

A machine learning calibration model using random forests to improve sensor performance for lower-cost air quality monitoring

Naomi Zimmerman¹, Albert A. Presto¹, Sriniwasa P.N. Kumar¹, Jason Gu², Aliaksei Hauryliuk¹, Ellis S. Robinson¹, Allen L. Robinson¹, R. Subramanian¹

5 ¹Center for Atmospheric Particle Studies, Carnegie Mellon University, Pittsburgh, 15213, USA

²Sensevere LLC, Pittsburgh, 15222, USA

Correspondence to: R. Subramanian (subu@cmu.edu)

Abstract. Low-cost sensing strategies hold the promise of denser air quality monitoring networks, which could significantly improve our understanding of personal air pollution exposure. Additionally, low-cost air quality sensors could be deployed to

10 areas where limited monitoring exists. However, low-cost sensors are frequently sensitive to environmental conditions and pollutant cross-sensitivities, which have historically been poorly addressed by laboratory calibrations, limiting their utility for monitoring. In this study, we investigated different calibration models for the Real-time Affordable Multi-Pollutant (RAMP) sensor package, which measures CO, NO₂, O₃, and CO₂. We explored three methods: 1) laboratory univariate linear regression, 2) empirical multiple linear regression and 3) machine-learning based calibration models using random forests (RF).

15 Calibration models were developed for 16-19 RAMP monitors (varied by pollutant) using training and testing windows spanning August 2016 through February 2017 in Pittsburgh, PA. The random forest models matched (CO) or significantly outperformed (NO₂, CO₂, O₃) the other calibration models, and their accuracy and precision was robust over time for testing windows of up to 16 weeks. Following calibration, average mean absolute error on the testing dataset from the random forest

20 models was 38 ppb for CO (14% relative error), 10 ppm for CO₂ (2% relative error), 3.5 ppb for NO₂ (29% relative error) and 3.4 ppb for O₃ (15% relative error), and Pearson r versus the reference monitors exceeded 0.8 for most units. Model performance is explored in detail, including a quantification of model variable importance, accuracy across different concentration ranges, and performance in a range of monitoring contexts including the National Ambient Air Quality Standards (NAAQS), and the US EPA Air Sensors Guidebook recommendations of minimum data quality for personal exposure measurement. A key strength of the RF approach is that it accounts for pollutant cross sensitivities. This highlights the

25 importance of developing multipollutant sensor packages (as opposed to single pollutant monitors); we determined this is especially critical for NO₂ and CO₂. The evaluation reveals that only the RF-calibrated sensors meet the US EPA Air Sensors Guidebook recommendations of minimum data quality for personal exposure measurement. We also demonstrate that the RF model calibrated sensors could detect differences in NO₂ concentrations between a near-road site and a suburban site less than 1.5 km away. From this study, we conclude that combining RF models with carefully controlled state-of-the-art multipollutant 30 sensor packages as in the RAMP monitors appears to be a very promising approach to address the poor performance that has plagued low cost air quality sensors.

1 Introduction

Historically, spatial coverage of air quality monitoring stations has been limited by the high cost of instrumentation; urban areas typically rely on a few reference-grade monitors to assess population scale exposure. However, air pollutant concentrations often exhibit significant spatial variability depending on local sources and features of the built environment (Marshall et al., 2008; Nazelle et al., 2009; Pugh et al., 2012; Tan et al., 2014), which may not be well captured by the existing monitoring networks. In the past several years, there has been a significant increase in the development and applications of low-cost sensor-based air quality monitoring technology (Lewis and Edwards, 2016; McKercher et al., 2017; Moltchanov et al., 2015; Snyder et al., 2013). The use of low-cost air quality sensors for monitoring ambient air pollution could enable much denser air quality monitoring networks at a comparable cost to the existing regime. Increasing the spatial density of air quality monitoring would help quantify and characterize exposure gradients within urban areas and support better epidemiological models. Additionally, more highly resolved air quality information can assist regulators with future policy planning, with identification of hot spots or potential areas of concern (e.g., fracking in rural areas) where more detailed characterization is needed, and with risk mitigation for noncompliant zones. Furthermore, low-cost air quality sensors are generally characterized by their compact size and low power demand. These features enable low-cost sensors to be moved with relative ease to rural areas or developing regions where limited monitoring exists.

The two primary requirements of low cost sensors for ambient measurement are 1) hardware that is sensitive to ambient pollutant concentrations, and 2) calibration of the sensors. The latter is the focus of this study. The challenge with low-cost air quality sensor calibration is that the sensors are prone to cross-sensitivities with other ambient pollutants (Bart et al., 2014; Cross et al., 2017; Masson et al., 2015b; Mead et al., 2013). The most common example is for ozone electrochemical sensors, which also undergo redox reactions in the presence of NO₂. Additionally, NO has also been observed to interfere with NO₂, and CO sensors have exhibited some cross-sensitivity to molecular hydrogen in urban environments (Mead et al., 2013). Furthermore, low-cost sensors can be affected by meteorology (Masson et al., 2015b; Moltchanov et al., 2015; Pang et al., 2017; Williams et al., 2013). Most electrochemical sensors are configured such that the reactions are diffusion-limited, and the diffusion coefficient can be affected by temperature (Hitchman et al., 1997); Masson et al. (2015b) have shown that at relative humidity exceeding 75% there is significant error, possibly due to condensation on potentiostat electronics. Lastly, the stability of low-cost sensors is known to degrade over time (Jiao et al., 2016; Masson et al., 2015a). For example, in electrochemical cells, the reagents are consumed over time and have a typical lifetime of 1-2 years.

Deconvolving the effects of cross-sensitivity and stability on sensor performance is complex. Linear calibration models developed in the laboratory perform poorly on ambient data (Castell et al., 2017). Attempts to build calibration models from first principles have shown some success, but the models are difficult to construct and their transferability to new environments remains unknown (Masson et al., 2015b). Accurate and precise calibration models are particularly critical to the success of

dense sensor networks deployed in urban areas of developed countries where concentrations are on the low end of the spectrum of global pollutant concentrations, as poor signal-to-noise ratios and cross-sensitivities may hamper their ability to distinguish between intra-urban sites. As such, there has been increasing interest in more sophisticated algorithms (e.g., machine learning) for low cost sensor calibration. To date, there have been published studies using high-dimensional multi-response models 5 (Cross et al., 2017) and neural networks (Esposito et al., 2016; Spinelle et al., 2015, 2017, De Vito et al., 2008, 2009). Spinelle et al. (2015) showed that artificial neural network calibration models could meet European data quality objectives for measuring ozone (uncertainty < 18 ppb); however, meeting these objectives for NO₂ remained a challenge. In De Vito et al. (2009), the neural network calibration approach was applied to CO, NO₂ and NO_x metal oxide sensors in Italy with encouraging 10 results; in general mean relative error was approximately 30%. Cross et al. (2017) built high-dimensional multi-response calibration models for CO, NO, NO₂ and O₃ which had good agreement with reference monitors (slopes 0.47-0.94, R² 0.39-0.88). Esposito et al. (2016) demonstrated excellent performance with dynamic neural network calibrations of NO₂ sensors (mean absolute error < 2 ppb); however, the same performance for O₃ was not observed. Furthermore, these calibrations have 15 only been tested on a small number of sensor packages. For example, Cross et al. (2017) tested two sensor packages, each containing one sensor per pollutant over a four-month period, of which 35% was used as training data. Spinelle et al. (2015) tested a cluster of sensors in a single enclosure, testing 22 individual sensors in total over a period of 5 months, of which 15% was used as training data. Esposito et al. (2016) reported calibration performance on a single sensor package (5 gas sensors per package for measuring NO, NO₂ and O₃) and the model was tested on four weeks of data.

In this study, we aim to improve the calibration strategies of low-cost sensors using a random-forest-based machine learning 20 algorithm, which, to our knowledge, has not been previously applied to low-cost air quality monitor calibrations. To ensure calibration model robustness, they were developed for 16-19 RAMP monitors and validated for 10-16 RAMP monitors (depending on pollutant), with each monitor containing one sensor per species (CO, CO₂, NO₂, SO₂ and O₃). Furthermore, the study was conducted over a six-month period (August 2016 – February 2017) spanning multiple seasons and a wide range of 25 meteorological conditions. During this period, RAMP monitors were intermittently deployed for air quality monitoring campaigns, resulting in collocation periods ranging from 5.5 to 16 weeks (median 9 weeks). The fitting of the machine learning algorithms is discussed in detail to determine ideal calibration datasets to maximize performance and minimize overtraining. The performance of the random forest models is compared to traditional laboratory univariate linear models, multiple linear regression models, and EPA performance guidelines. The performance of a given model over time is also discussed.

2 Experimental methods

30 2.1 Measurement sites

Measurements were made from August 3, 2016 to February 7, 2017 on the Carnegie Mellon University campus in the Oakland neighbourhood of Pittsburgh, PA. The outdoor ambient testing environment (40°26'31.5"N, 79°56'33"W) is located within a

small (< 100 vehicles) limited access, open air parking lot near the center of campus. It consisted of a mobile laboratory equipped with reference-grade instrumentation (Section 2.3) and adjacent lawn space where the RAMP monitors were mounted on tripods (Section 2.2). The dominant local source at the site is vehicle emissions when vehicles enter and exit the parking lot during the morning and evening rush hours. There was also occasional truck traffic and restaurant emissions from nearby 5 on-campus restaurants. The small size of the parking lot (< 100 cars) and few other local sources means that for most of the day the location is essentially an urban background site. During the measurement period, the site mean (range) ambient temperature and relative humidity were 13°C (-15 to 34 °C) and 71% (27 to 98%), respectively.

10 The RAMP monitors have also been intermittently deployed across the Pittsburgh region as part of ongoing air quality monitoring research. To demonstrate the accuracy of the calibrated RAMP, we also show data from a RAMP monitor which was first calibrated at Carnegie Mellon University and then moved to the Allegheny County Health Department (ACHD, 40°27'55.6"N, 79°57'38.9"W) from February – May 2017. The ACHD site has independent reference monitors for CO, NO₂ and O₃ and thus comparing data from these two sites enables an independent real-world assessment of model performance. The ACHD site is characterized by increased traffic volume, restaurant density and industry relative to the Carnegie Mellon 15 site.

2.2 Real-time Affordable Multi-Pollutant (RAMP) monitor

The study uses the Real-time Affordable Multi-Pollutant (RAMP) monitor, which was developed in a collaboration between Carnegie Mellon University and SenSevere. The RAMP uses the following commercially-available electrochemical sensors from Alphasense Ltd: carbon monoxide (CO, Alphasense ID: CO-B41), nitrogen dioxide (NO₂, Alphasense ID: NO2-B43F), 20 sulfur dioxide (SO₂, Alphasense ID: SO2-B4), and total oxidants (O_x, Alphasense ID: Ox-B431). The unit also includes a nondispersive infrared (NDIR) CO₂ sensor (SST CO2S-A) which contains built-in T (method: bandgap) and RH (method: capacitive) measurement. The experiments involved 95 individual pollutant sensors mounted in 19 unique RAMP monitors. While the collocation period spanned August 2016–February 2017, many sensors were intermittently deployed for air quality campaigns in Pittsburgh, so the collocation period ranged from 30 days to the full study period, depending on the unit. 25 Additionally, calibrations were not built for sensors for which reference data was below detection limits or if reference monitoring units were malfunctioning, reducing the total number of sensors in this experiment to 73, due to issues with the SO₂ and NO₂ reference monitors.

30 The electrochemical sensor outputs were measured using electronic circuitry custom designed by SenSevere optimized for signal stability. The circuitry includes custom electronics to drive the device, multiple stages of filtering circuitry for specific noise signatures, and an analog-to-digital converter for measurement of the conditioned signal. The RAMP monitors are housed in a NEMA-rated weather proof enclosure (Figure 1A) and equipped with GSM cards to transmit data using cellular networks to an online server. The RAMP monitors also log data to an SD card as a fail-safe in case of wireless data transfer issues. The

data is logged to the server at ~15 second resolution and down-sampled to 15-minute averages, which was deemed to be an appropriate time resolution for assessing spatial variability in air pollution exposure and to reduce the size of the dataset and increase computational efficiency. Regulatory bodies typically make their data available at hourly resolution. The sensors sample passively from the bottom of the unit (Figure 1B), with screens installed to protect the sensors. Roughly 3 weeks of 5 measurements of gaseous species, T, and RH are possible on single charge of a built-in 30 amp-hour NiMH battery. The RAMP monitors are either mounted to a steel plate for easy pole mounting or are deployed on tripods approximately 1.5 m above the ground (Figure 1C). In this study, all the RAMP monitors were tripod-mounted at a consistent height.

In their simplest configuration, electrochemical sensors function based on a redox reaction within an electrochemical cell in 10 which the target analyte oxidizes the anode and the cathode is proportionally reduced (or vice versa, depending on target analyte). The subsequent movement of charge between the electrodes produces a current which is proportional to the analyte reaction rate, which can be used to determine the analyte concentration. The Alphasense electrochemical sensors utilize a more complex configuration by using four electrodes (working, reference, counter and auxiliary) to account for zero current changes. 15 Essentially, the auxiliary electrode, which is not exposed to the target analyte, accounts for changes in the sensor baseline signal under different meteorological conditions. Additional details on the theory of operation for electrochemical sensors can be found in Mead et al. (2013).

The RAMP monitors log two output signals from each of the Alphasense sensors: one from the auxiliary electrode and the other from the working electrode. The net sensor response is determined by subtracting the auxiliary electrode signal from that 20 of the working electrode. In theory, for a target analyte a linear relationship should exist between the net sensor signal for that analyte and ambient analyte concentrations, and this expectation forms the basis of univariate linear regression models built from laboratory calibrations. However, as noted in the introduction, even with an auxiliary electrode, electrochemical sensors may 25 insufficiently account for the impacts of temperature (which affects the rate of diffusion) and relative humidity under high humidity conditions where condensation is possible. This has motivated researchers to construct multiple linear regression models (MLR) to account for these temperature and humidity effects (Jiao et al., 2016). While these calibration models typically improve performance relative to univariate linear models (Spinelle et al., 2015, 2017), they typically do not incorporate any cross-sensitivities to other pollutants or any non-linearities in the response. In this study, we attempt to build a calibration model for each analyte with no underlying assumptions regarding the calibration model structure and allow the models to consider directly the full suite of data being reported by the RAMP monitors using a machine learning approach.

30 2.3 Reference instrumentation

Reference measurements were made on ambient air continuously drawn through an inlet on the roof of the mobile laboratory located approximately 2.5 m above ground. Gaseous pollutants were drawn through approximately 4 m of 0.953 cm outer diameter Teflon fluorinated ethylene propylene (FEP) tubing with a six-port stainless steel manifold for flow distribution to

the gas analyzers. Measurements were made using direct absorbance at 405 nm for NO₂ (2B Technologies Model 405 nm), a gas filter correlation infrared analyzer for CO (Teledyne T300U), a non-dispersive infrared analyzer for CO₂ (LICOR 820), UV absorption for O₃ (Teledyne T400 Photometric Ozone Analyzer) and by UV fluorescence for SO₂ (Teledyne T100A UV Fluorescence SO₂ Analyzer). The time resolution for all reference measurements was 1 s.

5

The reference gas analyzers were checked and calibrated weekly using calibration gas mixtures, except for O₃ which is calibrated biannually at a nearby regulatory monitoring site. The CO and NO₂ analyzers experience modest baseline drift between weekly calibrations, on the order of approximately 40 ppb for CO and 2 ppb for NO₂. Hence, baseline pollutant concentrations were normalized to a nearby regulatory monitoring site (Allegheny County Health Department, Air Quality 10 Division, Pittsburgh, PA). The baseline correction was done using a linear regression between the beginning and end of the week on the baseline signals (local source spikes removed). The regression was based on daytime differences, as night time inversions may cause real differences in the baseline signals between the two sites. The gas analyzers at the regulatory monitoring site are checked daily and thus this normalization helped correct for any baseline drift during the days between calibration. No significant drift was observed for CO₂ or O₃.

