comparisons to the 1-minute comparisons as a function of buffer distance elasses 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. Thefor the Non-Highway road class. Generally, the results for O₃ are more-or-less identical. The correlation for NO₂ improved with time averaging, with the 1-minute and 1-hour aggregation are similar in terms of both bias and r². For O₃ and O_x, the differences are minor. For NO₂ and NO there is a slight increase in the median bias as well as an increase in r² for the 1-hour r²-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 elasses. 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 versus the 1-minute 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 585 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. The r^2 at 1 hour for NO₂ and NO is variable as a function of distance buffer, likely due to the limited number of days, and thus will size of the data set and the variable distribution of road type with distance, so this may not reflect the influence 590 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 2020be an accurate assessment. The increase in r^2 for NO₂ and NO suggest that there is a modest reduction in the impact of random spatial variability at hourly aggregations; however, the increase in bias suggests that there is additional apparent bias as a result of aggregating. This could be due to the incomplete hourly aggregates from the mobile platform being compared to 595 the full hourly observations from the stationary site. However, the Denver dataset is too limited to explore this hypothesis adequately. The number of data points per hourly mobile collocation will be explored more thoroughly in Section 6.

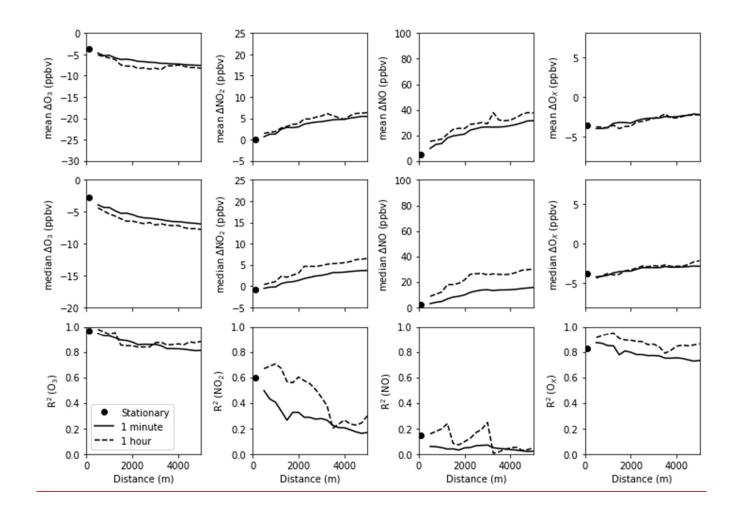


Figure 4: Mean and median ΔX and coefficient of determination (R^2) for 1-minute and 1-hour car versus La Casa comparisons as a function of the maximum distance between the car and the La Casa site. Results from stationary collocations (Section 3) are shown as black dots. All Non-Highway roads are included in this analysis.

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

5.1 Methods

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Aclima-operated fleet vehicles are equipped with a mobile sensing device, the Aclima Mobile Node₅ (AMN), which measures CO, CO₂carbon monoxide, carbon dioxide, O₃, NO, NO₂, PM_{2.5}, and TVOC. As part of the AMN calibration procedure, up to 16-total VOC. The AMNs at a time are deployed incalibrated using the Aclima Mobile Calibration Laboratory (AMCL), a gasoline-powered Ford Transit van equipped with laboratory-grade air pollution measurement instrumentation. The AMCL iswas driven around the San Francisco Bay Area of California to calibrate the sensors within the AMNs through

comparison of the AMN sensor response measurements with the laboratory-grade equipment collocated in the same van. The

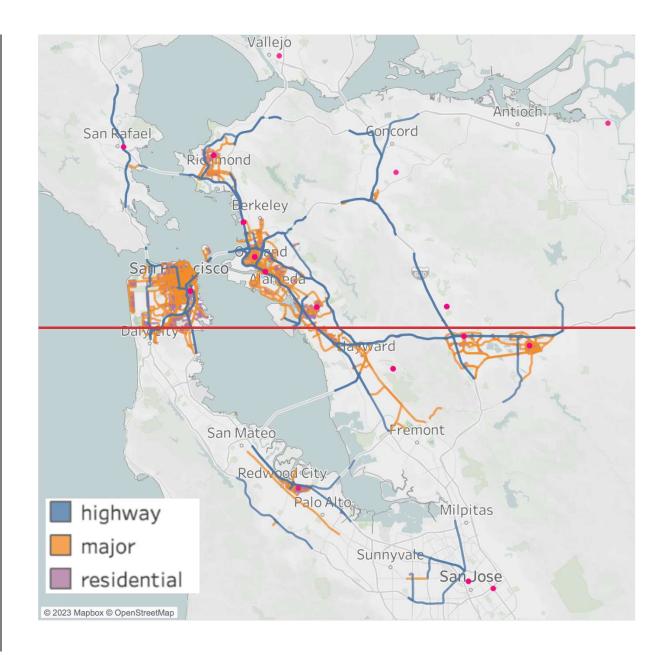
610 <u>laboratory-grade instrumentation in the AMCL are calibrated regularly using reference gases to maintain bias and precision</u> objectives. The calibration procedures have been described in Solomon et al. (2020). Bias and precision results across approximately 25 zero and span checks are shown in Table 2.

