The next generation of low-cost personal air quality sensors for quantitative exposure monitoring

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Abstract

Advances in embedded systems and low-cost gas sensors are enabling a new wave of low cost air quality monitoring tools. Our team has been engaged in the development of low-cost wearable air quality monitors (M-Pods) using the Arduino platform. The M-Pods use commercially available metal oxide semiconductor (MO$_x$) sensors to measure CO, O$_3$, NO$_2$, and total VOCs, and NDIR sensors to measure CO$_2$. MO$_x$ sensors are low in cost and show high sensitivity near ambient levels; however they display non-linear output signals and have cross sensitivity effects. Thus, a quantification system was developed to convert the MO$_x$ sensor signals into concentrations.

Two deployments were conducted at a regulatory monitoring station in Denver, Colorado. M-Pod concentrations were determined using laboratory calibration techniques and co-location calibrations, in which we place the M-Pods near regulatory monitors to then derive calibration function coefficients using the regulatory monitors as the standard. The form of the calibration function was derived based on laboratory experiments. We discuss various techniques used to estimate measurement uncertainties. A separate user study was also conducted to assess personal exposure and M-Pod reliability. In this study, 10 M-Pods were calibrated via co-location multiple times over 4 weeks and sensor drift was analyzed with the result being a calibration function that included drift.

We found that co-location calibrations perform better than laboratory calibrations. Lab calibrations suffer from bias and difficulty in covering the necessary parameter space. During co-location calibrations, median standard errors ranged between 4.0–6.1 ppb for O$_3$, 6.4–8.4 ppb for NO$_2$, 0.28–0.44 ppm for CO, and 16.8 ppm for CO$_2$. Median signal to noise (S/N) ratios for the M-Pod sensors were higher for M-Pods than the regulatory instruments: for NO$_2$, 3.6 compared to 23.4; for O$_3$, 1.4 compared to 1.6; for CO, 1.1 compared to 10.0; and for CO$_2$, 42.2 compared to 300–500.
The user study provided trends and location-specific information on pollutants, and affected change in user behavior. The study demonstrated the utility of the M-Pod as a tool to assess personal exposure.

1 Introduction

1.1 Background and motivation

Health effects such as asthma, cardio-pulmonary morbidity, cancer, and all-cause mortality are directly related to personal exposure of air pollutants (EPA ISA Health Criteria, 2010, 2013a, b). To comply with the US Clean Air Act, state monitoring agencies make ongoing measurements in centralized locations that are intended to be representative of the conditions normally experienced by the majority of the population. Because these measurements require sophisticated, costly, and power-intensive equipment, they can only be taken at a limited number of sites. Depending on the pollutant, individual, and location, this limitation can lead to misleading personal exposure assessments (HEI, 2010). Low-cost, portable, and autonomous sensors have the potential to take equivalent measurements while more effectively capturing spatial variability and personal exposure. Thus, we set out to survey such sensors, analyze their performance, and understand the feasibility of using them. We describe the M-Pod hardware and quantification system, and personal exposure results.

1.2 Low-cost portable air pollution measurement techniques

Quantitative measurements of pollutant concentrations generally require techniques to be sensitive at ambient concentrations and unique to that particular compound (i.e. free from interference from other pollutants). Regulatory agencies use EPA-approved methods and equipment to meet these criteria, and some examples are given below. Rather than provide an exhaustive report of all available measurement techniques, we
provide brief descriptions of the various techniques (along with their measurements, costs, and potential).

### 1.2.1 Carbon monoxide

Federal Reference Method (FRM) measurements of CO are made using infrared absorption instruments, which use ~200 W power, cost $15,000–20,000, and require frequent calibrations and quality control checks (EPA Quality Assurance Handbook Vol. II, 2013). By comparison, metal oxide semiconductor (MO\textsubscript{x}) sensors cost $5–15 and require less than 1 W of power. One example of this kind of device is the SGX 5525 sensor used for CO measurements that uses approximately ~80 mW power. MO\textsubscript{x} sensors have fast responses, low detection limits, and require simple measurement circuitry. However, they can have high cross-sensitivities to other reducing gases, and can be poisoned by certain gases or high doses of target gases. Daily or weekly calibrations are recommended.