15 3 Calibration methods

Three calibration methods were evaluated: (1) a laboratory-based univariate linear regression based on net sensor response when exposed to calibration gases, (2): an empirical multiple linear regression of net sensor response, T and RH regressed against reference monitor concentrations, and (3): a random forest machine learning model using net responses from all sensors, T, and RH to predict reference monitor concentrations. Calibration models were constructed for the CO, NO₂, CO₂ 20 and O₃ sensors in each RAMP monitor. In this study, no calibration models were built for SO₂ due to a combination of reference instrument malfunction and SO₂ concentrations measured with the reference instrumentation being below the instrument detection limit (<0.4 ppbv) for most of the campaign (no nearby sources of SO₂). While lab calibrations were conducted for the SO₂ sensors, this data will be the subject of a future publication on air quality in industrial areas where SO₂ is more commonly detected.

25 3.1 Laboratory-based univariate linear regression (LAB)

Prior to outdoor collocation, the sensors inside the RAMP monitors were calibrated in a laboratory environment using a custom manufactured sensor bed and calibration gas mixtures. The sensors were exposed to each step in the calibration window (Table 1) for 20 minutes and a face velocity of 1.2 m/s flowed perpendicular to the sensor surface. This face velocity is in the lower end of the wind speed range in Pittsburgh, PA (e.g. average monthly windspeed over Jan-May 2017 at 2m height is estimated 30 at 2.4-3.4 m/s). The sensor response at each calibration step was averaged once the signal had stabilized (steady sensor output voltage). Temperature and relative humidity were not controlled during the calibration due to lack of available infrastructure

at the time of the study. The temperature was at levels typical of indoor laboratory environments (approx. 20 °C), and the dry calibration gas provided very little humidity (RH <5%). Calibrations were built for CO, NO₂ and CO₂. Laboratory calibrations for O₃ were not performed due to a lack of suitable O₃ calibration gas.

5 The laboratory calibration follows a standard univariate linear regression model of regression net (CO, NO₂) or raw (CO₂) signal against the reference gas concentration (Eq. 1)

$$y_{\text{reference}}(t) = \beta_0 + \beta_1 \times [\text{Net Sensor Response (CO, NO}_2\text{)} \text{ or Raw Sensor Response (CO}_2\text{)}], \quad (1)$$

10 Model performance was evaluated by comparing the calibrated response to reference measurements. We refer to the laboratory univariate linear regression calibration as LAB. Separate LAB calibrations were developed for each sensor (37 individual calibrations, 9-14 per pollutant). Due to difficulty controlling temperature and RH over a wide range of known ambient conditions, we focused on the relationship between analyte response and the calibration gas concentration, which any user with access to basic lab infrastructure can do. While beyond the scope of this study, an improved LAB calibration would
15 involve a chamber with variable T and RH to better match ambient conditions.

3.2 Empirical multiple linear regression (MLR)

Following laboratory calibration, the individual sensors were mounted in the RAMP monitors and deployed outdoors adjacent to the Carnegie Mellon University supersite. The collocation period varied by RAMP, with a minimum collocation period of 6 weeks and a maximum collocation period of the entire 6-month study period. The collocation window varied due to
20 intermittent deployment of some RAMP monitors for ongoing air quality monitoring campaigns in the Pittsburgh area. To build calibration models, the collocation period was separated into a training and testing period identical to that used for the random forest calibration (see Section 3.3). Due to the previously established influence of T and RH on sensor response (Jiao et al., 2016; Masson et al., 2015b; Spinelle et al., 2015, 2017), a multiple linear regression (MLR) model was used to calibrate the output from each sensor using net sensor response to the target analyte (e.g. CO for the CO sensor), T and RH as explanatory
25 variables (Eq. 2), similar to the approach described in a recent a European Union report on protocols for evaluating and calibrating low-cost sensors (Spinelle et al., 2013).

$$y_{\text{reference}}(t) = \beta_0 + \beta_1 \times [\text{Net Sensor Resp. (CO, NO}_2\text{, O}_3\text{)} \text{ or Raw Sensor Resp. (CO}_2\text{)}] + \beta_2 \times T + \beta_3 \times RH, \quad (2)$$

30 The training data was used to calculate the model coefficients (β_0 through β_3) and the model performance was evaluated on withheld testing data. Separate multiple linear regression models were developed for each sensor (73 individual models). We refer to these models as MLR.

3.3 Random forest model (RF)

A random forest (RF) model is a machine learning algorithm for solving regression or classification problems (Breiman, 2001). It works by constructing an ensemble of decision trees using a training data set; the mean value from that ensemble of decision trees is then used to predict the value for new input data. Briefly, to develop a random forest model, the user specifies the 5 maximum number of trees that make up the forest, and each tree is constructed using a bootstrapped random sample from the training data set. The origin node of the decision tree is split into sub-nodes by considering a random subset of the possible explanatory variables (m_{try}). The training algorithm splits the tree based on which of the explanatory variables in each random subset is the strongest predictor of the response. The number of random explanatory variables considered at each node (denoted m_{try}) is tuned by the user. This process of node splitting is repeated until a terminal node is reached; the user can specify the 10 maximum number of sub-nodes or the minimum number of data points in the node as the indication to terminate the tree. For our random forest models, the terminal node was specified using a minimum node size of 5 data points per node.

To illustrate the method, consider building a random forest model for one RAMP monitor using a single decision tree and a subset of 100 training data points to build a CO calibration model (Figure 2). In this highly simplified example, at the first 15 node, the net CO sensor signal is the strongest predictor of the CO reference monitor concentration, with a natural split in the data at a net CO sensor voltage of 255.9 a.u. If sensor voltage exceeds 255.9 a.u., a cluster of 7 data points from the training data predicts an average CO concentration of 357 ppb, if CO net sensor voltage is ≤ 255.9 a.u. then the data goes to the next decision node, in which net CO sensor signal is again the strongest predictor of the CO reference monitor concentration, with a natural break in the data at a net CO sensor voltage of 167.3 a.u. The splitting proceeds until all the training data are assigned 20 to a terminal node. The prediction value for each terminal node is the average reference monitor concentration of training points assigned to that node. To apply the algorithm (i.e., predict the CO concentration from a set of measured inputs), the user takes the measured T and the net CO, NO₂ and O₃ signals and follows the path through the tree to the appropriate terminal node. The predicted CO concentration for that tree is then the average training value associated with that terminal node. This process is then repeated through multiple trees (Figure 2 shows only one simple tree) and the predictions from each tree are 25 averaged to determine the final output from the entire random forest model. In this simple example, there are only six possible CO concentrations the random forest model will output. In practice, each tree has hundreds of terminal nodes and the forest typically comprises hundreds of trees, which means that there are thousands of possible answers. The model prediction for a given set of inputs is the average prediction across all the hundreds of trees that comprise the forest.

30 The random forest model's critical limitation is that its ability to predict new outcomes is limited to the range of the training data set; in other words, it will not predict data with variable parameters outside the training range (no extrapolation). Therefore, a larger and more variable training data set should create a better final model. In this study, our collocation window covered a broad range of concentrations and meteorological conditions; however, in situations where shorter collocation

windows with less diverse training ranges are desired, the RF model may not be suitable as a standalone model. This is discussed further in Section 4.3.2. To maximize utilization of the training data set to avoid missing any spikes during the training window, a k-fold cross validation approach was used. A k-fold cross-validation divides the data into k equal sized groups (where k is specified by the user) and k repeats are used to tune the model. Consider an example where k is equal to 5 (a 5-fold cross-validated random forest model). With a 5-fold validation, five unique random forest models are constructed, one for each fold. In building the first random forest, the first 20% (1/k) of the data will be the testing data, and the remaining 80% [(1-k)/k] of the data will be used as training. In building the second random forest, the next 20% of the data will be used as test data, and the first 20% and remaining 60% will be used to train. This is repeated until the data are fully covered, at which point the random forest model is created by combining the five (k) individual models into one large random forest model. This helps to minimize bias in training data selection when predicting new data, and ensures that every point in the training window is used to build the model.

In this study, reference gas data, RAMP net sensor data for CO, NO₂, SO₂, O₃, and RAMP raw sensor data for CO₂, T, and RH were collected at 15 second resolution, time-matched, and down-averaged to 15 min intervals (IGOR Pro v6.34), which is a higher temporal resolution than the 1 h intervals at which typical regulatory monitoring information are reported and minimized computational cost. The down-sampled data were then imported into R (ver. 3.3.3, “Another Canoe”) for random forest model building. R is an open-source package for tuning and cross-validating many classes of statistical models, including random forest models. The cross-validated random forest models were compiled using the open-source “caret” package (Kuhn et al., 2017). The model considered all RAMP data (net voltage outputs from the five gas sensors plus T and RH, 7 possible variables total) as potential explanatory variables to predict the reference monitor gas concentration. The number of trees was capped at 100 per fold, and a five-fold cross-validation was used for a total of 500 trees. Therefore, the predicted value for a given set of measured inputs is the average value from this set of 500 trees (each tree provides one prediction). The k-value was chosen by identifying the minimum number of folds for which an increase in the fold size increased model performance less than 5% on the held-out data. The number of trees was chosen based on the work of Oshiro et al., (2012), who suggested that the number of trees range from 64-128. The computation time to train a complete RAMP monitor with five sensors was approximately 45 minutes. This was another motivating factor for 15-minute resolution data, as building models at higher time resolutions would have significantly increased computational demand.

When fitting the random forest models with the training data, the main tuning parameter is the number of explanatory variables to consider at each decision node (m_{try}). To determine the optimal m_{try} , the root mean square error (RMSE, equation in Supplemental Information) and the coefficient of determination (R^2) were calculated on the withheld folds of the training data (Figure 3, step 2) for m_{try} equal to 2, 4 or 7 to span the complete variable range. The random subset of explanatory variables considered at each node was chosen based on which value of m_{try} minimized RMSE. The cross-validation and the subset of explanatory variables randomly considered at each node (m_{try}) was tuned using the caret package in R (Kuhn et al., 2017).

Following random forest model generation and tuning, the five 100 tree models were combined to create a final model with 500 trees. This process was repeated for each sensor to create 73 separate random forest models. The final models convert the RAMP output signals into calibrated concentrations. The model conversion was done within R, where it exists as a standalone object compatible with the standard R configuration.

5

Data from three RAMP monitors (15 individual gas sensors) were used to investigate the optimal training period, which was determined by comparing the training data size to mean absolute error (MAE, the average of the absolute value of the residuals). The optimal training period was the period beyond which increases in the length of the training window (and therefore size of the training dataset) no longer resulted in significant reductions in the MAE. The initial training window 10 evaluated was 1 week, and 1 week increments in training period duration were considered until MAE was minimized. The optimal collocation window was determined to be 4 weeks (or 2688 data points at 15-minute resolution). This was evaluated for a consecutive collocation window and for 8 non-consecutive collocation windows equally distributed throughout the whole collocation period (August 2016 – February 2017) in half week increments. Details of this evaluation are provided in the 15 Supplemental Information, but the non-consecutive collocations generally performed slightly better, with reductions in MAE of 12 ppb (4% relative error) for CO, 2 ppm for CO₂ (0.4% relative error), 0.4 ppb for NO₂ (4% relative error), and 1.6 ppb for O₃ (7% relative error) compared to the consecutive four-week collocation. The motivation for exploring non-consecutive 20 collocation windows dispersed throughout the study period was to ensure that the training period covered a complete range of gas species concentrations, temperatures and relative humidity. In practice, the training data utilized in this study is equivalent to collocating the RAMP monitors with reference monitors for 3-4 days every 1-2 months. If non-consecutive collocation is inconvenient or not possible, consecutive collocation may be satisfactory as determined by MAE and other accuracy parameters needed for the application at hand.

3.4 Metrics for performance evaluation

The evaluation of the different models was conducted on 15-minute averaged testing data (i.e., data withheld entirely from model building). Metrics to quantitatively compare the LAB, MLR and RF model output to the reference monitor 25 concentrations included Pearson r, which is a measure of the strength and direction of a linear relationship, and the coefficient of variation of the mean absolute error (CvMAE, Eq. 3). For comparing the RF model performance to other published studies, we also evaluated mean bias error, mean absolute error, slope of the linear regression of RF model calibrated RAMP data and reference data, and coefficient of determination (R²).

$$\text{CvMAE} = \frac{\text{MAE}}{\text{Avg. Reference Conc.}} = \frac{1}{\text{Avg. Reference Conc.}} \times \left[\frac{1}{n} \sum_{i=1}^n |\text{Model}_i - \text{Reference}_i| \right], \quad (3)$$

30

Another useful tool for visually comparing competing models is a target diagram (Jolliff et al., 2009). A target diagram illustrates the contributions of the centered root mean square error (CRMSE, which is RMSE corrected for bias) and the mean

bias error (MBE) towards total RMSE. In a target diagram, the x-axis is the CRMSE, the y-axis is the MBE and the vector distance to the origin is the RMSE. Since CRMSE is always positive, a further dimension is added: if the standard deviation of the model predictions (calibrated sensor data) exceeds the standard deviation of the reference measurements, the CRMSE is plotted in the right quadrants and vice versa. To match previously constructed target diagrams (Borrego et al., 2016; Spinelle et al., 2015, 2017), the CRMSE and MBE were normalized by the standard deviation of the reference measurements, and thus the vector distance in our diagrams is $\text{RMSE}/\sigma_{\text{reference}}$ (nRMSE). The resulting diagram enables visualization of four diagnostic measures: (1) whether the model tends to overestimate ($\text{MBE} > 0$) or underestimate ($\text{MBE} < 0$), (2) whether the standard deviation of the model predictions (calibrated sensor data) is larger (right plane) or smaller (left plane) than the standard deviation of the reference measurements, (3) whether the variance of the residuals is smaller than the variance of the reference measurements (inside circle of radius 1) or larger than the variance of the reference measurements (outside circle), and (4) the error (nRMSE), the vector distance between the coordinate and the origin. Details of equations required to build a target diagram are provided in the Supplemental Information. Model performance metrics were calculated in R (ver. 3.3.3, “Another Canoe”) using the “tdr” package (Perpinan Lamigueiro, 2015).

4 Results and Discussion

15 4.1 Calibration model goodness of fit: comparing model predictions to training data

Following model building, the goodness of fit between the model output concentrations and the reference monitor concentrations during the training window (i.e. the data used to build the model) were evaluated for all three calibration model approaches (laboratory univariate linear regression “LAB”, field-based multiple linear regression “MLR” and field-based random forest “RF”). For the training period, the calibrated CO and O₃ concentrations were all highly correlated (Pearson $r > 0.8$) with the reference monitor concentrations for all the calibration model approaches (Table 2). However, only the RF model achieved strong correlations between the reference monitor and the RAMPs for NO₂ and CO₂ (Pearson $r: 0.99$). Furthermore, CvMAE for each species was $\leq 5\%$ during the training window for the RF models, substantially outperforming the other models.

25 Regression plots for 19 RAMP monitors and for CO, CO₂ and O₃ and 16 RAMP monitors for NO₂ illustrating the goodness of fit of the RF model are provided in the Supplemental Information (Figures S3-S6). Only 16 of the 19 RAMP monitors had an NO₂ calibration, since the NO₂ monitor malfunctioned during the period when three RAMPs were collocated and so a calibration model could not be built for NO₂ for these three RAMPs. For the RF models, Table 2 also provides the random subset of explanatory variables sampled for splitting at each decision node (m_{try}) to achieve the lowest model RMSE. In 30 general, the larger the m_{try} , the simpler the underlying structure of the model. For example, if there is one dominant variable but the model is permitted to consider all 7 explanatory variables at each decision node (i.e., $m_{\text{try}}=7$), then the model will most frequently split the data based on the dominant variable. By contrast, the advantage of a lower m_{try} is that subtle relationships

between explanatory variables and the response can be probed. When randomly selecting fewer explanatory variables ($m_{try}=2$ or 4) at each decision node, the probability of selecting a dominant variable decreases and the model is forced to split the data into sub-nodes based on variables which may have a smaller (but real) effect on the response. If the goodness of fit of the calibration model is improved by decreasing m_{try} , this suggests more complex variable interactions with the response (Strobl et al., 2008).