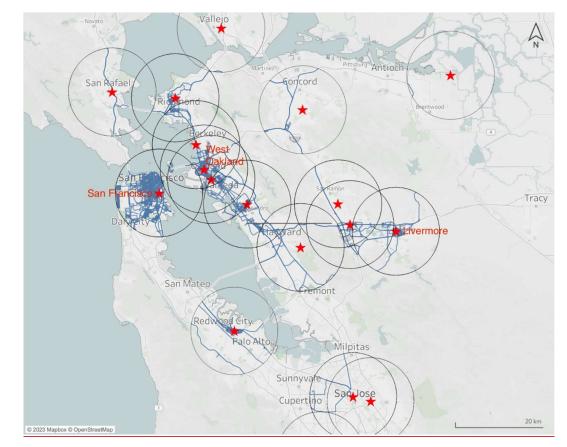
As part of the validation process, the AMCL regularly drives around several Bay Area Air Quality Management District (BAAQMD) regulatory monitoring sites (Figure 5). These BAAQMD sites are equipped with EPA-approved

615 reference FRM and FRM measurements of NO, NO₂, and O₃ (or a subset of these species), along within addition to other regulatory measurements.pollutants. We present an analysis comparing the AMCL reference measurements with nearby BAAQMD reference measurements between November 2019 and October 2020. All measurements within 10 km of the regulatory sites were included in the data set used for analysis, as displayed in Figure 5.

Calibrations using reference gases were performed throughout the collection period to assess bias and precision.

620 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.





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

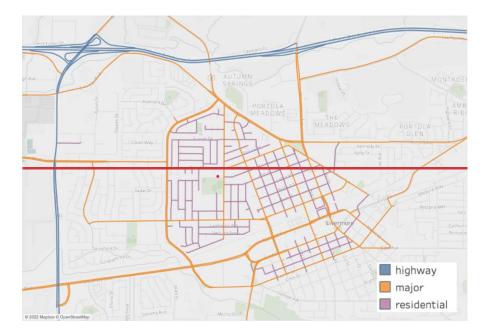


Figure 6: Zoomed in view of the street-by-street Aclima Mobile Calibration Laboratory driving pattern<u>The circles</u> 630 <u>delineate a 10 km radius</u> around the Livermore site in the San Francisco Bay Areaeach regulatory station. The roads within each circle shown in blue are roads with measurements used in the analysis.

 Table 42: Precision and bias of measurements in the Aclima Mobile Calibration LaboratoryAMCL from approximately 25

 quality assurance (QA) checks during the San Francisco Bay Area study period. Each QA check consisted of a zero and span point for each pollutant.

Pollutant	Bi	as	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%		
O3O3 (ppbv)	0.5	2.5%	0.4	2.0%		

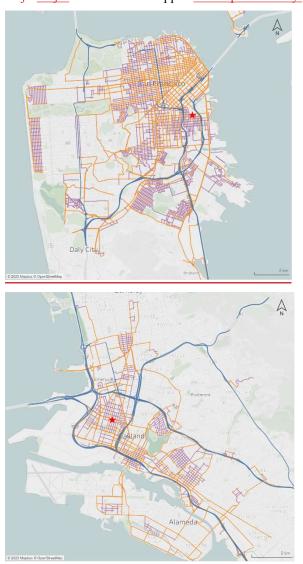
_____The dataset included comparisons to 19 different BAAQMD sites that represent a number of differentseveral spatial representativeness scales (40 CFR 58 Appendix D), but most). Most of the data was collected near the Livermore, San Francisco, and West Oakland sites (Figure 5)... These three regulatory stations wereare specifically labelled in Figure 5, and details of the road locations and road types mapped around each site are shown in Figure 6. Aclima selected these three stations to represent different climatological and land use regimes in the San Francisco Bay Area—. The San Francisco (station is in a warm summer Mediterranean climate with marine influence, cool winds and fog in summer, little_overall little_seasonal temperature variation, and mixed residential with light industrial), land use. The Oakland (station has a warm summer

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Mediterranean climate with marine influence, overnight fog in the summer, and mixed industrial and residential), and land

645 use. The Livermore (station has hot summer Mediterranean climate, inland with some marine influence, and predominantly residential, land use while being upwind of a large fraction of the urban Bay Area emissions). In contrast to the mapping performed in Denver, measurements near these three monitoring sites generally involved mapping essentially all publicly available streets within a few kilometersa significant fraction of the roads near the site (Figure 6). Measurements near other regulatory sites are also included and were generally chance encounters due to the AMCL driving past these sites on its way to its daily mapping assignments, as illustrated in Figure 5. As a result, this data tends to be from highwaysHighways or majorMajor roads and not mapped as comprehensively on a street-by-street basis.