The typical reducing gas MO\textsubscript{x} sensor uses a heated tin-oxide n-type semi-conductor surface, on which oxygen can react with reducing gases, thus freeing electrons in the semiconductor. This lowers the electrical resistance proportional to the concentration of the reducing gas (Moseley, 1997). These sensors suffer from cross-sensitivities to temperature, humidity, and other pollutants. Korotcenkov (2007) provides a comprehensive review of MO\textsubscript{x} materials and their characteristics for gas sensing, while Fine et al. (2010) and Bourgeois et al. (2003) review the use of MO\textsubscript{x} sensors and arrays in environmental monitoring.

Electrochemical sensors are relatively low in cost, ~$50–100, and have been used in multiple studies that required low power sensors for measuring CO (Milton and Steed, 2007; Mead et al., 2013). They exhibit high sensitivity, low detection limit (sub-ppm for some models), fast response, low cross-sensitivity, and consume power in the hundreds of µW range. However, they have more complicated and expensive measurement circuitry, are susceptible to poisoning, have a shorter life span (generally 1–3 years), are more expensive than MO\textsubscript{x}, and are generally larger in size than MO\textsubscript{x}.
1.2.2 Ozone

FRM measurements of O$_3$ are made using the principle of chemiluminescence (EPA ISA, 2013a). Chemiluminescence instruments typically cost $10 000–20 000 and use approximately 1 kW. A Federal Equivalence Method uses UV absorption to measure an O$_3$ concentration. Such instruments have prices in the low $1000$s.

MO$_x$ O$_3$ sensors have been commercialized and can cost anywhere from $5 to $100, with power consumption as low as 90 mW. A tungsten oxide semiconductor sensor board has been commercialized by Aeroqual, and costs $\sim$250. Power consumption is 2–6 W, and this material is reported to have less cross-sensitivity and drift than other MO$_x$ materials (Williams et al., 2009). Electrochemical sensors are also available with reported detection limits less than 5 ppb.

1.2.3 Nitrogen oxides (NO$_x$)

FRM measurements of NO$_x$ are made using the chemiluminescence reaction of O$_3$ with NO along with the catalytic reduction of NO$_2$ to NO (EPA ISA, 2013b). These instruments typically cost $10 000–20 000 and consume approximately 1 kW power. NO$_2$ can also be measured with electrochemical sensors ($80–210$) and MO$_x$ sensors ($4–54$).

1.2.4 Carbon dioxide (CO$_2$)

CO$_2$ is the primary anthropogenic greenhouse gas, as well as a proxy for assessing ventilation conditions in indoor environments. Elevated concentrations have also been found to affect decision-making and exam performance (Satish et al., 2012). Portable non-dispersive infrared (NDIR) carbon dioxide sensors are precise, easy to calibrate, easy to integrate into a mobile sensing system (Yasuda et al., 2012), and are available in the $40$ range. The sensors operate by emitting a pulse of infrared radiation across a chamber. A detector at the other end of the chamber measures light intensity.
Absorption of light by CO$_2$ accounts for the difference between expected and measured intensity. Interference can occur due to absorption by water vapor and other gasses and drift can occur due to changes in the light source (Zakaria, 2010). Electrochemical sensors are also available to measure CO$_2$. They are inexpensive and have low power requirements, but generally have slower response times, shorter lifespans, and are more susceptible to poisoning and drift, than NDIR type sensors.

1.3 Instruments for personal air quality monitoring

Personal exposure has been characterized extensively using filter samplers, particle counters, and sorbent tubes. These methods can provide simple, accurate, and comprehensive speciation results, but the time series information is lost since each filter or adsorbent tube typically samples for durations of a day or more. Relatively recent sampling techniques allow for higher time-resolution personal measurement of pollutants.

Electrochemical sensors have been used to monitor CO in many works, including Kaur et al. (2007), Mead et al. (2013), Honicky et al. (2008), and Milton et al. (2006). Shum et al. (2011) developed a wearable CO, CO$_2$, and O$_2$ monitor. O$_3$ and NO$_x$ have both been monitored by Mead et al. and Honicky et al., using electrochemical and MO$_x$ sensors. Williams et al. (2009) developed and deployed a portable tungsten oxide-based ozone sensor and NO$_2$ sensor. Hasenfratz (2012) also monitored O$_3$ in a train-mounted instrument study using metal oxide semiconductor sensors. Hasenfratz's work tested collaborative calibration performance, in which sensor nodes were periodically co-located to check and improve calibrations.