Using the m_{try} metric, we observed that the underlying RF model structure is the simplest for CO, that some model explanatory variable complexities exist for the O₃ and NO₂ models, and that the CO₂ model is the most complex and relies on subtle relationships between the explanatory variables to best fit the data (lowest m_{try} had the best results). This finding matches our expectations based on the LAB and MLR models; these simpler models performed best for CO and worst for CO₂. The trends in the m_{try} metric highlights the value of the RF model approach which directly accounts for multiple pollutants. This appears to be critical for O₃, NO₂ and CO₂ sensors because they are cross-sensitive to other pollutants. Cross-sensitivities have been shown to have a minimal impact on CO sensors, with the only notable cross-sensitivity being to molecular hydrogen (Mead et al., 2013). The poor performance of linear models at predicting CO₂ concentration is not surprising, as the sensor was observed to measure high concentrations under periods of high relative humidity (e.g., during rain) and in some cases during heavy rain will be saturated at 2000 ppm, the upper limit of the sensor, and then is reset to 400 ppm daily, as per manufacturer recommendations. The increase in CO₂ under high humidity conditions is likely due to the interference of water with CO₂ in the NDIR signal. Linear models are poorly suited to describe this behaviour.

4.2 Evaluation of models using testing data

To test the performance of the three different calibration models, the models were applied to the testing data that were not used for model fitting. The RAMP monitor concentrations after correction using the calibration models were compared to the actual measured reference concentrations (Figure 3, step 5). To illustrate the approach, in Figure 4, we show a very short time-series of the testing data (~48-hour window) for RAMP #1. This RAMP monitor's performance is representative of the average model performance across the RAMP monitors and therefore illustrates the quality of an average model. Figure 4 also shows the calibrated RAMP #1 output regressed against the reference monitor concentration for the entire testing period for all three calibration models (LAB, MLR, and RF). For this period, the RF model outperformed the LAB and MLR models for all pollutants except for CO. Differences between the different models were smallest for CO and O₃ and largest for CO₂ and NO₂; the LAB models essentially did not reproduce the reference concentrations for CO₂ and NO₂. To illustrate the consistency of the RF model calibrated RAMP monitors across the entire suite of monitors, regressions for all the RAMP monitors for O₃ are shown in Figure 5. Regression plots for all RAMP monitors across the other gases are provided in the Supplemental Information (Figures S7-S10).

In this study, any data remaining after training were used to test model performance, provided there were at least 48 hours of testing data (192 data points, each point a 15-minute average). The RAMP sensors that met this threshold and are used to test the model – 16 for CO and O₃, 15 for CO₂, and 10 for NO₂ – had at least 1.4 weeks of testing data and a maximum of 15 weeks, with a median testing dataset of 5 weeks. The amount of data used to test model performance varied by RAMP monitor and 5 by pollutant because reference monitors were occasionally offline for maintenance and calibration, and some RAMP monitors were intermittently deployed for concurrent air quality monitoring campaigns in Pittsburgh. Figure S11 shows examples of testing periods for two RAMPs, one at the low end (#19 with ~2300 testing data points) and one at the high end (#4 with ~10,000 data points), interspersed with training periods (2,688 data points for each sensor.)

10 To assess the overall model performance, two performance metrics (Pearson r and CvMAE) were calculated for each RAMP monitor using the entire testing dataset (Figure 6). The aggregate assessment shows that the MLR and RF models are interchangeable for CO, as both models achieved Pearson $r > 0.9$ and CvMAE < 15%. The LAB model achieved a similar Pearson r , but CvMAE doubled to ~30%. For CO₂, NO₂, and O₃, the RF model substantially outperforms the LAB and MLR calibration models on the testing data. On average, Pearson r exceeded 0.8 for the RF model for CO₂ and NO₂ versus < 0.6 for 15 the LAB and MLR calibration models. Furthermore, the RF model performance was more consistent across the RAMP monitors than the MLR and LAB models. For example, the Pearson r for O₃ ranged from 0.92 to 0.95 for the RF models versus 0.74 to 0.89 for the MLR models. This means that essentially all the RF models for O₃ performed well versus only a subset of the MLR models. The consistency of the different models is indicated by the smaller range in the box plots of Figure 6.

20 To compare the LAB, MLR and RF models, target diagrams were constructed for the four gases using all three calibration models for each RAMP monitor (Figure 7). The target diagrams show that, on average, across the RAMP monitors the random sensor error (distance to origin) was smaller for RF models and the RF models showed the least RAMP-to-RAMP variability (less disperse). This contrasts with the MLR models, whose bias and extent of model standard deviation varied much more widely between RAMP monitors, especially for CO₂. For the LAB models, the error for CO₂ and NO₂ was approximately an 25 order of magnitude larger than for the RF and MLR models and had to be plotted on a separate inset due to their poor performance. Across all gases, the RF models on average were biased towards predicting concentrations slightly lower than the reference (i.e., slight tendency to under-predict, $MBE/\sigma_{reference} < 0$). Thus, we conclude that the low CvMAE, high Pearson r correlations, lowest bias and lowest absolute error characteristics of the RF models for all four gases are significant improvements compared to conventional calibration approaches (LAB and MLR).

30 **4.3 Detailed assessment of RF model performance**

To investigate the performance of the RF models in greater detail, we assessed the effect of amount of testing data on model performance, the relative importance of the seven explanatory variables, the performance of the models across the different concentration ranges, and the number of data points needed in each concentration range to optimize the fit.

4.3.1 Drift over amount of testing data

To assess the effect of testing window size on conclusions regarding RF model performance, we compare the MAE to the number of weeks in the testing window (Figure 8). For all the gas species, the MAE was essentially flat across the RAMP monitors and the 95% confidence interval on the slope included 0; RAMP monitors with more testing data did not have substantially higher (worse) MAE, suggesting the RF models are robust over the study period. For NO₂, the most data available for testing was approximately 8 weeks due to instrument maintenance and repair taking the NO₂ reference monitor offline for 6 weeks of the study. Figure 8 also shows MAE over time from one RAMP, RAMP #4, which remained at the Carnegie Mellon supersite for the entirety of the six-month study. For RAMP #4, MAE was calculated for an increasing cumulative number of weeks forward in time, and again, MAE was consistent (and in some weeks improved) over time.

10 4.3.2 RF model explanatory variable importance

While RF models are non-parametric, some sense of the model structure can be gained by examining the relative importance of the explanatory variables. The importance of each variable was quantified by comparing the percent increase in mean square error (MSE) when an explanatory variable signal is permuted - i.e. the values of the selected variable are randomly shuffled, effectively eliminating this variable from the model (Pearson 2017).. If an explanatory variable strongly affects the model 15 performance, permuting that variable results in a large increase in MSE. Conversely, if a variable is not a strong predictor of the response, then permuting the variable does not significantly increase the MSE. Figure 9 shows for each of the gases (CO, CO₂, NO₂ and O₃) the increase in MSE when the explanatory variables were permuted. For both CO and O₃, the signal from the sensor measuring the target analyte (CO or O₃) is the most important explanatory variable, as expected. For the O₃, the second most important variable was the NO₂ signal, an expected cross-sensitivity, as the ozone sensor measures total oxidants 20 (O₃ + NO₂) (Spinelle et al., 2015).

The explanatory variable importance is more complex for CO₂ and NO₂. For CO₂, all variables are roughly equally important, with CO being the most important. This is likely due to the strong meteorological effect of humidity on the measured CO₂ concentration; the model must rely on other primary pollutants to predict the CO₂ signal when the measured CO₂ has reached 25 full-scale (i.e. becomes saturated in periods of high humidity), and short-term fluctuations of CO₂ are likely from combustion sources (e.g., vehicular traffic in urban areas) which also emit CO. This highlights the value of having sensors for multiple pollutants in the same monitor. Including measurements of additional pollutants helps the RF model correct for cross-sensitivities. However, the drawback of this cross-sensitivity in the model is that the RF model may not perform well in areas where the characteristic source ratios of CO and CO₂ have changed. For example, this model was calibrated in an urban 30 environment with many traffic and combustion-related sources nearby. Such a model would be expected to perform poorly for CO₂ in a heavily vegetated rural environment where CO and CO₂ are not strongly linked. For the NO₂ model, RH was the most important explanatory variable followed by the NO₂ sensor signal, highlighting again the importance of including

meteorological data within sensor packages. The NO₂ model was also more strongly affected by temperature than the other pollutants. We hypothesize that the sensitivity of the NO₂ sensor to ambient NO₂ is suppressed in Pittsburgh, which has low ambient NO₂ concentrations compared to other cities where these sensors have been evaluated (see Table 3). NO₂ is lowest when O₃ is highest in the summer, and thus the NO₂ RF model effectively uses T and RH as indicators for seasonality when NO₂ is low and the sensor response is suppressed. Furthermore, the relatively equal variable importance of several of the explanatory variables within a model suggests that a cluster of sensors measuring many different species is critically important to build robust calibration models. Interestingly, despite low SO₂ concentrations, there was some contribution from the RAMP SO₂ sensor. This may be due to cross-sensitivities within the SO₂ sensor itself, as the SO₂ sensor may respond to more than ambient SO₂, warranting future investigation. However, in general the SO₂ sensor contributed the least to model performance, thus this sensor could be replaced with a more relevant sensor, such as NO, in future iterations of the RAMP monitor. These findings highlight the value of bundling sensors for measuring a suite of pollutants together, as the different sensors can capture (at least to some extent) cross-sensitivities to other pollutants and improve the model performance for other sensors.

4.3.3 RF model performance as a function of ambient concentration

In Section 4.2, predicted concentrations were normalized to average reference monitor concentration to quantitatively compare differences between the calibration models (CvMAE). To evaluate the RF model performance at different reference concentrations, the testing data were divided into deciles for which the median reference monitor concentration, the absolute residual, and the residual normalized to the reference monitor concentration were calculated (Figure 10). For all species, the RF models tended to overestimate at lower concentrations, and underestimate at the highest concentrations. For the CO RF model, the normalized residual is within 10% of the reference monitor concentration by the 20th percentile of the data (>100 ppb), and continues to improve until the 50th percentile when it plateaus at a normalized residual of about 5%. The US EPA requires a limit of detection of 100 ppb for CO instruments used for regulatory monitoring (United States Environmental Protection Agency, 2014), thus our performance meets that goal. In the top decile, the average absolute CO residual for the RF models approximately doubles but the relative error is still around 5%. However, the top decile spans the broadest concentration range due to the lognormal shape of the CO concentration distribution, and these points are difficult to capture in training data sets.

For the CO₂ RF model, agreement with the reference monitor data are within a few percent up to the 90th percentile, when agreement drops to within 5%. This is possibly due to the RF model actively suppressing high CO₂ sensor signals, as the sensor is prone to reading erroneously high concentrations during rain events. Additionally, the top decile of the data spans a wide range of CO₂ concentrations due to the lognormal shape of the CO₂ distribution. As with CO, the NO₂ RF model agreement with the reference monitor plateaus around the 50th percentile mark; however, the NO₂ RF-model error exceeds 100% for the lowest decile (<5 ppb), suggesting an effective sensitivity of the sensor of 5 ppb. For the O₃ RF model, the effective sensitivity

is also around 5 ppb; when the average reference monitor concentration increased from 5 ppb to 10 ppb (from first to second decile), the normalized residual decreased from over 100% to about 20%. The US EPA limit of detection for federal regulatory monitors is 10 ppb for both NO₂ and O₃, suggesting that as with CO, the RF model performance is within 20% of regulatory standards (United States Environmental Protection Agency, 2014).

5

Systematic underprediction at the highest concentrations was also observed and is likely a consequence of the training dataset used to fit the RF model. Unless the range of concentrations in the training data encompasses the range of concentrations during model testing, there will be underpredictions for concentrations in exceedance of the training range due to the RF model's inability to extrapolate. This is also what causes the horizontal feature for some RAMP monitors at high O₃ 10 concentrations in Figure 5, as the model will not predict beyond its training range. Additionally, the performance of the RF model is sensitive to the number of data points at a given concentration and the model performance. To build a robust model, many data points are required at a given concentration to probe the extent of the ambient air pollutant matrix. In this study, the training windows were dispersed throughout the collocation period to ensure good agreement of gas species and meteorological conditions during both the training and testing windows (see Supplemental Information). The RF model may 15 not work well in cases where such a diverse collocation window is not possible or where concentrations are routinely expected to exceed the training window. In such situations, hybrid calibration models such as combined RF-MLR, where MLR is used for concentrations higher than the RF training window range, may be suitable as MLR tends to perform better when concentrations are higher. An example of this approach is provided by Hagan et al. (2017).

20 To illustrate the impact of number of training data points on the RF model, we binned the data for the representative RAMP (RAMP #1) by concentration and the average concentration measured by the reference monitors was plotted against the average concentration from the calibrated RAMP (Figure 11). The uncertainty in the RF model was plotted as the standard deviation of the model solutions from the 500 trees and the bins were colour coded by the number of data points within each bin. Figure 25 11 illustrates that for every pollutant, agreement with the reference monitor and uncertainty in the model prediction was larger for concentration bins containing fewer than 10 data points. This disproportionately impacted the upper end of the pollutant distribution where fewer data points were collected due to the intermittent and variable nature of high pollutant episodes. This suggests that a minimum of 10 data points at a given concentration are needed to adequately train the RF model, which may inform future RF model building. At NO₂ concentrations below 5 ppb, deviations from the 1:1 line were also observed despite the training dataset containing more than 100 data points at these concentrations. As was concluded from Figure 10, 5 ppbv 30 appears to be the sensitivity limit of these low-cost sensors for NO₂.

4.4 Comparison of results to other published studies

In this section, we compare the performance of our RF models to results from other recent studies including the EuNetAir project in Portugal (Borrego et al., 2016) and EPA Community Air Sensor Network (CAIRSENSE) project (Jiao et al., 2016).

Additionally, a handful of studies have tested the field performance of low-cost sensors both ‘out of the box’ with factory calibrations (Castell et al., 2017; Duvall et al., 2016), and after a machine-learning-based calibration (Cross et al., 2017; Esposito et al., 2016; Spinelle et al., 2015, 2017). We compare the performance of our RF models to these studies in Table 3. While several low-cost sensor calibration studies have investigated calibration models within laboratory environments 5 (Masson et al., 2015a; Mead et al., 2013; Piedrahita et al., 2014; Williams et al., 2013), we have elected to limit our comparison to field data.

There was not a substantial difference in performance of the RF model calibrated vs. LAB calibrated RAMP for CO, and 10 performance was best for this pollutant on the ‘out-of-the-box’ factory calibrated performance assessments in EuNetAir and CAIRSENSE, suggesting that rigorous calibration models may not be critical for CO. However, the RAMP CO RF model did provide improved performance (smallest MAE, 38 ppb) at lower average concentrations compared to the EuNetAir study. Similarly, the ‘out-of-the-box’ performance of the CO sensors tested as part of CAIRSENSE and by the 24 AQMesh sensors tested in Castell et al. (2017) was poorer than the RF model calibrated RAMP. Of those studies that used an advanced algorithm 15 to calibrate the sensors (Cross et al., 2017; Spinelle et al., 2017), the CO RF model resulted in the highest R^2 values and slightly lower slopes; the slope closest to 1 was reported by Cross et al. (2017).

For NO₂, the performance of ‘out-of-the-box’ low-cost sensors varied widely and half the sensors in the EuNetAir study (Borrego et al., 2016) reported errors larger than the average ambient concentrations. While the quality of the baseline gas 20 sensing unit remains critical (in which case no calibration should work), we suggest that advanced calibration models, such as those using machine learning, may be critical for accurate measurements of ambient NO₂. Furthermore, sensor performance was correlated with average ambient concentration; studies in areas with higher NO₂ concentrations had the best performance, consistent with our observations (Figure 10). For studies using advanced NO₂ sensor calibration models (Cross et al., 2017; Esposito et al., 2016; Spinelle et al., 2015), Esposito et al. (2016) had the best performance, with a MAE of < 2 ppb; however, this evaluation was done in a location with high NO₂ concentrations, 45 ppbv (Air Quality England, 2015), more than three 25 times higher than the 12 ppbv in Pittsburgh. In addition, they only evaluated one sensor array, so the robustness of the approach is unknown. In our study, the MAEs across the NO₂ RF model RAMPs ranged from 2.6-3.8 ppb, which is almost as good as Esposito et al. (2016), but at less than one third the ambient concentrations. The slope of the HDMR model for NO₂ of Cross et al. (2017) does exceed that of the RAMP RF model, but the R^2 and MAE values are similar between both studies. Similarly, the annual average NO₂ concentrations in 2015 were 15 ppb at the Massachusetts regulatory site used as a reference in Cross 30 et al. (2017) (Massachusetts Department of Environmental Protection, 2016), 3 ppb higher than the average concentration observed in our study. As shown in Figure 10, an increase of a few ppb of NO₂ can result in almost 100% reductions in relative residuals in our model, potentially explaining discrepancies in the slope between our study and Cross et al. (2017). Furthermore, for identical factory calibrated sensors out of the box, such as the Cairclip and AQMesh, a 5 ppb increase in

average NO₂ concentration results in R² values more than doubling. As such, the excellent performance of the RF model for NO₂ at average ambient concentrations of 12 ppbv shows promise.