655 Figure 6: Detail of the road locations and road types mapped by the Aclima Mobile Calibration Laboratory around the San Francisco (top), West Oakland (middle), and Livermore (bottom) regulatory sites (indicated by red stars) in the San Francisco Bay Area. Residential roads are shown in purple, major roads are yellow, and highways are blue

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 660 measurements in the US,USA 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.local traffic emissions. This is in contrast to Sections 3 and 4 where the mean was used to aggregate the one second data up to one minute or one hour. In general, using the median versus the mean produce similar results for O₃, NO₂, and O₃; 665 however, using the hourly medians versus means significantly reduces the impact of high NO outliers (peaks) on the NO aggregation. 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 <u>\$4\$10</u> and <u>\$5\$11</u>). 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 any individual hourly aggregate may include anywhere between

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a few seconds and a full hour of 1-Hz 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 Non675 Highway roads is shown in Figure 7 for O₃, NO₂, NO, and O_X, and ordinary least squares regression statistics (for both the Residential and Non-Highway subsets) are provided in Table 5. O₃ and O_X-have strong correlations (r² > 0.90) for both Residential and Non-Highway roads within all buffer distances up to 3 km, and the 10 km buffer distance has r² values that are only slightly lower (above 0.88, Table 5). For all buffer distances, O₃ and O_X-produced slopes within the range of (0.95, 1.05) and intercepts in the range of (-2.1, 1.6). NO₂ regressions produced lower r²-values than O₃ and O_X, ranging between 680 0.62 and 0.85. Ordinary least squares regressions from NO₂ 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.

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Table 5: Regression statistics, median ΔX values (m ΔX) 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.

0.20	6	9	0
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		Res	idential Roa		All No	n-Highway					
Species	Distance	Slope ¹	Intercept ¹	₽ ²	<u>*m∆X</u> ¹	N	Slope ¹	Intercept ¹	₽ ²	$*m\Delta X^1$	N
O 3	100 m	1.02	0.02	0.943	0.70	206	1.03	-0.23	0.963	0.80	4 69
O 3	300 m	1.04	-0.98	0.963	0.90	704	1.03	-0.71	0.953	0.57	786
Q 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	0.2 4	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	216 4
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
Θ_{X}	100 m	1.03	0.30	0.9 44	1.53	199	$\frac{1.00}{1.00}$	1.5 4	0.958	1.43	4 61
Θ_{X}	300 m	1.02	0.03	0.961	1.27	691	1.00	1.00	0.946	1.09	771
θx	1 km	1.03	-0.23	0.962	1.16	890	1.01	0.32	0.961	1.06	102 4
Θ_X	3 km	1.03	-0.23	0.9 44	1.11	1108	1.01	0.49	0.922	1.01	1623
Θ_X	10 km	1.03	-0.58	0.909	0.91	1503	1.01	-0.10	0.894	0.82	1888

* $m\Delta X$ is the median ΔX value.

⁴ units are in ppbv.

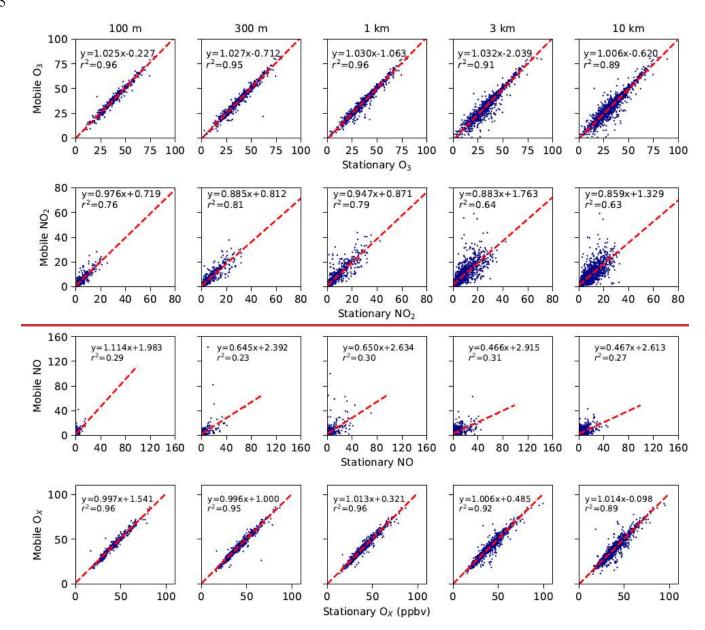


Figure 7: Scatterplots of 1-hour median mobile platform versus 1-hour fixed reference site O₃, NO₂, NO, and O_X for Non-Highway roads within five distance buffers of the fixed reference sites in the California study.