Tsow et al. (2011) developed wearable monitors to measure benzene, toluene, ethylbenzene and xylene at ppb levels. The measurement is based on a MEMS tuning fork design that provides good selectivity and low detection limits, but the device is not yet commercially available. Electronic nose systems for sensing VOCs are commercially available, often designed to detect specific gas mixtures from processes. Such systems use a variety of sensing techniques, including those mentioned above, as well as polymer coated sensors, mass spectrometry, ion mobility spectrometry, and gas...
Advancements in technology and increasing concern about air quality in many regions have produced a wave of low-cost personal exposure instruments. Reliable results are needed for users of these low-cost monitors before they take action to reduce their exposure. We describe our novel quantification system that includes co-location calibration, modeling of sensor responses with environmental parameters, and uncertainty estimation for these measurements. We demonstrate the quantification system by presenting results from a user study where six users wore monitors for 10–20 days.

2 Methods

2.1 MAQS – Mobile Air Quality Sensing System

The requirements for our mobile sensing system included wearability, low-cost, multi-pollutant (sensing of as many NAAQS criteria pollutants at typical ambient concentrations as possible), wireless communication, and enough battery life to wear for an entire day. The result of our development effort is the M-Pod, shown in Fig. 1.

The Mobile Air Quality Sensing (MAQS) system is designed to collect, analyze, and share air quality data (Jiang et al., 2011). An Android mobile phone application, MAQS3 (Mobile Air Quality Sensing v.3, 2014) pairs with the M-Pod via Bluetooth, and the M-Pod data is transmitted to the phone periodically. This data is then sent to a server for analysis. A web-based analysis and GIS visualization platform can access new data from the server. Wi-Fi fingerprints can also be used to identify an M-Pod’s indoor locations (Jiang et al., 2012).

The M-Pod has four metal oxide sensors, measuring CO, total VOCs, NO₂, and O₃ (SGX Corporation models MiCS-5525, MiCS-5121WP, MiCS-2710, and MiCS-2611). Socket-mount MOₓ sensors were used because of the difficulty replacing the surface-mount sensors and possible poisoning of the surface-mount sensors due soldering
(hot-air reflow). An NDIR sensor (ELT, S100) measures CO$_2$ and a fan (Copal F16EA-03LLC) provides steady flow through the device. Data is logged using an Arduino-based platform. The M-Pod also has a light sensor, and a relative humidity and temperature sensor (Sensirion, SHT21).

2.2 Calibration system

MO$_x$ sensors represent the lowest cost sensing solution but hold significant quantification challenges. MO$_x$ sensor responses are non-linear with respect to gas concentration, and are affected by ambient temperature and, to a lesser degree, by humidity (Sohn et al., 2008; Barsan and Weimar, 2001; Delpha et al., 1999). Baseline drift and changes to sensitivity over time are also common. These are typically due to aging and irreversible changes to the sensing surface (Romain and Nicolas, 2009). As such, using MO$_x$ sensors quantitatively requires that a model be developed which not only characterizes the relationship between sensor resistance and gas concentration, but also includes the impacts of these other parameters. Below we describe our calibration system and strategies for overcoming these challenges.

Our calibration system uses automated mass flow controllers (MFCs, Coastal Instruments FC-2902V) and solenoidal valves to inject specific mixtures of gas standards into a Teflon-coated aluminum chamber equipped with, temperature and relative humidity control. Custom LabVIEW software and Labjack data acquisition devices are used for instrument control and data logging. The CO and NO$_2$ sensors were calibrated for changes in temperature and humidity. The CO$_2$ sensors were only calibrated for temperature, as they show a small non-linear response to temperature. Humidity effects have been reported for NDIR sensors, but are not a significant issue in this case. Temperature is controlled using a heat lamp and by performing calibrations inside a refrigerated chamber. Routing a portion of the airflow through deionized water controls relative humidity, using a 3-way valve. Conditions reach steady state in 90 s or less.

Calibrations are performed after sensors have operated continuously for at least a week, to ensure adequate sensor warm-up and stabilization time. Each calibration
run consists of different gas concentrations, temperature and humidity set points. Sensors are held at each state for periods of 15 min to allow them to reach steady state. The last 30 s of each 15 min period are averaged, and these points are used for the calibration. Initially, the sensors were calibrated by mounting them on large arrays, but we found that the sensor response is highly dependent on the position in the array and air flow conditions. The convective cooling of the sensors is thus an important variable. To ensure that calibration temperature and flow conditions about each sensor are the same as during operating conditions, they are calibrated in their individual M-Pods.