For O₃, the RF model, the calibrated data from Spinelle et al., (2015), and the measurements from the Aeroqual SM50 (Jiao et al., 2016) performed the best. Good performance from the Aeroqual when measuring NO₂ has also been previously observed (Delgado-Saborit, 2012). However, the results were the most consistent across the RAMP monitors calibrated with RF models, with relative standard deviations of <20% across the 16 RAMPs for all markers of statistical performance. This performance consistency also holds for the CO and NO₂ RF models. The O₃ RF models were built in Pittsburgh, PA, which has historically had issues with NAAQS ozone compliance, thus while our model was seemingly one of the most accurate and robust, some of this performance may be attributed to the higher ambient O₃ concentrations. From this comparison, we conclude that the RAMP monitor calibrated with a RF model is unique in that it is more accurate when considering the combined suite of pollutants (i.e., all pollutants were accurately measured), it is consistent between many units (<20% relative standard deviation in performance metrics across 10-16 monitors), and is precise even at lower ambient concentrations.

15 4.5 RF model calibrated RAMP performance in a monitoring context

We further assess the RAMP monitor performance against three metrics: 1) comparison of a RAMP monitor calibrated at Carnegie Mellon against an independent set of regulatory reference monitors at the Allegheny County Health Department, 2) for NAAQS compliance, and 3) for suitability for exposure measurements as per the US EPA Air Sensor Guidebook (Williams et al., 2014). We also demonstrate the benefit of improved performance of the RF models in a real-world deployment at two nearby sites in Pittsburgh, PA.

From February through May 2017, a RAMP calibrated at the Carnegie Mellon Campus was deployed at the Allegheny County Health Department (ACHD) to test the performance of the RAMP relative to an independent reference monitor (Figure 12). The ACHD site reports data hourly, so RAMP data were down-sampled to hourly averages and the CO, NO₂ and O₃ concentrations were compared (no measurement of CO₂ is made at ACHD). For all pollutants, R² was ≥ 0.75 (CO: 0.85, NO₂: 0.75, O₃: 0.92) and points were clustered around the 1:1 line. NO₂ performed the most poorly, with a large cluster of points in the 5-10 ppb range where the model is known to underperform. The MAE was 49 ppb (17% CvMAE) for CO, 4.7 ppb for NO₂ (39% CvMAE) and, 3.2 ppb for O₃ (16% CvMAE), in line with the performance metrics in Figure 6. At the time of this submission, RAMPs have been collocated with reference monitors at three additional ACHD sites; these comparisons will be the subject of a forthcoming publication.

Regulatory agencies must also report compliance with National Ambient Air Quality Standards (NAAQS). In this study, the time resolution and methods used to assess the effectiveness of the RF models (15 min) do not match the metrics used for

NAAQS. For example, the NAAQS standard for O₃ is based on the maximum daily maximum 8-hour average, and compliance for NO₂ is based on the 98th percentile of the daily maximum 1-hour averages. While acknowledging that the RAMP monitor collocation period was shorter than typical NAAQS compliance periods (e.g. annually for O₃ and across 3 years for NO₂) it is still worth characterizing the RAMP performance using the LAB, MLR and RF models (Figure 13). For the representative 5 RAMP monitor used previously (RAMP #1), daily maximum 8-hour O₃ was in good agreement between the RF calibrated RAMP and the reference monitor, with all data points falling roughly along the 1:1 line (slope: 0.82, 95% CI: 0.81-0.83), while for the MLR model, concentrations were skewed slightly low (slope of 0.65, 95% CI: 0.63-0.67). For NO₂, the 98th percentile of the daily maximum 1-hour averages was 34 ppb for the RF model versus 35 ppb measured using a reference monitor compared to 25 ppb for the MLR model and 51 ppb for the LAB model. The RF model was substantially closer to the reference 10 monitor estimate and the underestimation was only by 1 ppb. Other RF model calibrated RAMP monitors performed similarly, all agreeing within 5 ppb.

Air sensor performance goals by application area are also provided by the US EPA Air Sensor Guidebook (Williams et al., 2014). The performance criteria include maximum precision and bias error rates for applications ranging from education and 15 information (Tier I) to regulatory monitoring (Tier V). The precision estimator is the upper bound of a 90% confidence interval of the coefficient of variation (CV) and the bias estimator is the upper bound of a 95% confidence interval of the mean absolute percent difference between the sensors and the reference (full equations in the Supplemental Information). An overarching goal of RAMP monitor deployments is to use low-cost sensor networks to quantify intra-urban exposure gradients, thus our benchmark performance was Tier IV (Personal Exposure), which recommends that low-cost sensors have precision and bias 20 error rates of less than 30%. For the testing (withheld) periods, we compared the performance of the RF, MLR and LAB models for all the RAMP monitors used in this study to the precision and bias estimators recommended by the US EPA (Figure 1). The performance across the RAMP monitors was summarized using box plots, and only the RF model calibrated RAMPs are suitably precise and accurate for Tier IV (personal exposure) monitoring across CO, NO₂ and O₃. Furthermore, both RF model calibrated CO and O₃ RAMP monitor measurements were below the even more stringent Tier III (Supplemental 25 Monitoring) standards, which recommends precision and bias error rates of <20%. The RF model NO₂ RAMP measurements may reach Tier III in locations with higher NO₂ concentrations.

To demonstrate the improved performance of the RF models in a real-world context, two of the RAMPs used in the evaluation study were deployed for a 6-week period at two nearby sites in Pittsburgh, PA. One RAMP monitor was located on the roof 30 of a building at the Pittsburgh Zoo in a residential urban area, and another was placed approximately 1.5 km away at a near-road site located within 15 m of Highway 28 in Pittsburgh (Figure 15). NO₂ concentrations are known to be elevated up to 200 m away from a major roadway compared to urban backgrounds due to the reaction of fresh NO in vehicle exhaust with ambient O₃ (Zhou and Levy, 2007). Figure 15 shows the diurnal profiles of the RAMPs at the two locations evaluated using the RF and MLR models. The RF model indicates an NO₂ enhancement of approximately 6 ppb at the near-road site (Figure 15, red trace)

compared to the nearby urban residential site (Figure 15, blue trace) and there are notable increases in NO₂ during morning and evening rush hour periods, as expected. However, applying the MLR model to the RAMP data reveals no significant difference between the two sites (Figure 15, bottom diurnal). In fact, the MLR model predicts negative concentrations during the day. The results of this preliminary deployment suggest that the RF model calibrated RAMPs could be suitable for 5 quantification of intra-urban pollutant gradients.

5 Conclusions

This study demonstrates that the RF model applied to the RAMP low-cost sensor package can accurately characterize air pollution concentrations at the low levels typical of many urban areas in the United States and Europe. The fractional error of the models at a 15-minute time resolution was <5% for CO₂, approximately 10-15% for CO and O₃ and approximately 30% 10 for NO₂, corresponding to mean absolute errors of 10 ppm, 38 ppb, 3.4 ppb and 3.5 ppb, respectively. This performance meets the recommended precision and accuracy error metrics from the US EPA Air Sensor Guidebook for Personal Exposure (Tier IV) monitoring. We demonstrate that this degree of sensitivity allows quantification of intra-urban gradients. Furthermore, the calibration models were well-constrained across 10-16 RAMP units (all performance metrics <20% relative standard deviation), and showed minimal degradation over the duration of the collocation study (August 2016 – February 2017),

15

While the iteration of the RAMP used in this study was equipped with an SO₂ sensor, no calibration model was possible due to SO₂ concentrations at our supersite being below reference instrument detection limits. One feature of the RAMP monitor is that the sensors are modular and can be readily replaced. The assessment of explanatory variable importance combined with the sub-detection limit levels of SO₂ during the study suggests that the RAMP monitor did not substantially benefit from the 20 presence of the SO₂ sensor in this urban background environment. Future iterations of the RAMP will be equipped with NO sensors, which may be more relevant in an urban context.

The RF-models described here were built on four weeks of training data equally distributed in 3.5-day periods throughout the entire collocation (examples shown in Figure S11.) This is nominally equivalent to 3-4 days of calibration every 2 months. As 25 previously mentioned, the low-cost sensor modules within the RAMP monitors can be readily replaced, and as such, we recommend for a large urban deployment to prepare a set of sensors at a regulatory monitoring site and to exchange sensors as they malfunction or as calibration models drift. Since the completion of this study, the sensors have been deployed in Pittsburgh for over 4 months, and changes in the calibration models over longer periods of deployment (1 year or more) will be discussed in a future work. Additionally, the sensors were first opened in July 2016, and characterized over the first 7 30 months of exposure to ambient environments. During this period, no significant temporal drift or sensor degradation was observed, but longer observational studies are likely needed to characterize sensor decay and end-of-life.

The calibration models were developed in Pittsburgh, which had higher O₃ and lower NO₂ compared to several published field-based calibrations and measurements with low-cost sensors. Our results and those of other studies demonstrate that low-cost sensor performance generally increases with increasing ambient concentration, but despite this, the RF models for NO₂ had the second lowest mean absolute error (<4 ppbv) even at low NO₂ concentrations. The good performance of the RF models across all pollutants can likely be attributed to the ability of the RF models to account for pollutant and meteorological cross-sensitivities, highlighting the importance of building multipollutant sensor packages.

Overall, we conclude that with careful data management and calibration using advanced machine learning models, low-cost sensing with the RAMP monitors may significantly improve our ability to resolve spatial heterogeneity in air pollutant concentrations. Developing highly resolved air pollutant maps will assist researchers, policymakers and communities in developing new policies or mitigation strategies to enhance human health. Going forward, a random forest calibrated RAMP network of up to 50 nodes will be deployed in Pittsburgh, PA. This robustly calibrated network will help support better epidemiological models, aid in policy planning, and identify areas where more assessment is needed.

Competing interests

Author J. Gu is the CEO of SenSevere, the developer and manufacturer of the RAMP hardware. The extent of J. Gu's involvement was solely in development management, and improvement of the hardware in the RAMP monitors, and not in data analysis. Authors N. Zimmerman and R. Subramanian may in the future act as consultants for SenSevere on low-cost sensor calibration. The data output from the SenSevere hardware in conjunction with the calibration algorithms presented in this paper yields significantly more accurate measurements than previously reported, and are the subject of provisional patent application. The authors declare no other competing interests.

Acknowledgements

Funding for this study was provided by the Environmental Protection Agency (Assistance Agreement Nos. RD83587301 and 83628601), and the Heinz Endowment Fund (Grants E2375 and E3145). N. Zimmerman's funding was provided by the NSERC Postdoctoral Fellowship (PDF-487660-2016). The authors also wish to thank A. Ellis and J.S. Apte for helpful conversations, and M. Schurman Boehm for her assistance with the laboratory calibrations.

References

Air Quality England: Air Pollution Report, 1st January to 31st December 2016, Cambridge Parker Street (Site ID: CAM 1), , 1–4 [online] Available from: <http://www.airqualityengland.co.uk/assets/downloads/airqualityengland-statistics-report->

CAM1-2015.pdf, 2015.

Bart, M., Williams, D. E., Ainslie, B., McKendry, I., Salmond, J., Grange, S. K., Alavi-Shoshtari, M., Steyn, D. and Henshaw, G. S.: High density ozone monitoring using gas sensitive semi-conductor sensors in the lower Fraser valley, British Columbia, *Environ. Sci. Technol.*, 48(7), 3970–3977, doi:10.1021/es404610t, 2014.

5 Borrego, C., Costa, A. M., Ginja, J., Amorim, M., Coutinho, M., Karatzas, K., Sioumis, T., Katsifarakis, N., Konstantinidis, K., De Vito, S., Esposito, E., Smith, P., Andre, N., Gerard, P., Francis, L. A., Castell, N., Schneider, P., Viana, M., Minguillon, M. C., Reimringer, W., Otjes, R. P., von Sicard, O., Pohle, R., Elen, B., Suriano, D., Pfister, V., Prato, M., Dipinto, S. and Penza, M.: Assessment of air quality microsensors versus reference methods: The EuNetAir joint exercise, *Atmos. Environ.*, 147(2), 246–263, doi:10.1016/j.atmosenv.2016.09.050, 2016.

10 Breiman, L.: Random Forests, *Mach. Learn.*, 45, 5–32, 2001.

Castell, N., Dauge, F. R., Schneider, P., Vogt, M., Lerner, U., Fishbain, B., Broday, D. and Bartonova, A.: Can commercial low-cost sensor platforms contribute to air quality monitoring and exposure estimates?, *Environ. Int.*, 99, 293–302, doi:10.1016/j.envint.2016.12.007, 2017.

15 Cross, E. S., Lewis, D. K., Williams, L. R., Magoon, G. R., Kaminsky, M. L., Worsnop, D. R. and Jayne, J. T.: Use of electrochemical sensors for measurement of air pollution: correcting interference response and validating measurements, *Atmos. Meas. Tech. Discuss.*, 2017–138, 1–17, doi:10.5194/amt-2017-138, 2017.

Delgado-Saborit, J. M.: Use of real-time sensors to characterise human exposures to combustion related pollutants, *J. Environ. Monit.*, 14(7), 1824, doi:10.1039/c2em10996d, 2012.

20 De Vito, S., Massera, E., Piga, M., Martinotto, L. and Di Francia, G.: On field calibration of an electronic nose for benzene estimation in an urban pollution monitoring scenario, *Sensors Actuators, B Chem.*, 129(2), 750–757, doi:10.1016/j.snb.2007.09.060, 2008.

De Vito, S., Piga, M., Martinotto, L. and Di Francia, G.: CO, NO₂ and NO_x urban pollution monitoring with on-field calibrated electronic nose by automatic bayesian regularization, *Sensors Actuators, B Chem.*, 143(1), 182–191, doi:10.1016/j.snb.2009.08.041, 2009.

25 Duvall, R. M., Long, R. W., Beaver, M. R., Kronmiller, K. G., Wheeler, M. L. and Szykman, J. J.: Performance evaluation and community application of low-cost sensors for ozone and nitrogen dioxide, *Sensors (Switzerland)*, 16(10), 1–14, doi:10.3390/s16101698, 2016.

Esposito, E., De Vito, S., Salvato, M., Bright, V., Jones, R. L. and Popoola, O.: Dynamic neural network architectures for on field stochastic calibration of indicative low cost air quality sensing systems, *Sensors Actuators, B Chem.*, 231, 701–713, doi:10.1016/j.snb.2016.03.038, 2016.

Hagan, D. H., Issacman-Vanwertz, G., Franklin, J. P., Wallace, L. M. M., Kocar, B. D., Heald, C. L., and Kroll, J. H.: Calibration and assessment of electrochemical air quality sensors by co-location with reference-grade instruments, *Atmos. Meas. Tech. Discuss.*, <https://doi.org/10.5194/amt-2017-296>, in review, 2017.

Hitchman, M. L., Cade, N. J., Kim Gibbs, T. and Hedley, N. J. M.: Study of the Factors Affecting Mass Transport in

Electrochemical Gas Sensors, *Analyst*, 122(November), 1411–1417, doi:10.1039/a703644b, 1997.