700 5.3 Using driving data for ongoing performance evaluation

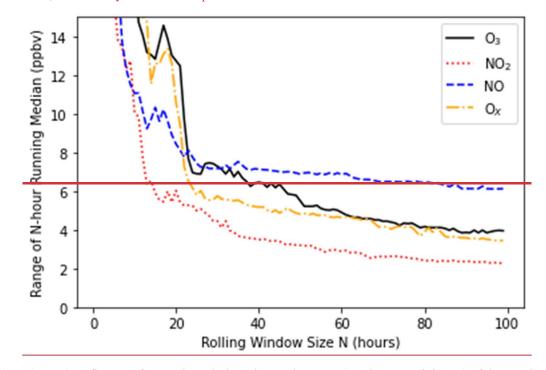
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We considered the possibility of using mobile versus fixed site measurements for calibration corrections, using metrics such as running slopes and intercepts of ordinary least squares linear regressions to provide a "correction" model to apply to the 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 ∆X 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 O₃₇ NO₂₇ and O_{X7} 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 715 of the hourly ΔX values (m ΔX 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 AX 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 720 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 AX 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 725 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} , ANO, and AO_x. Thus, we choose a 40-hour running median window as optimal for the following analysis, where the range of 730 AX values is around 3.5 ppbv for NO₂, 6.4 ppbv for O₃, 5.2 ppbv for O_x, and 7.1 ppbv for NO. Larger running window sizes (N) will decrease the range of AX 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 NO₂) 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 Using the approach described in Section 4.2, both the mean and median bias values and r² values are shown as a function of road type and distance buffer from the fixed regulatory sites in Figure 7. Similar to Section 4.2, we associated the central tendency of the bias to reflect both instrument biases as well as persistent spatial biases and the r² values to reflect random spatial biases. The California study is bolstered by a much larger data set (note that N in Figure 7 represents the number of hourly aggregates, whereas the N in Figure 3 represents the number of 1-minute aggregates). This dataset was also collected over a full year and includes comparisons with multiple stationary sites. Despite the different geographic locations and scale of data collection between the California and Denver studies, the general patterns observed in Figure 7 are highly consistent with the patterns observed in Figure 3. For example, higher magnitude of biases and lower r² at larger buffer distances and significantly worse agreement for Highways than other road classes. The California results do show somewhat higher r² and smaller magnitude
 of biases in general than the equivalent results for the Denver study (i.e., the hourly traces in Figure 4). The higher r², in
- particular for NO_2 , could be due to the larger dataset used, collected over a full year of atmospheric and climatological

conditions at multiple sites. This led to both a more representative dataset and a wider range of sampled concentrations than the one-month, single-site Denver analysis done in Section 4. The smaller bias, in particular for O₃ and O_X, could be ue to better inter-lab comparability in the California dataset, but aggregating data across multiple sites may also explain a reduction

755 in the systematic bias. For example, if one site has a slightly positive bias and another site has a slightly negative bias (due to monitor siting or random calibration variability), those biases will partially cancel each other out.

Figure 7 also shows close agreement between the results for the Major roads and the Non-Highway roads, reflecting the large number of Major roads (versus Residential roads) included in this study compared to in Denver. It is interesting to note that the number of datapoints (N) is almost identical for the Major and Non-Highway road classes. This is because most

760 hour periods that included driving on Residential roads also included driving on Major roads, so those hour periods were counted separately for the separate Residential and Major road classes but only one time for the Non-Highway road classes. Unlike the Denver dataset, this dataset shows remarkable consistency in the r^2 values between the Residential, Non-Highway, and Major road class subsets, suggesting that large scale application of these comparisons provide similar random spatial biases $(r^2 \text{ values})$ for Residential and Non-Highway roads. As with the Denver results, there is also minimal variability in these metrics with increasing distance buffers up through 3000 m (and higher).

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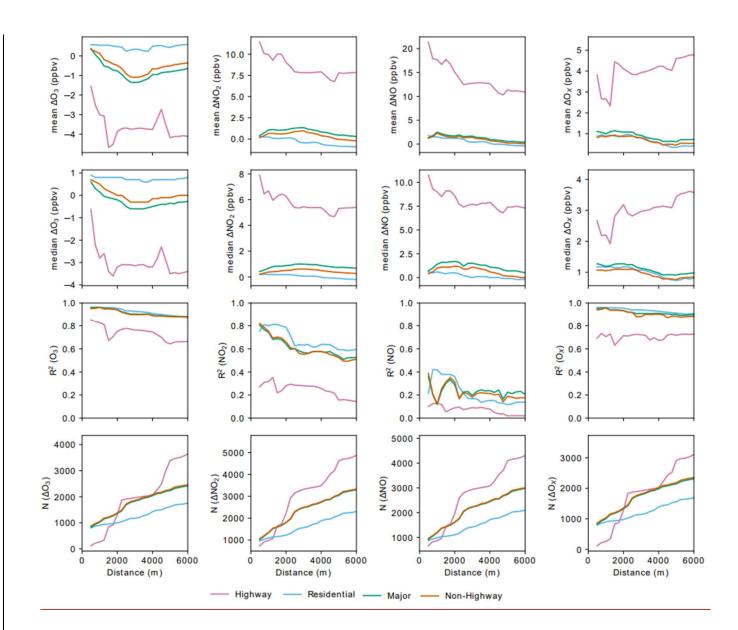


Figure 7: Mean and median ΔX , coefficient of determination (R^2), and number of datapoints (N) for 1-hour AMCL versus regulatory site comparisons as a function of the maximum distance between the AMCL and the regulatory site. Depending upon the buffer distance, the comparisons can include data for up to 19 BAAQMD regulatory sites. Different road classes are shown in different colors.