2.3 Development of quantification models

To simplify the inter-comparison of MOx sensors (which are often highly heterogeneous from sensor to sensor), it is common practice to normalize a sensor’s resistance by a reference resistance, $R_0$. The reference resistance is the sensor’s unique response to a given environment, for example, clean air at 25°C, standard atmospheric pressure, and 20% relative humidity. As such, a sensor quantification model relates $R_s/R_0$ to concentration, temperature and humidity. Previous works have used machine-learning techniques to determine concentration values (Kamionka et al., 2006) and identify pollution sources (Gardner and Bartlett, 1994). However, to our knowledge, a parametric regression based model has yet to be developed for these sensors. We believe this type of model is preferable for ease of implementation.

Two sensor models were chosen for the majority of the analysis conducted thus far: the MiCS-5121WP CO/VOC sensor and MiCS-5525 CO sensor (both manufactured by SGX Sensortech). The VOC sensor was chosen because of our strong initial interest in indoor air pollution. The MiCS-5525 was the logical next step because it has the same semiconductor sensor surface as the MiCS-5121, but with an activated charcoal pre-filter. The models to convert sensor signal to concentration were developed using both lab data and ambient co-location data. In lab experiments, the sensors were calibrated in the Teflon-coated chamber. The chamber and calibration system are described in the Supplement. The model derived from this data was then applied to each M-Pod.
CO sensor used in the co-location. Our results show that the CO, NO₂ and O₃ MOₓ sensors can detect ambient concentrations when frequently calibrated.

Figure 2 illustrates the MiCS-5525 CO sensor response to changing temperature at various concentrations of CO. The absolute humidity has a lesser effect on signal response and was held constant so as to minimize the degrees of freedom within the model response. From experimental observation, the sensor response appears to change linearly with respect to temperature for a given CO concentration between concentrations of 0–2.8 ppm. The slope and intercept of the linear temperature trends also appear to decrease with increasing CO concentration. Equation (1) was chosen as the best fit for the observed sensor response to CO concentration and temperature. A third term of the same form was added to the model to account for changes in absolute humidity (H).

\[
\frac{R_s}{R_0} = f(C)(T - 298) + g(C) + h(C)H
\]  

(1)

In this model, \(f(C)\) describes the change in temperature slope with respect to pollutant concentration; \(g(C)\) describes the change in resistance in dry air at 298 K due to concentration; and \(h(C)\) describes the change in absolute humidity slope with respect to concentration. The terms \(f(C)\), \(g(C)\), and \(h(C)\) were chosen to be of the form \(p_1 \exp(Cp_2)\).

This model form performed well for all MOₓ sensors used, but is computationally challenging to work with because it is not algebraically invertible. Instead, we used a 2nd order Taylor approximation for this model. However, an even simpler model in temperature, absolute humidity, and concentration (Eq. 2) was found to perform similarly in many cases. The comparable performance of the models is likely due to the low variation in CO concentration observed throughout the field experiments. Though we did not perform the same lab calibration tests with the NO₂ and O₃ sensors, we found that in co-location calibrations, Eqs. (2) and (3) also fit the data comparably to
the model in Eq. (1).

\[
\frac{R_s}{R_o} = p_1 + p_2 C + p_3 T + p_4 H
\]  

(2)

In cases with longer time series and multiple calibrations, a time term, \(p_5 t\), was added to correct for temporal drift.

\[
\frac{R_s}{R_o} = p_1 + p_2 C + p_3 T + p_4 H + p_5 t
\]  

(3)

Equation (3) was used throughout the results unless otherwise noted.

We determined concentration uncertainty by propagating the error in the calibration model through the inverted calibration function (NIST Engineering Statistics Handbook 2.3.6.7.1). The calculation included co-variance terms, but did not include the propagated uncertainty of the temperature, humidity, nor voltage measurements, as those are expected to be insignificant relative to the other sources of error. The calculated uncertainty does not directly account for sources of error such as convection heat loss or cross sensitivities that may be seen in field measurements but not during calibration. However, co-location calibration should account for some cross-sensitivity effects since there is simultaneous exposure to various measured pollutants.