Jiao, W., Hagler, G., Williams, R., Sharpe, R., Brown, R., Garver, D., Judge, R., Caudill, M., Rickard, J., Davis, M., Weinstock, L., Zimmer-Dauphinee, S. and Buckley, K.: Community Air Sensor Network (CAIRSENSE) project: Evaluation of low-cost sensor performance in a suburban environment in the southeastern United States, *Atmos. Meas. Tech.*, 9(11), 5281–5292, 5 doi:10.5194/amt-9-5281-2016, 2016.

Jolliff, J. K., Kindle, J. C., Shulman, I., Penta, B., Friedrichs, M. A. M., Helber, R. and Arnone, R. A.: Summary diagrams for coupled hydrodynamic-ecosystem model skill assessment, *J. Mar. Syst.*, 76(1–2), 64–82, doi:10.1016/j.jmarsys.2008.05.014, 2009.

Kuhn, M., Wing, J., Weston, S., Williams, A., Keefer, C., Engelhardt, A., Cooper, T., Mayer, Z., Kenkel, B., The R Core 10 Team, Benesty, M., Lescarbeau, R., Ziem, A., Scrucca, L., Tang, Y., Candan, C. and Hunt., T.: caret: Classification and Regression Training, [online] Available from: <https://cran.r-project.org/package=caret>, 2017.

Lewis, A. and Edwards, P.: Validate personal air-pollution sensors, *Nature*, 535, 29–31, 2016.

Marshall, J. D., Nethery, E. and Brauer, M.: Within-urban variability in ambient air pollution : Comparison of estimation methods, , 42, 1359–1369, doi:10.1016/j.atmosenv.2007.08.012, 2008.

15 Massachusetts Department of Environmental Protection: Massachusetts 2015 Air Quality Report. [online] Available from: <http://www.mass.gov/eea/docs/dep/air/priorities/15aqrpt.pdf>, 2016.

Masson, N., Piedrahita, R. and Hannigan, M.: Approach for quantification of metal oxide type semiconductor gas sensors used for ambient air quality monitoring, *Sensors Actuators, B Chem.*, 208, 339–345, doi:10.1016/j.snb.2014.11.032, 2015a.

Masson, N., Piedrahita, R. and Hannigan, M.: Quantification method for electrolytic sensors in long-term monitoring of 20 ambient air quality, *Sensors (Switzerland)*, 15(10), 27283–27302, doi:10.3390/s151027283, 2015b.

McKercher, G. R., Salmond, J. A. and Vanos, J. K.: Characteristics and applications of small, portable gaseous air pollution monitors, *Environ. Pollut.*, 223, 102–110, doi:10.1016/j.envpol.2016.12.045, 2017.

Mead, M. I., Popoola, O. A. M., Stewart, G. B., Landshoff, P., Calleja, M., Hayes, M., Baldovi, J. J., McLeod, M. W., Hodgson, T. F., Dicks, J., Lewis, A., Cohen, J., Baron, R., Saffell, J. R. and Jones, R. L.: The use of electrochemical sensors for 25 monitoring urban air quality in low-cost, high-density networks, *Atmos. Environ.*, 70, 186–203, doi:10.1016/j.atmosenv.2012.11.060, 2013.

Moltchanov, S., Levy, I., Etzion, Y., Lerner, U., Broday, D. M. and Fishbain, B.: On the feasibility of measuring urban air pollution by wireless distributed sensor networks, *Sci. Total Environ.*, 502, 537–547, doi:10.1016/j.scitotenv.2014.09.059, 2015.

30 Nazelle, A. De, Rodríguez, D. A. and Crawford-brown, D.: Science of the Total Environment The built environment and health : Impacts of pedestrian-friendly designs on air pollution exposure, *Sci. Total Environ.*, 407(8), 2525–2535, doi:10.1016/j.scitotenv.2009.01.006, 2009.

Oshiro, T. M., Perez, P. S. and Baranauskas, J. A.: How Many Trees in a Random Forest?, in Machine Learning and Data Mining in Pattern Recognition: 8th International Conference, MLDm 2012, Berlin, Germany, July 13-20, 2012. Proceedings,

edited by P. Perner, pp. 154–168, Springer Berlin Heidelberg, Berlin, Heidelberg., 2012.

Pang, X., Shaw, M. D., Lewis, A. C., Carpenter, L. J. and Batchellier, T.: Electrochemical ozone sensors: A miniaturised alternative for ozone measurements in laboratory experiments and air-quality monitoring, *Sensors Actuators, B Chem.*, 240, 829–837, doi:10.1016/j.snb.2016.09.020, 2017.

5 Pearson, R.: Assessing Variable Importance for Predictive Models of Arbitrary Type. Available at <https://cran.r-project.org/web/packages/datarobot/vignettes/VariableImportance.html>, 2017.

Perpinan Lamigueiro, O.: tdr: Target Diagram, [online] Available from: <https://cran.r-project.org/package=tdr>, 2015.

Piedrahita, R., Xiang, Y., Masson, N., Ortega, J., Collier, A., Jiang, Y., Li, K., Dick, R. P., Lv, Q., Hannigan, M. and Shang, L.: The next generation of low-cost personal air quality sensors for quantitative exposure monitoring, *Atmos. Meas. Tech.*, 7(10), 3325–3336, doi:10.5194/amt-7-3325-2014, 2014.

10 Pugh, T. A. M., Mackenzie, A. R., Whyatt, J. D. and Hewitt, C. N.: Effectiveness of Green Infrastructure for Improvement of Air Quality in Urban Street Canyons, *Environ. Sci. Technol.*, 46(14), 7692–7699, 2012.

Snyder, E. G., Watkins, T. H., Solomon, P. A., Thoma, E. D., Williams, R. W., Hagler, G. S. W., Shelow, D., Hindin, D. A., Kilaru, V. J. and Preuss, P. W.: The changing paradigm of air pollution monitoring, *Environ. Sci. Technol.*, 47(20), 11369–15 77, doi:10.1021/es4022602, 2013.

Spinelle, L., Aleixandre, M. and Gerboles, M.: Protocol of evaluation and calibration of low-cost gas sensors for the monitoring of air pollution, *Eur. Comm. JRC Technical Reports*, EUR 26112, doi:10.2788/9916, 2013.

Spinelle, L., Gerboles, M., Villani, M. G., Aleixandre, M. and Bonavitacola, F.: Field calibration of a cluster of low-cost available sensors for air quality monitoring. Part A: Ozone and nitrogen dioxide, *Sensors Actuators, B Chem.*, 215, 249–257, 20 20 doi:10.1016/j.snb.2015.03.031, 2015.

Spinelle, L., Gerboles, M., Villani, M. G., Aleixandre, M. and Bonavitacola, F.: Field calibration of a cluster of low-cost commercially available sensors for air quality monitoring. Part B: NO, CO and CO₂, *Sensors Actuators, B Chem.*, 238, 706–715, doi:10.1016/j.snb.2016.07.036, 2017.

Tan, Y., Lipsky, E. M., Saleh, R., Robinson, A. L. and Presto, A. A.: Characterizing the Spatial Variation of Air Pollutants 25 and the Contributions of High Emitting Vehicles in Pittsburgh, PA, 2014.

United States Environmental Protection Agency: Appendix D, Measurement Quality Objectives and Validation Templates, in *QA Handbook Volume II*, pp. 5–12., 2014.

Williams, D. E., Henshaw, G. S., Bart, M., Laing, G., Wagner, J., Naisbitt, S. and Salmond, J. a: Validation of low-cost ozone measurement instruments suitable for use in an air-quality monitoring network, *Meas. Sci. Technol.*, 24(6), 65803, 30 doi:10.1088/0957-0233/24/6/065803, 2013.

Williams, R., Kilaru, V. J., Snyder, E. G., Kaufman, A., Dye, T., Ruttler, A., Russell, A. and Hafner, H.: Air Sensor Guidebook, EPA/600/R-14/159., 2014.

Zhou, Y. and Levy, J. I.: Factors influencing the spatial extent of mobile source air pollution impacts: a meta-analysis, *BMC Public Health*, 7(1), 89, doi:10.1186/1471-2458-7-89, 2007.

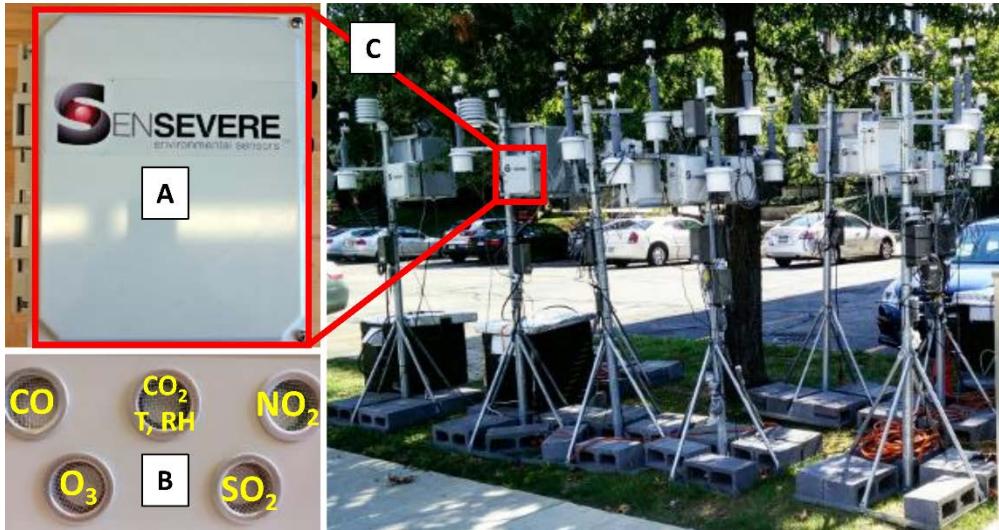


Figure 1: Photographs of the RAMP monitors and the sampling set up. (A) Front view of the RAMP unit in the NEMA-rated enclosure. (B) Bottom view of the RAMPs with sensor layout labelled in yellow. (C) Example of collocation set-up using tripod mounting (not pictured: supersite containing the reference monitors, immediately beside the tripods).

5

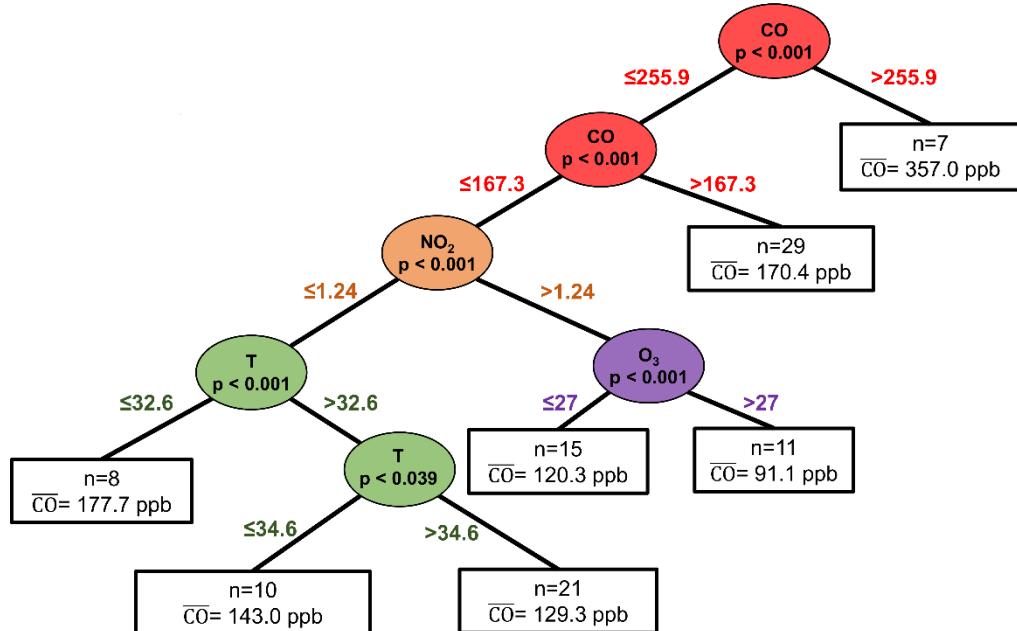


Figure 2: Simplified illustration of one potential CO random forest tree for one RAMP using 100 data points (the trees within the actual models are significantly more complex and 500 such trees are included in the final models). Tree nodes are coloured by splitting variable and split point is overlaid on the branch (e.g., at first split, points with CO sensor signal > 255.9 a.u. are sent to a terminal node, the remaining points go to the next splitting node). \bar{CO} is the average CO reference monitor concentration (ppb) in each terminal node; n = number of data points in each terminal node.

10

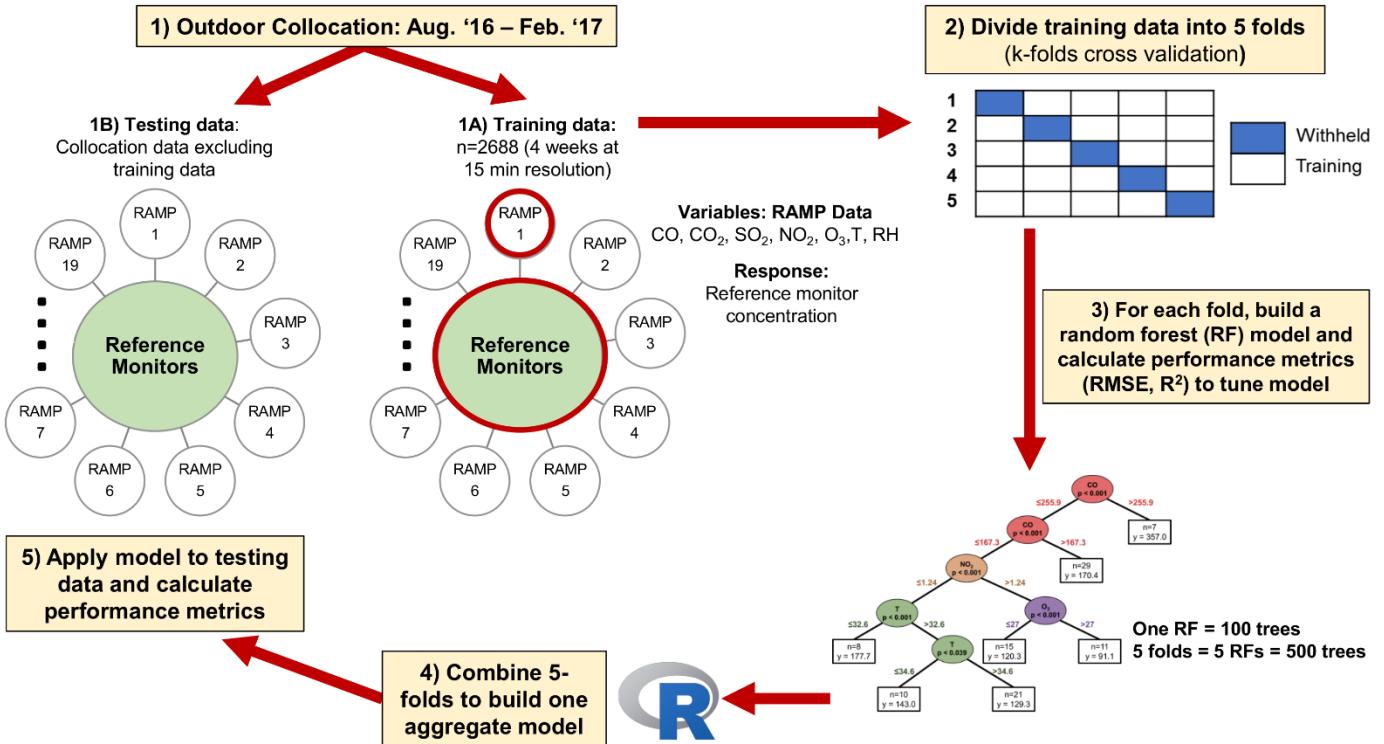


Figure 3: Flow path for data collection and RF model fitting and testing. From collocation period, 2688 points were sub-selected as training (1A) data while the remaining data were used for model testing (1B). The training data were further divided into 5 cross-validation folds and each fold was used to tune and build an RF model. All five models were then combined in R to build one cumulative model and the predictive power of the model was assessed for the withheld testing data.