6 Operationalization of mobile-versus-fixed-site comparisons for ongoing instrument assessment

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These results in both Denver and California provide a blueprint for how operational decisions can be made for efficiently collecting collocation data to determine systematic measurement bias while accounting for the tradeoffs between

the rate of data collection and uncertainty tolerance for attributing changes in ΔX to measurement bias. The optimal approach

- 775 will balance these tradeoffs in a way that maximizes N, maximizes r^2 (i.e., minimizing random spatial variability), and minimizes the spatial bias component of ΔX . Based on our results in both Denver and California, it seems advantageous to remove Highway road segments from the data set. This would result in a decrease to both the random and the persistent spatial variability with minimal cost on the number of data points collected. The optimization of buffer distance and non-Highway roads to include is not as straightforward. As a thought exercise, consider two different examples of operationally feasible
- mobile collocations that are more scalable than parked collocations. In Scenario 1, we use only Residential roads and a distance buffer of 500 m. From the Denver results, this scenario would give results that are approximately equivalent to the parked collocation in terms of bias and random variability while simultaneously allowing the collection of hyperlocal air pollution data to meet monitoring objectives. It would also reduce the need to identify optimal parking locations near the stationary sites. In Scenario 2, we use all non-Highway roads and a distance buffer of 3000 m. Scenario 2 would provide increased bias and random variability over Scenario 1 and the parked collocations. However, it would increase the size of the dataset by a factor

of 2-3 and allow for simultaneously mapping a larger area to meet the monitoring objectives more efficiently.

The choice of which scenario to use requires an analysis along an additional dimension that has not yet been considered - the uncertainty with which ΔX can be determined. While there is likely a close relationship between the magnitude and the uncertainty of ΔX, the uncertainty in ΔX is more important than the absolute value of ΔX in the context of using mobile to stationary comparisons to attribute changes in measurement bias over time. Assuming that random spatial variability is the primary source of error in determining the true systematic spatial bias, the rate at which collocation data can be collected becomes a factor that may need to be weighted more heavily than the absolute magnitude of the spatial bias. This will be especially true for cases where r² is expected to be low (i.e., for NO₂ or NO), which indicates a higher degree of random spatial variability and, therefore, requires additional data collection to achieve an equivalent uncertainty reduction in ΔX. With this context, Scenario 2 appears to be a more attractive operational scenario despite the increased ΔX due to spatial bias and in the following section, we quantify the uncertainty in ΔX as a function of the amount of data collected and aggregated under Scenario 2.

6.1 Using running median bias values to identify instrument issues

800 Here we explore how the amount of data aggregated over a specified amount of collection time impacts the instrument bias calculated for the instruments that we know have a stable calibration over time. Our objective is to find the minimum temporal window required to determine reasonable uncertainty bounds for the determination of measurement bias. For the analysis, we use the California dataset of hourly ΔX values and calculate a running median of hourly median ΔX as a function of the number of hours of data in the running average, N. We use median ΔX as the results are less affected by outliers resulting
 805 from local emission plumes and thus produce estimates of the bias with smaller magnitude. For this analysis, we focus on a buffer distance of 3000 m and all Non-Highway roads (i.e., Scenario 2, as discussed earlier in Section 6). Figure 8 shows the

minimum and maximum bounds for observations of the running median ΔX for the entire dataset as a function of window size N. Note that each individual one-hour ΔX is the median of one-second differences during that one hour time window. Therefore, we are taking a running median (over individual hours) of hourly median ΔX values. The range between the upper