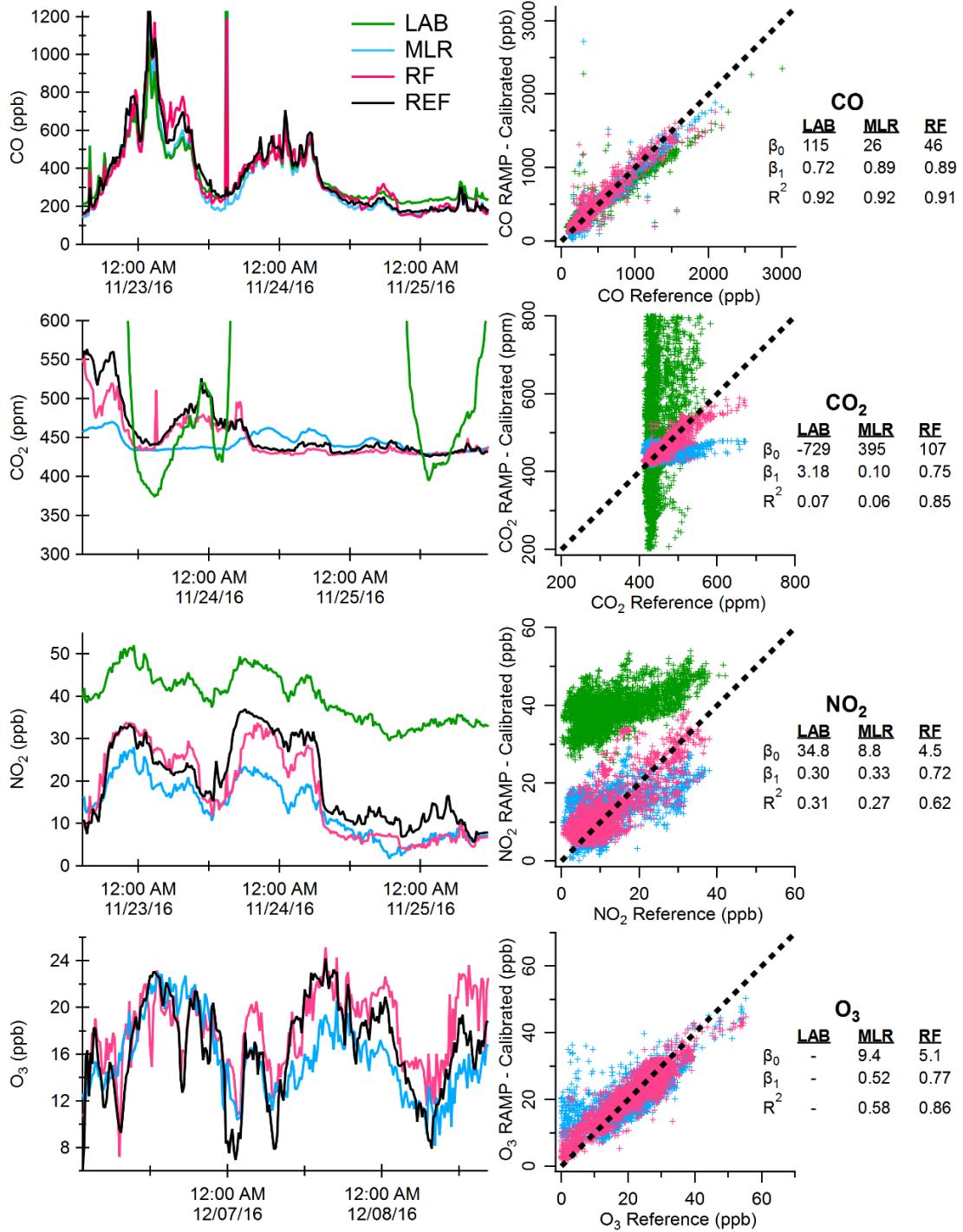
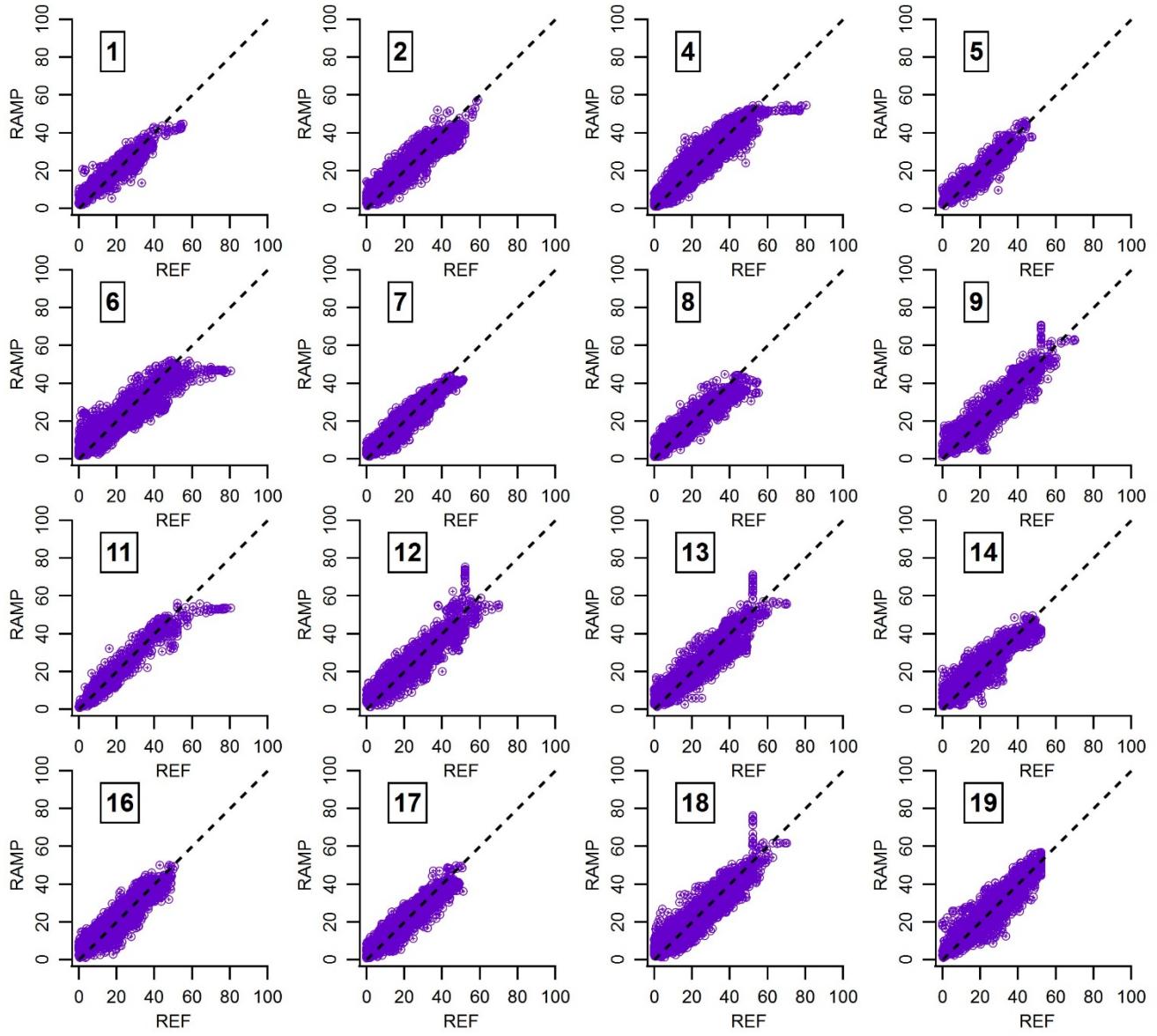


Figure 4: Example time series and regressions comparing the reference monitor data (black) to statistically average RAMP (RAMP#1) using LAB model (green), multiple linear regression (MLR) model (blue) and random forest (RF) model (pink). The left panel shows only 48 hrs of time series data to illustrate approach; the full evaluations (Table 3) were performed with much larger testing datasets; example regressions from the full data set for RAMP #1 are shown in the right panel.



O₃ (ppb)
1:1 line: - - -

Figure 5: RF model performance for ozone evaluated using the testing data (data withheld from building model). Correlation plots show predicted ozone concentration (“RAMP”) versus the reference monitor concentration (“REF”) for 16 RAMP units. All values are in ppb, and the 1:1 line is drawn as a black dashed line.

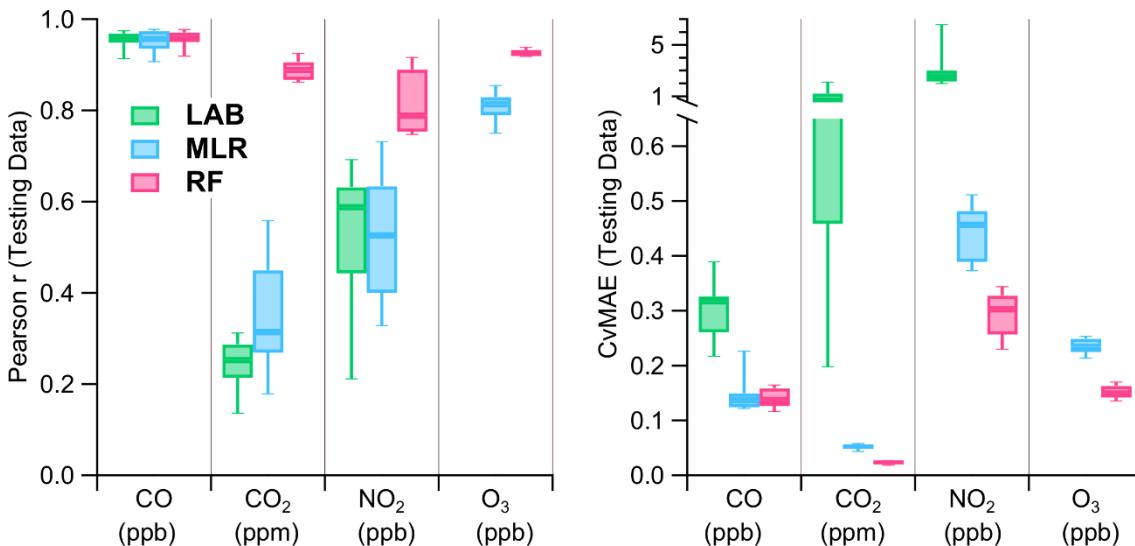


Figure 6: Performance of different calibration models against reference monitor testing data (data not included in model fitting). *Left:* Pearson r correlation coefficient (higher = better, maximum of 1) of different calibration models ('LAB', green; 'MLR', blue; 'RF', pink) versus reference monitor. *Right:* The CvMAE (coefficient of variation of the MAE; MAE normalized by average reference concentration, lower = better) for the three calibration methods. The box plots show the range across the 10-16 RAMPs (whiskers: 10th and 90th percentile, box edges: 25th and 75th percentile).

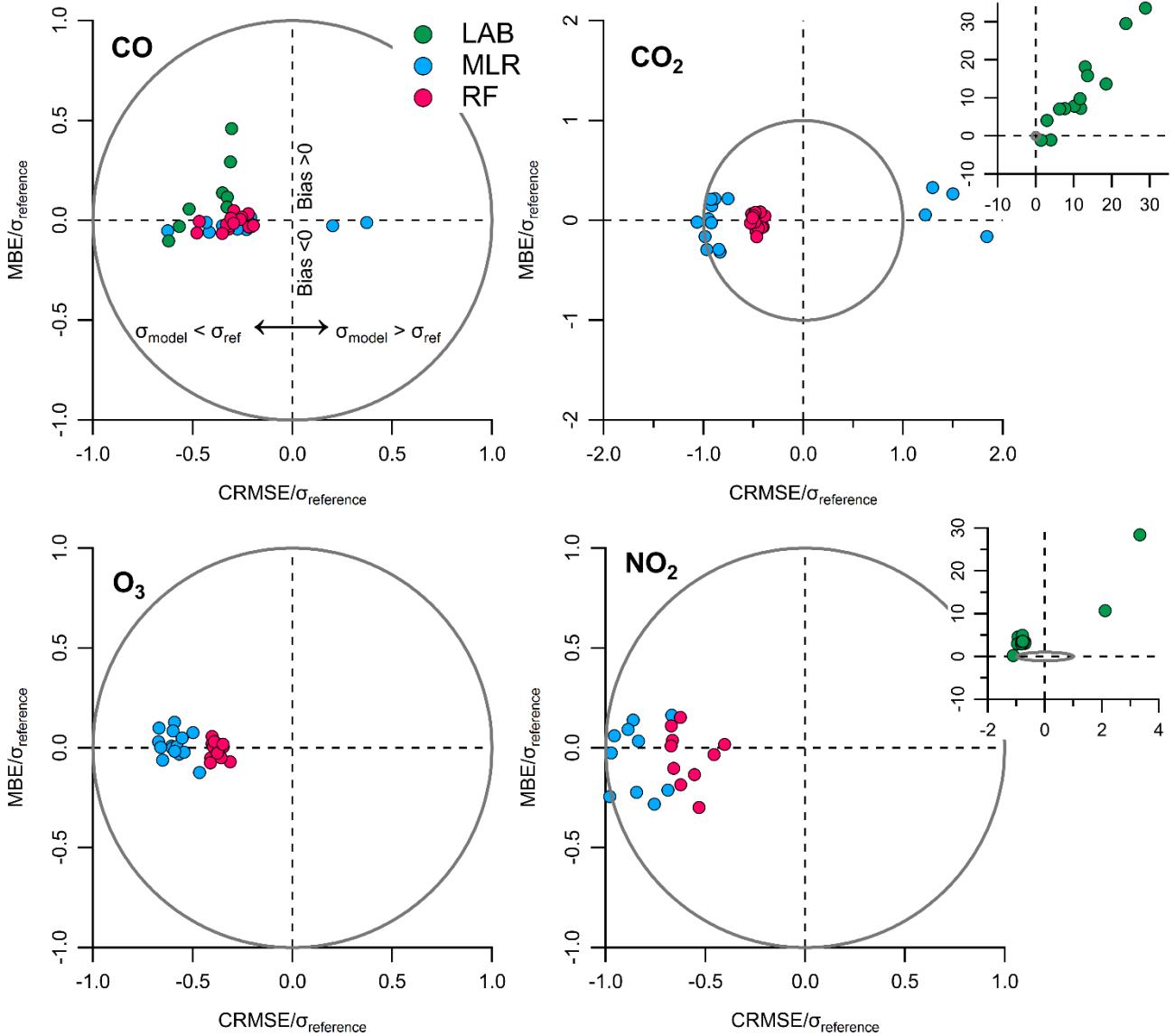


Figure 7: Target diagrams for CO, CO₂, NO₂ and O₃ to compare the LAB, MLR and RF model performance. The y-axis is the bias relative to the reference and the x-axis is the bias-adjusted RMSE (CRMSE) normalized by reference monitor standard deviation; the vector distance between any given point and the origin is the RMSE normalized by the standard deviation of the reference measurements. The CRMSE is in the left plane if model standard deviation is smaller than the standard deviation of the reference observations, and vice versa. If data falls within the circle, then the variance of the residuals is smaller than the variance of the reference measurements. The target diagram for the LAB model for CO₂ and NO₂ is shown in the inset figure because of the order of magnitude difference in MBE and CRMSE compared to the MLR and RF models.