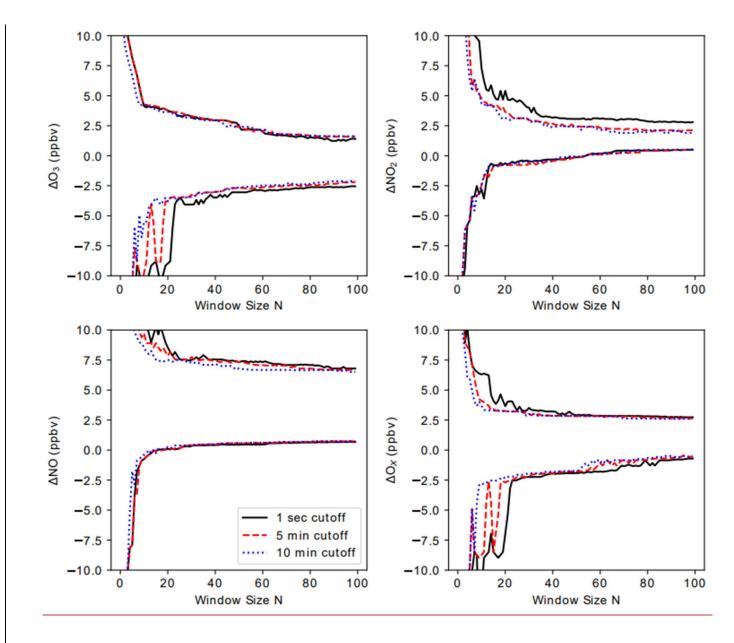
- 810 and lower traces in Figure 8 provides an estimate of the uncertainty in median ∆X due to random spatial variability, and thus a measure of the magnitude of change in systematic measurement bias that we can expect to be observable by this approach. As expected, the range of values decreases with increasing window size as the influence of random spatial and temporal variability on the calculated bias is reduced. At around 30 to 40 hour window size, the range between the minimum and maximum stabilizes and does not reduce appreciably with further aggregation.
- 815 The number of 1-second data points contributing to each 1-hour ΔX value in this analysis is highly variable (Figures S10 and S11). For data within a 3 km buffer distance, the number of 1-second data points in each hour aggregate ranges from just a few seconds to a full hour and is fairly evenly distributed. The time-resolved data from mobile mapping, by its nature, can have significant temporal and spatial variability and using only a few seconds of data to compare to regulatory data with hourly time resolution may result in additional noise and perhaps invalid assessments of instrument bias in the analysis. To
- 820 explore the degree to which including hourly aggregates with only a few seconds of data impact the resulting analysis, we repeated the calculation with a restriction on the amount of mobile data necessary during each hour period for inclusion in the analysis. We used completeness criteria of 5 and 10 minutes, which would require at least 5 minutes (or 10 minutes) of valid data during each hour period for that period to be included in the sample data set for analysis. Note that these 5 or 10 minutes need not have been consecutive. Results are shown alongside the base case (no time-base restriction) in Figure 8. While there
- 825 are only subtle differences in the results between the three completeness criteria, the use of 5 or 10 minute minimum cutoff for each hourly aggregation does result in the convergence of median ΔX values at somewhat smaller window sizes, particularly for O₃. However, atby a rolling window size of around N = 40 hours or higher, the improvement is marginal (e.g., less than a fraction of a ppbv) in most cases. This analysis has an important implication for the design of a mobile data collection plan that incorporates monitoring in the vicinity of stationary monitors for quality assurance purposes while simultaneously meeting
- 830 mobile monitoring objectives. The results in Figure 8 imply that spending only 5-10 minutes, or even less, within 3 km of a monitoring site can be an effective collocation data point, and the more critical parameter is the number of distinct hourly comparisons. This minimizes the effort needed to collect 40 distinct hours near the monitoring site and, thus, the impact on mobile data collection efficiency in areas of interest farther away from stationary monitors. For the California data used in this analysis, the range of ΔX values of around ±4 ppbv for O₃, NO₂, and O_X and ±8 ppbv for NO represent the minimum instrument
- 835 drift we can expect to detect using this method. This is determined as the range between the upper and lower traces in Figure 8 for each pollutant. We find that it is possible to detect this magnitude of instrumental drift over the time with which it takes to collect approximately 40 distinct hourly collocation data points. The time it takes in practices to acquire a 40 hour median ΔX depends heavily on the specific data collection plan. For our dataset, we have ~1600 (for O₃) to ~1900 (for NO₂) hourly datapoints in our 3 km, Non-Highway dataset from one year of driving in California. For a rolling window size of 40, this
- 840 gives an "effective response time" on the order of one week (7 to 9 days), where the response time is the study period (365
 - 41

days) divided by the number of discrete hour periods (1600 to 1900) times the rolling window size (40). This response time is likely atypical, since the objectives of this monitoring plan was specifically to collect data in close proximity to a predetermined set of stationary monitors. On the other hand, most typical mobile monitoring deployments would aim to collect over a much broader area and there would be much higher value in collecting in areas farther removed from where stationary

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monitors exist. Nevertheless, it does provide a framework for planning purposes. For example, if it is desirable to detect instrument drift over a quarterly time period, the mobile collection plan would need to incorporate about 3 separate visits per week to Non-Highway roads within a 3 km radius of a stationary monitor. Importantly, these "visits" do not need to be exclusively 1-hour long visits, and visits of only 5 - 10 minutes or even a few seconds can be viable. While imposing a minimum data completeness criteria might improve the range of running median ΔX values at lower N values, it comes at the

850 cost of a reduction in the amount of data and thus an increase in the "effective response time" (for similar values of N).



855 Figure 8: Influence of window size on the range (maximum and minimum) of running median ΔO₃, ΔNO₂, ΔNO, and ΔO_X values for all Non-Highway roads within 3 km of a fixed reference site. We show the relationship for three "minimum data" cutoff values – 1 second, 5 minutes, and 10 minutes – which define the minimum length of valid data necessary during the hour-long averaging period necessary to include that hour in the sample dataset.

Using a running 40 hour time window, we show amean AO3, ANO2, ANO, and AOx values for 3 km, non-highway

860 roads.

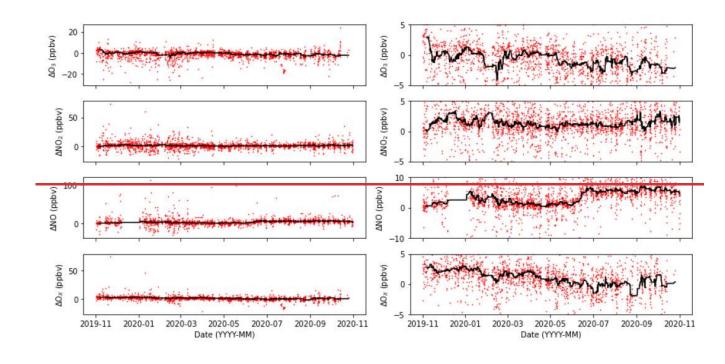


Figure 9: Timeseries of 3 km AO₃, ANO₂, ANO₂, and AO_x, showing individual 1-hour measurements (red points) and 40-sample running median (solid line). The right column truncates many of the 1-hour measurements and scales the axis to highlight the running median.

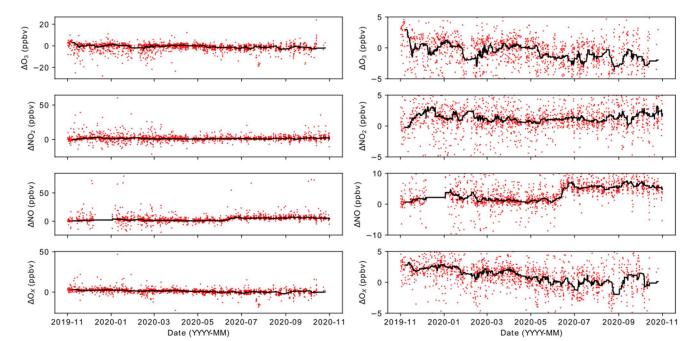
Table 6: Number of hour averaged measurements in the California dataset (non highway roads).

Distance Class	⊖ ₃	NO 2	NO	O *
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

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We examined the timeseries of ΔO₃, ΔNO₂, ΔNO, and ΔO_x for Non-Highway roads for the 3 km distance class (Figure 8), including both hourly median ΔO₃, ΔNO₂, ΔNO, and ΔO_x in Figure 9. Both the raw 1-hour data (red points) and aas well as the 40-sample (40-hour) running median (black line). Figure 9 shows that, despite the-) are included. While there is a large rangedegree of 1-hour ΔX values across the timeseries variability for any individual ΔX value, the running median tends to converge to a small range around zero, reduces that random variability and provides an estimate with values within ±4 ppbv for O₃, NO₂, and O_xquantified uncertainty bounds that can be used to identify drift. 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. systematic bias in mobile measurements in an ongoing basis. In Section 6.2, we apply this method to an example of two NO₂ sensors in Aclima's mobile fleet to show an example of how this method could identify real drift in lower-cost sensors.

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 885 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.



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895 <u>6.2 Case study: NO₂ sensors in Aclima's mobile fleet</u>

To illustrate how the running median method can be used to identify drift in sensors deployed for mobile monitoring, we show an example using NO₂ sensors deployed as part of Aclima's mobile collection fleet. We analyzed mobile versus stationary differences for two NO₂ sensors deployed in two different vehicles across a multi-month deployment in California. In contrast to the AMCL results shown in Section 5 and Section 6.1, these sensors are not regularly calibrated during their mobile deployments. Instead, they are calibrated initially via collocation with the reference instruments in the AMCL (prior to deployment) and calibrated a second time in the AMCL at the end of their deployment period. For Sensor A, the original calibration was found to have held well during the post-deployment calibration check, with a mean bias of +2.2 ppbv. This was well within our ±6 ppbv acceptance criteria for the NO₂ sensor. Sensor B, on the other hand, was found to have a mean bias of -15.5 ppbv during its post-deployment calibration check, indicating significant drift beyond what we consider acceptable performance.

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

While the sensors are deployed, the only possible in-situ evaluation of the calibration of these sensors was using mobile-versus-stationary comparisons with fixed reference sites. Over the course of the deployment for both vehicles, they made frequent passes within 3000 m of various regulatory sites. These sites were not specifically targeted - rather these encounters occurred over the course of typical data collection for hyperlocal air pollution mapping. The hour-averaged results

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for each of these sensors is shown in Figure 10, along with the 40-hour running median. The in situ running median biases compared to the regulatory sites is consistent with the post-deployment bias determinations, indicating that mobile versus stationary comparisons could be used to detect drift in mobile NO₂ sensors while deployed. In addition, it is apparent in Figure 10 that Sensor B had significant discrepancies compared to the fixed reference sites that were outside of the expected ± 6 ppbv uncertainties. Therefore, this method could have been used to identify potential issues with Sensor B during deployment 915 without waiting until a post-deployment calibration. While a more detailed analysis across multiple devices and deployments would be required to establish this approach as an accepted method, this case study demonstrates how such an approach might

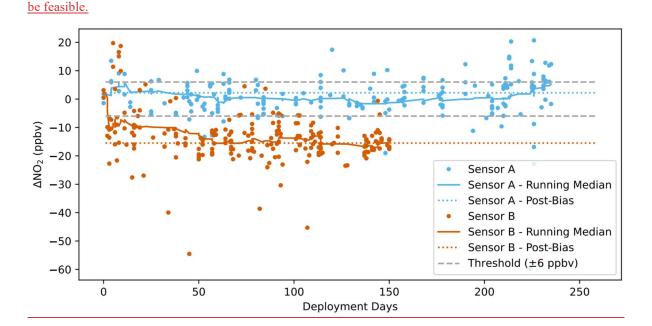


Figure 10: Timeseries of 1-hour ANO₂ and 40-hour running medians for two sensors (Sensor A and Sensor B) deployed 920 as part of Aclima NO₂ sensor deployment. Uncertainty threshold (±6 ppbv) and post-campaign sensor biases also shown.