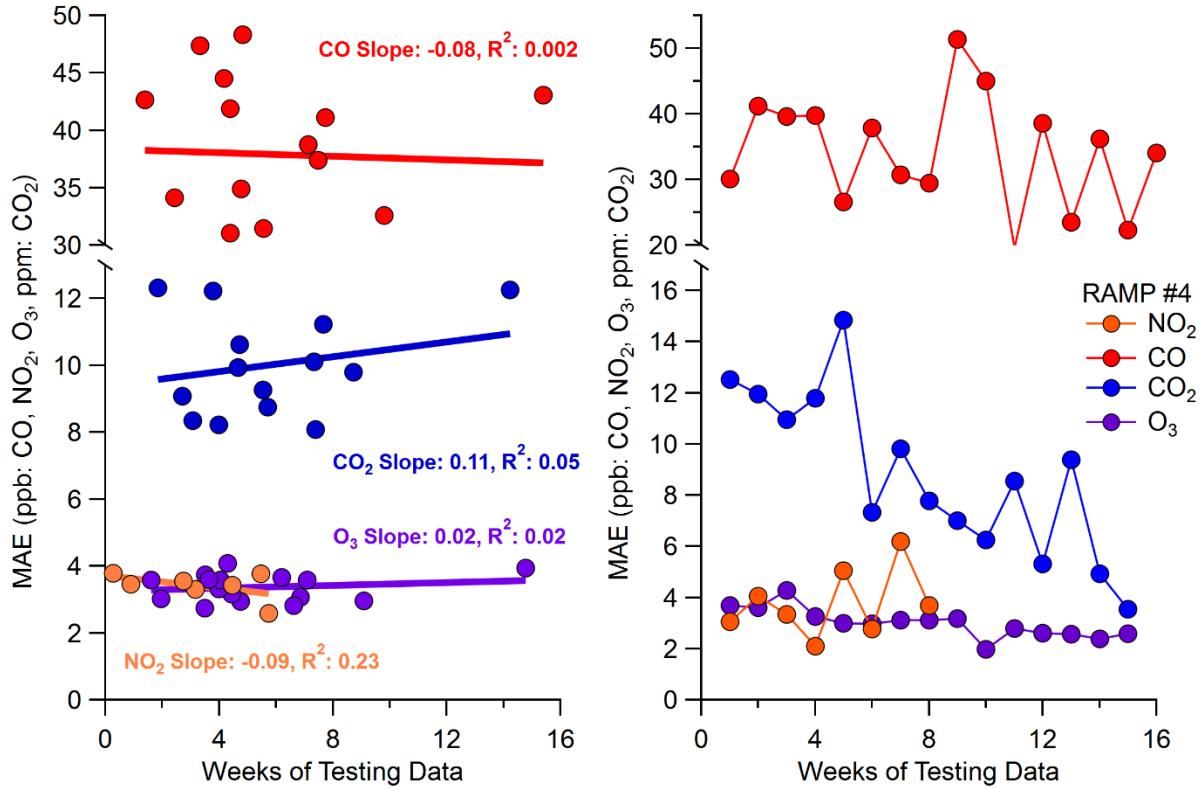
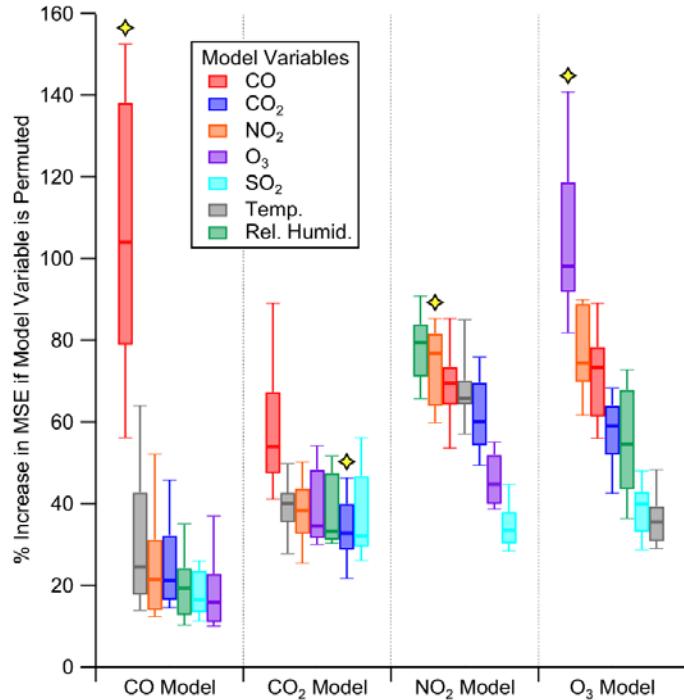


Figure 8: Left: Mean absolute error (MAE) versus the length of the testing period for CO (red), CO₂ (blue), NO₂ (orange) and O₃ (purple) for all the RAMPs. **Right:** Changes in MAE over time for the RAMP with the longest testing window (RAMP #4). The figure shows that the MAE is generally unchanged (or in some cases improves) as the amount of testing data increases, suggesting the RF models are stable over the study period.



5 **Figure 9: Importance of the explanatory variables to each of the RF models. For each model, the explanatory variables are rank ordered from most to least important, and the sensor response corresponding to the target analyte is marked with a yellow star. The box plots represent the range of importance across the 10-16 RAMPs (whiskers: 10th and 90th percentile, box edges: 25th and 75th percentile). The relative importance is determined by calculating the increase in mean square error if the explanatory variable is permuted (i.e., randomly shuffled).**

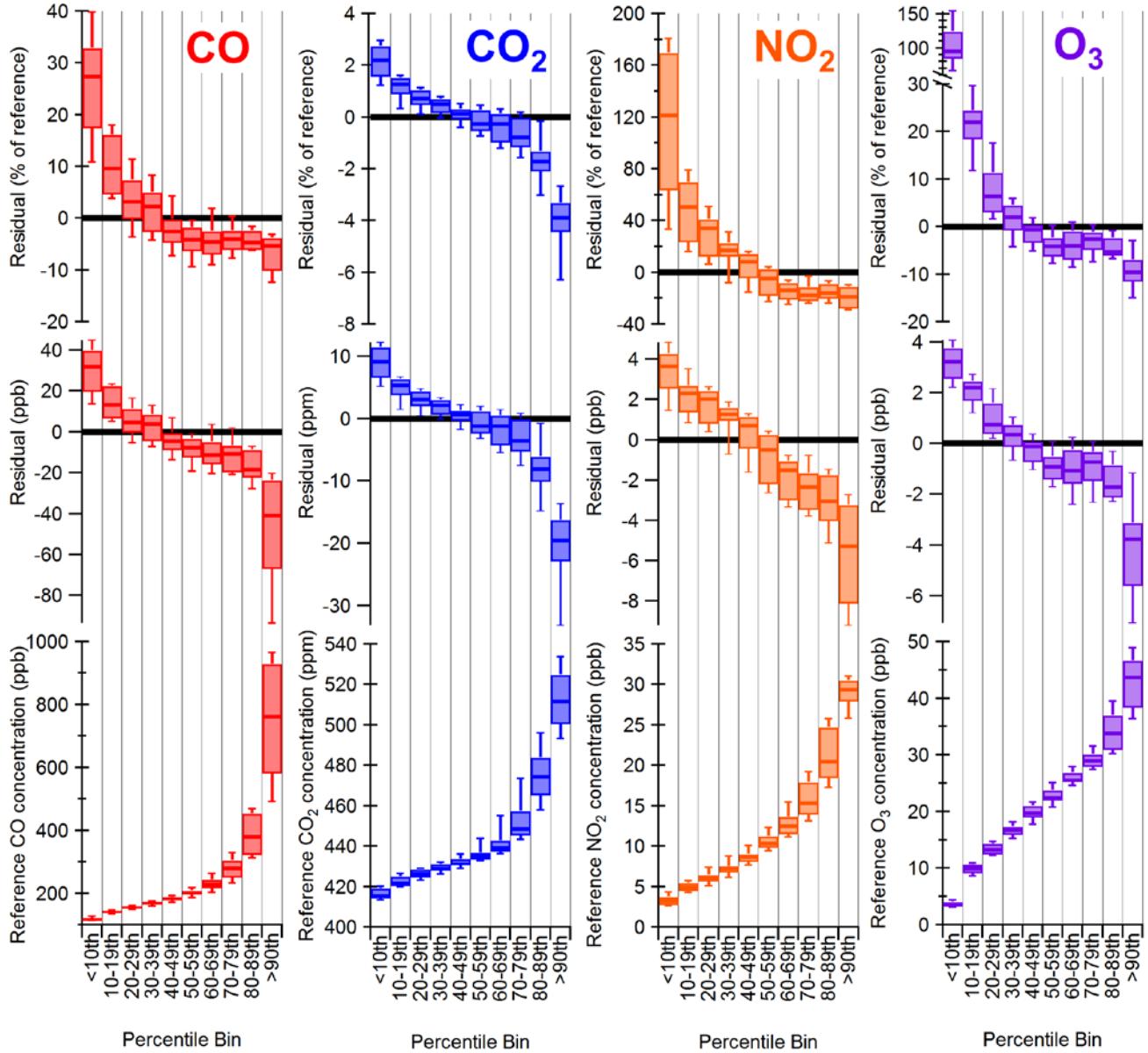
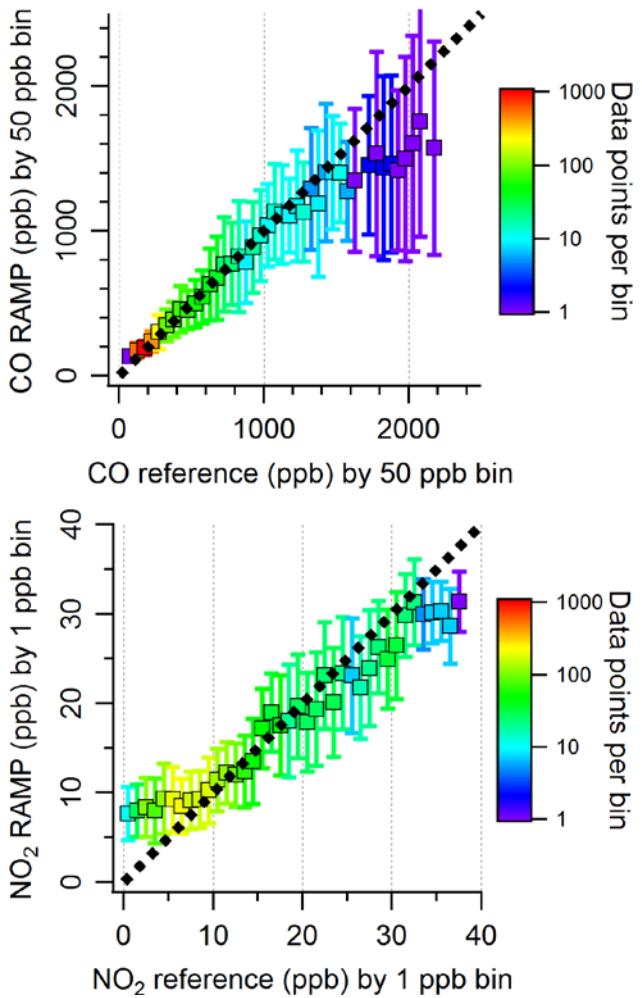


Figure 10: Box plots from the 10-16 RAMP monitors of median concentrations measured by monitors (bottom) and median model residuals (middle) and model residuals normalized to the reference concentration (top) for each pollutant, divided into deciles. The box plots provide the range of medians by the different RAMP monitors.



5

Figure 11: Illustrating the range of predictions from the 500 trees for RAMP #1. The testing data were binned and averaged. The concentration measured by the calibrated RAMP monitors is then plotted against the average concentration from the reference monitor. The error bars represent the standard deviation of the answers from the 500 trees and the bins are colour coded by the number of data points within each bin. The dashed black line is the 1:1 line.

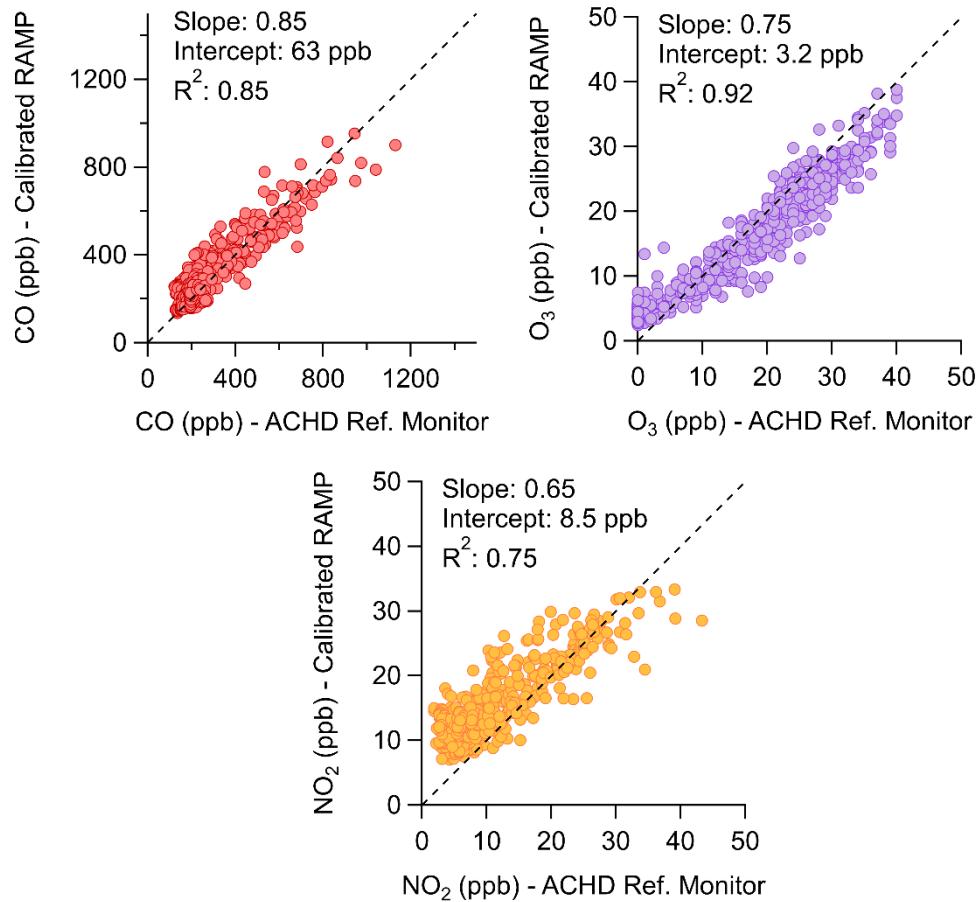
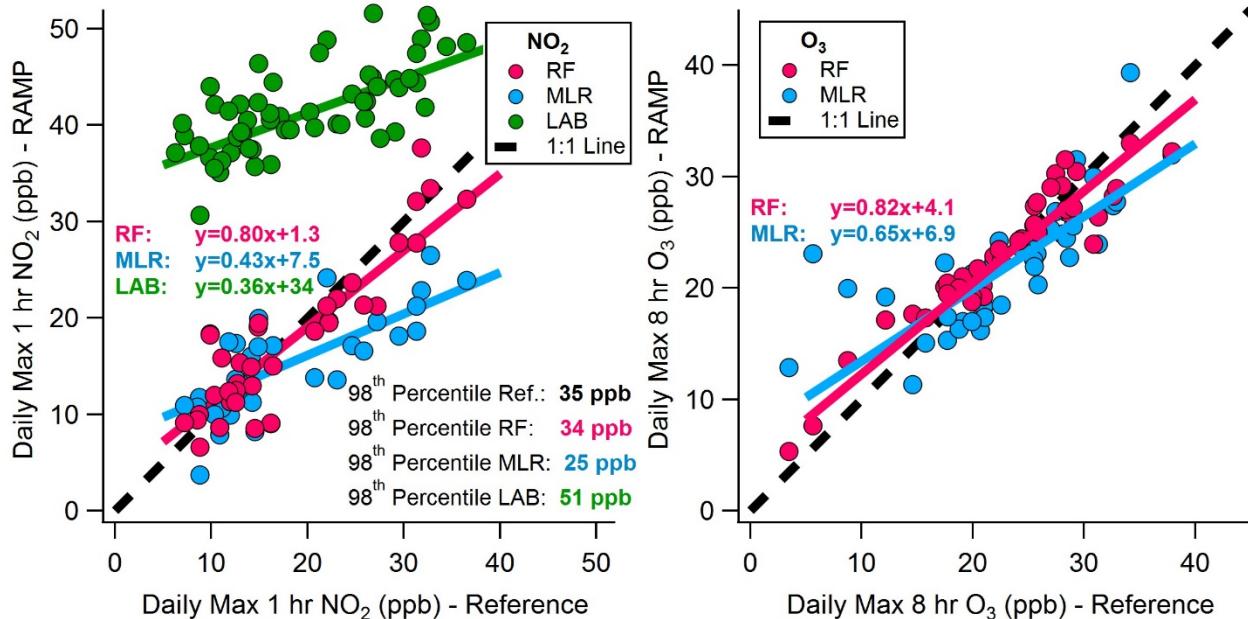


Figure 12: Comparison of CO, NO₂ and O₃ hourly average concentrations measured by a co-located RAMP monitor and the reference monitors at the Allegheny County Health Department (ACHD). The RAMP monitor was first calibrated on the Carnegie Mellon campus prior to deployment.