7 Spatial heterogeneity and implications for applicability to additional pollutants

In this manuscript, we focus on three major pollutants (O3, NO2, and NO) with very different behaviors in the atmosphere. O₃ is predominantly a regionally-distributed secondary pollutant with high background concentrations and negative deviations in direct emission plumes, especially those containing NO (which rapidly titrates O₃). NO₂ is both a

primary and secondary pollutant that has moderate regional background concentrations and falls into the category of a coemitted pollutant with high regional background (Brantley et al., 2014), along with species like PM_{2.5} and PM₁₀. NO is a primary pollutant with a short lifetime, especially in the presence of O₃, and has high peak concentrations and low background concentrations. NO falls into the category of a co-emitted pollutant with low regional background (Brantley et al., 2014), which also includes pollutants such as carbon monoxide, black carbon, and ultra-fine particles.

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In addition to providing valuable insights into deployed sensor data quality, our analysis also has implications for the spatial heterogeneity of these atmospheric pollutants and the spatial representativeness of measurements at stationary sites. If we consider the coefficient of determination (r^2) of the mobile vs stationary regressions to be a simple proxy for spatial homogeneity (setting aside for now the important temporal component to this variability), with higher r^2 indicating a more

- 935 spatially homogeneous pollutant, then we conclude that O_3 is more spatially homogeneous, NO is more heterogeneous and NO₂ is between the two. This is consistent with our understanding of emission sources and atmospheric lifetimes of these different species under typical urban conditions. The temporal component of this spatial variability is, of course, a key consideration and, as such, the linear regressions described by the r^2 values (and shown in Figure S12) are sensitive to the spatial variability within an hourly snapshot, while the relationships shown in Figure 8 describe the spatial variability over
- 940 different temporal aggregations (i.e. the median window size). The field of hyperlocal air quality monitoring is predicated on the fundamental principle that aggregating many samples over time is required to reduce the impact of temporal variability to observe persistent spatial trends in concentration (Apte et al, 2017; Messier et al., 2018; Van Poppel et al., 2013). While the hourly variability between mobile and stationary measurements may be a reasonable proxy for spatial heterogeneity, aggregations over longer durations provide a more accurate measure.
- 945 <u>The high degree of correlation between hourly mobile and stationary measurements might suggest that O₃ has minimal spatial variability. However, our analysis shows that there are measurable spatial gradients in O₃. This is particularly true for Highways, and to a lesser degree Major roads, as compared to Residential roads and stationary sites. Figure 3 for example, clearly shows observable differences in the mobile measurements in Denver compared to the stationary measurements both as a function of distance from the site and as a function of road type. For this reason, our analysis intentionally removes highways</u>
- 950 specifically to reduce the spatial heterogeneity in the data set so that the mobile to stationary comparison can be more readily indicative of measurement bias. However, the full data set could offer a means to more closely investigate the interplay between the hyperlocal spatial patterns of these 3 closely related pollutants (O₃, NO₂, and NO) and the implications for effective emissions controls at hyperlocal scales.
- This relationship between spatial heterogeneity, mobile vs stationary correlation coefficients, and the practicality of using mobile vs stationary comparisons as a quality control method suggests that it is possible to make some educated assumptions about how other pollutants would behave. For example, PM_{2.5} has both primary and secondary sources and would likely show mobile to stationary correlations similar to NO₂, or possibly O₃ in some areas. Conversely, black carbon, carbon monoxide, or ultrafine particles would likely behave more similarly to NO, given their variability in near-source areas results primarily from direct emissions. While we expect significant differences in correlation with stationary sites for different

960 pollutants across the spectrum of spatial variability, the results in Figure 8 for NO, NO₂, and O₃ suggest that all pollutants would likely follow a similar pattern of decreasing range of ΔX with increasing median window size. The key differences that would be expected for different pollutants would be the optimal median window size (although the optimal median window size is similar for all 3 measured pollutants in this study) and the width of the confidence intervals around the central tendency of the differences between mobile and stationary measurements, and thus, the magnitude of systematic measurement bias that 965 could be detected.

8 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 970 inherent in the collocation or comparison process. We used a set of parked and moving mobile monitoring data from a onemonth study in Denver, CO (2014) and compared reference grade NO, NO₂, and O₃ 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 several kilometers 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 975 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 severalmany 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.

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Conflict of Interest

The authors declare no conflicts of interest.

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Data Availability

Data used for this manuscript is proprietary and owned by Aclima, Inc. (https://aclima.io).(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

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 Funding Acquisition ML, BL, PS
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 Methodology AW, ML, BL, and PS
 Project Administration ML, SK, PS
- 1000 Resources –ML, BL, PS Software – AW, ML, BL Supervision – ML, SK, PS Validation – ML, BL, and PS Visualization – AW, ML
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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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