5

Figure 13: Performance of one representative RAMP (RAMP#1) for NAAQS compliance metrics (O₃: Daily Max 8 h, NO₂: 98th percentile of Daily Max 1 h averages) Right: comparison of daily 8 hr maximum reference monitor ozone concentrations (x-axis) to MLR and RF models. Left: comparison of daily 1 h maximum reference monitor concentrations versus the LAB, MLR and RF models. The NO₂ standard is the 98th percentile of the daily 1 h maximums.

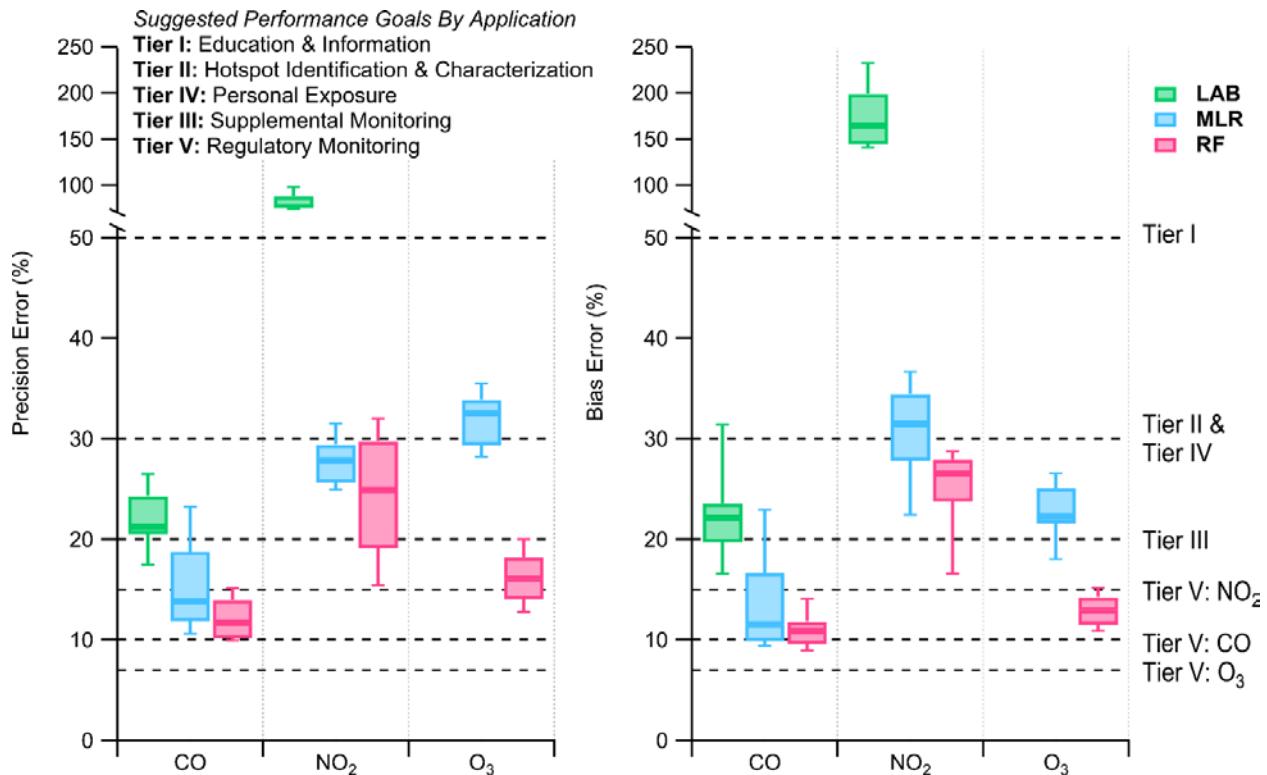


Figure 14: Precision (left) and bias (right) estimates of RAMP monitors calibrated using LAB, MLR, and RF models compared to the suggested performance goals by application as recommended in the EPA Air Sensor Guidebook. The precision estimator is the upper bound of the coefficient of variation (upper bound of the relative standard deviation, RSD). The box plots are the range of performance across the calibrated RAMP monitors (testing data only). The calibrated RAMP monitors meet the recommended error limits for exposure (Tier IV).

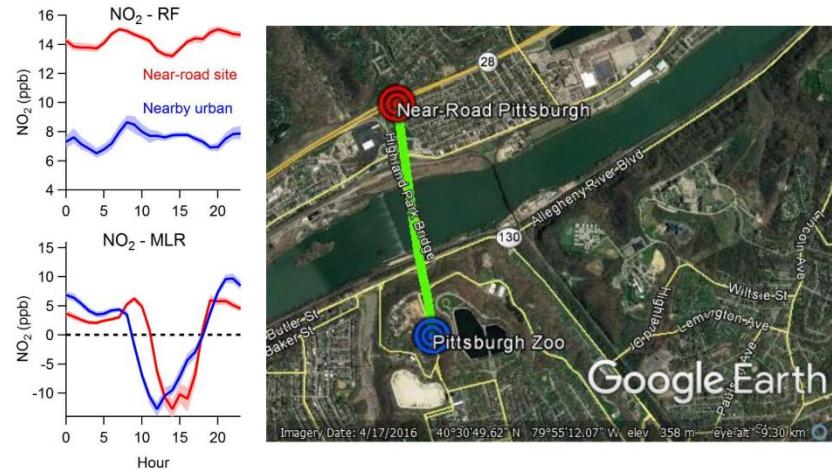


Figure 15: Left: Diurnal NO₂ patterns at two nearby sites (one urban, one near-road) measured by RAMP monitors calibrated using RF models (top) or MLR models (bottom), Right: Satellite view of the two sites, which were ~1.5 km apart. The urban site was at the Pittsburgh Zoo and the near-road site was within 15 m of Highway 28.

Table 1: Calibration ranges for laboratory-based calibration (LAB)

Pollutant	Calibration Range	Points per Calibration
CO	0 – 1600 ppb	3-4
NO ₂	0 – 50 ppb	3-4
CO ₂	0 – 500 ppm	3-4

5 **Table 2: Performance metrics for fits to training data (i.e., goodness of fit) discussed in Section 4.1**

Type	Species	# RAMPs	Avg. Pearson r (\pm SD)	Avg. MAE (\pm SD)	Avg. CvMAE (\pm SD)	β_0 (\pm SD)	β_1 (\pm SD)	β_2 (\pm SD)	β_3 (\pm SD)
LAB	CO	9	0.99 (\pm 0.01)	132 (\pm 32 ppb)	38% (\pm 17%)	-119 (\pm 53)	0.82 (\pm 0.69)	-	-
	CO ₂	14	0.99 (\pm 0.01)	28 (\pm 24 ppm)	24% (\pm 12%)	20 (\pm 36)	0.98 (\pm 0.13)	-	-
	NO ₂	14	0.99 (\pm 0.01)	35 (\pm 8 ppb)	188% (\pm 48%)	-14 (\pm 4.9)	0.62 (\pm 0.15)	-	-
Type	Species	# RAMPs	Avg. Pearson r (\pm SD)	Avg. MAE (\pm SD)	Avg. CvMAE (\pm SD)	β_0 (\pm SD)	β_1 (\pm SD)	β_2 (\pm SD)	β_3 (\pm SD)
MLR	CO	19	0.94 (\pm 0.06)	39 (\pm 13 ppb)	15% (\pm 5%)	32 (\pm 50)	1.3 (\pm 0.2)	-1.1 (\pm 2.8)	-0.1 (\pm 0.6)
	NO ₂	16	0.59 (\pm 0.17)	4.6 (\pm 0.7 ppb)	42% (\pm 5%)	3.9 (\pm 16)	1.2 (\pm 0.5)	0.1 (\pm 0.3)	-0.1 (\pm 0.2)
	O ₃	19	0.81 (\pm 0.06)	5.1 (\pm 0.6 ppb)	24% (\pm 2%)	9.4 (\pm 14)	0.92 (\pm 0.2)	0.1 (\pm 0.2)	-0.2 (\pm 0.2)
	CO ₂	19	0.49 (\pm 0.13)	19 (\pm 3 ppm)	4% (\pm 1%)	390 (\pm 72)	0.1 (\pm 0.1)	-0.8 (\pm 0.7)	0.1 (\pm 1.0)
Type	Species	# RAMPs	Avg. Pearson r (\pm SD)	Avg. MAE (\pm SD)	Avg. CvMAE (\pm SD)	Median m_{try}	$m_{try} = 2$	$m_{try} = 4$	$m_{try} = 7$
RF	CO	19	0.99 (\pm 0.00)	7.9 (\pm 1.5 ppb)	3% (\pm 0.5%)	7	11%	21%	68%
	NO ₂	16	0.99 (\pm 0.01)	0.5 (\pm 0.1 ppb)	5% (\pm 1%)	4	21%	74%	5%
	O ₃	19	0.99 (\pm 0.00)	0.7 (\pm 0.1 ppb)	3% (\pm 0.4%)	4	0%	84%	16%
	CO ₂	19	0.99 (\pm 0.00)	1.7 (\pm 0.3 ppm)	0.4% (\pm 0.1%)	2	74%	21%	5%

LAB: Laboratory calibration (Eq. 1), MLR: multiple linear regression (Eq. 2), RF: random forest model.

For the LAB and MLR models, the fit coefficients are provided.

For the RF models, the median mtry value across the 16-19 RAMPs and the breakdown of the mtry tuning results (m_{try} which minimized RMSE) across the 16-19 RAMPs results are provided.

Table 3: Comparison to other published studies.

	Project	Location	Sensor Node	Type	N (days)	Time Res. (min)	AvgConc (ppb)	Slope	R²	MAE (ppb)	MBE (ppb)
CO	EuNetAir ¹	Aveiro, PT	AirSensorBox	EC	6	60	330	NR	0.76	90	0
	EuNetAir ¹	Aveiro, PT	NanoEnvi	EC	9	60	330	NR	0.53	100	100
	EuNetAir ¹	Aveiro, PT	Cambridge CAM11	EC	14	60	330	NR	0.87	180	-200
	EuNetAir ¹	Aveiro, PT	AQMesh	EC	15	60	330	NR	0.86	50	0
	CAIRSENSE ²	Decatur, GA, US	AQMesh	EC	110-111	60	330	NR	0.77-0.87	NR	NR
	CAIRSENSE ²	Decatur, GA, US	Air Quality Egg	MOS	115-196	60	310	NR	<0.25	NR	NR
	Castell et al. ³	Kirkeveien, NO	AQMesh	EC	72	15	NR	0.88*	0.36	150	-150
	Spinelle et al. ⁴	Ispra, IT	Figaro, e2V	EC, MOS	85	60	230	1.01-1.38	0.29-0.37	NR	NR
	Cross et al. ⁵	Boston, MA, US	ARISense	EC	75	5	--	0.94	0.88	24.8	-10.4
This Study		Pittsburgh, PA, US	RAMP	EC	41 [10-108]	15	270 (±30)	0.86 (±0.09)	0.91 (±0.05)	38 (±6.5)	0.1 (±0.2)
NO ₂	EuNetAir ¹	Aveiro, PT	Cambridge CAM11	EC	14	60	16	NR	0.84	5.61	-2.3
	EuNetAir ¹	Aveiro, PT	AirSensorBox	EC	7	60	16	NR	0.06	20.2	17.7
	EuNetAir ¹	Aveiro, PT	NanoEnvi	EC	7	60	16	NR	0.57	14.9	13.1
	EuNetAir ¹	Aveiro, PT	ECN_Box_10	EC	11	60	16	NR	0.89	4.95	-1
	EuNetAir ¹	Aveiro, PT	AQMesh	EC	6	60	16	NR	0.89	1.46	0
	EuNetAir ¹	Aveiro, PT	ISAG	MOS	13	60	16	NR	0.02	16.2	349.5
	CAIRSENSE ²	Decatur, GA, US	Cairclip	EC	194-285	60	11	0.96	<0.25-0.57	NR	NR
	CAIRSENSE ²	Decatur, GA, US	AQMesh	EC	110-111	60	10	NR	<0.25	NR	NR
	CAIRSENSE ²	Decatur, GA, US	Air Quality Egg	MOS	115-196	60	11	NR	<0.25	NR	NR
	Duvall et al. ⁶	Houston, TX, US	Cairclip	EC	24	60	5.5	0.25	0.01	NR	NR
	Duvall et al. ⁶	Denver, CO, US	Cairclip	EC	30	60	5.1	0.04	<0.01	NR	NR
	Castell et al. ³	Kirkeveien, NO	AQMesh	EC	72	15	NR	0.2-0.38*	0.24	26.2	13.3
	Esposito et al. ⁷	Cambridge, UK	SnaQ	EC	28	1	NR	NR	0.83	1.27	NR
	Spinelle et al. ⁸	Ispra, IT	uSense, Citytech	EC	86	60	9	0.64-0.79	0.55-0.59	NR	NR
	Cross et al. ⁵	Boston, MA, US	ARISense	EC	89	5	NR	0.81	0.69	3.45	1.20
This Study		Pittsburgh, PA, US	RAMP	EC	24 [2-56]	15	12 (±1.4)	0.64 (±0.11)	0.67 (±0.12)	3.48 (±0.36)	-0.4 (±1.13)
O ₃	EuNetAir ¹	Aviero, PT	AirSensorBox	EC	6	60	17	NR	0.13	22.12	19.2
	EuNetAir ¹	Aviero, PT	NanoEnvi	MOS	9	60	17	NR	0.77	7.66	6.5
	EuNetAir ¹	Aviero, PT	Cambridge CAM11	EC	11	60	17	NR	0.14	21.5	15.7
	EuNetAir ¹	Aviero, PT	AQMesh	EC	6	60	17	NR	0.7	2.4	0
	EuNetAir ¹	Aviero, PT	ISAG	MOS	13	60	17	NR	0.12	360.12	356.1
	CAIRSENSE ²	Decatur, GA, US	Aeroqual SM50	GSS	168-281	60	18	0.81-0.96	0.82-0.94	NR	NR
	CAIRSENSE ²	Decatur, GA, US	Cairclip	EC	194-285	60	17	0.68-0.85	0.68-0.88	NR	NR
	CAIRSENSE ²	Decatur, GA, US	AQMesh	EC	110-111	60	15	NR	<0.25	NR	NR
	Duvall et al. ⁶	Houston, TX, US	Cairclip	EC	24	60	32	0.93	0.80	NR	NR
	Duvall et al. ⁶	Denver, CO, US	Cairclip	EC	30	60	46	1.19	0.77	NR	NR
	Castell et al. ³	Kirkevein, NO	AQMesh	EC	72	15	NR	0.11-0.26*	0.29	19.9	6.8
	Esposito et al. ⁷	Cambridge, UK	SnaQ	EC	28	1	NR	NR	0.69	7.45	--
	Spinelle et al. ⁸	Ispra, IT	uSense, Citytech	EC	82-84	60	30	1.02-1.12	0.86-0.91	NR	NR
	Cross et al. ⁵	Boston, MA, US	ARISense	EC	87	5	NR	0.47	0.39	7.34	0.78

This Study	Pittsburgh, PA, US	RAMP	EC	38 [11-103]	15	22 (± 1.4)	0.82 (± 0.05)	0.86 (± 0.02)	3.36 (± 0.41)	-0.14 (± 0.46)
¹ (Borrego et al., 2016), ² (Jiao et al., 2016), ³ (Castell et al., 2017), ⁴ (Spinelle et al., 2017), ⁵ (Cross et al., 2017), ⁶ (Duvall et al., 2016), ⁷ (Esposito et al., 2016), ⁸ (Spinelle et al., 2015)										
EC=electrochemical, MOS=metal oxide sensor, GSS=gas sensitive semiconductor. NR= not reported in manuscript.										
For RAMP data, bracketed data is range (for N days) or standard deviation (all other metrics) across all the RAMP units.										
*values for slopes only provided for a subset of 2 of 24 sensors										