To explore the validity of this uncertainty propagation, we employed duplicate M-Pods during a user study. For this user study data, when there were duplicate M-Pod measurements but no reference monitors, we used two additional methods to explore uncertainty, the average relative percent difference (ARPD), and the pooled pairwise standard deviation of the differences (SD\(_{\text{diff}}\)) (Table 4). These formulas are defined as follows:

\[
SD_{\text{diff}} = \sqrt{\frac{1}{2n} \sum_{i=1}^{n} (C_{i}^{\text{primary}} - C_{i}^{\text{duplicate}})^2}
\]  

(4)
\[ \text{ARPD} = \frac{2}{n} \sum_{i=1}^{n} \left| \frac{C_{i\text{primary}} - C_{i\text{duplicate}}}{C_{i\text{primary}} + C_{i\text{duplicate}}} \right| \times 100\% \] (5)

This approach, outlined in Dutton et al. (2009), provides an additional assessment of measurement uncertainty, and can be compared to the uncertainties calculated using propagation of error to understand if the propagation has captured most real sources of error. To calculate the ARPD, negative data were removed. In the future, zero replacement, or detection limit replacement for data with negative values will be considered. The ARPD was then multiplied by the average pooled concentration measurements to get units of concentration that could be directly compared with the uncertainty estimates derived through propagation.

### 2.4 Validation and user study

From 3–12 December 2012, and later from 17–22 January 2013, nine M-Pods were co-located with reference instruments at a Colorado Department of Public Health and Environment (CDPHE) air monitoring station in downtown Denver. Total system performance was assessed by comparing laboratory-generated calibrations with calibrations based on “real-world” ambient data, referred to as co-location calibrations. The 2nd co-location was performed with a fresh set of sensors and yielded slightly better results (Supplement). Reference instruments for calibration and validation were provided by CDPHE and the National Center for Atmospheric Research (NCAR). CO was measured using a Thermo Electron 48c monitor, \( \text{CO}_2 \) and \( \text{H}_2\text{O} \) were measured with a LI-COR LI-6262, \( \text{NO}_2 \) was measured using a Teledyne 200E, and \( \text{O}_3 \) was measured with a Teledyne 400E. The \( \text{CO}_2 \) instrument was calibrated before the deployment, while the others were span and zero checked daily as per CDPHE protocol. The M-Pods were positioned 8 feet from the sampling inlets. They operated continuously in a ventilated shelter on the roof of the facility.
In the user-study portion of the validation, nine M-Pods were carried for over two weeks, with three users each carrying two M-Pods. The objective of the user study was to demonstrate the use of the M-Pods for assessing personal exposure. Specifically, we were interested in M-Pod inter-comparisons and how they drift over time during personal usage. The M-Pods were calibrated before and after the deployment using co-location calibrations. They were worn on the user’s upper arm or attached to backpacks or bags, and were placed as close as possible to the breathing area when users were sitting or sleeping.

Measurement values were given as minute medians of the 1/10 Hz raw data. The raw data was filtered beforehand for electronic noise. Sensor-specific thresholds of two standard deviations on the differences between sequential values were used to identify and remove noise spikes. An upper bound threshold on sequential differences provided another layer of filtering for the noisiest data. To ensure that sensors were warmed up, 10 min of data were removed after power-on. Additional noise filtering was applied for the co-location tests due to a bad USB power supply. This data was filtered for noise by applying the Grubbs test for outliers to the differences between all the M-Pods and a “reference” M-Pod that displayed less electronic noise.

3 Results

3.1 Lab vs. co-location calibration results

A summary of the results from the 3–12 December co-location and lab-calibrated data are presented in Tables 1 and 2. Table 1 shows summary statistics for the first co-location calibration, while Table 2 shows the performance for the different calibration methods and models.
3.1.1 MO\textsubscript{x} sensor results

The MiCS-5525 CO sensor was found to have substantially higher error using lab-calibrations vs. co-location calibrations. As shown in Table 1, the median standard error for co-location calibration was 0.45 ppm (range 0.38–0.52 ppm), while the median lab calibration standard error was 3.56 ppm (range 2.85–5.33 ppm). Adding a linear time correction, as in Eq. (3), was found to improve the fit in most MO\textsubscript{x} sensor data sets. In this case, it improved the fit of the co-location calibrations slightly, giving a median standard error of 0.44 ppm (range 0.38–0.51 ppm). The median standard error for the exponential-based model from Eq. (1) was 0.39 ppm (range 0.34–1.78 ppm), but it actually provided a worse fit in some cases. The linear form of the equation, Eq. (2), is a good approximation of the exponential form shown in Eq. (1), likely because of the small parameter space spanned by the observed data. We have included residual plots (Fig. 3) to demonstrate model performance. Note the absence of a trend in these residual plots.

The relationship between co-location calibrated sensor readings and reference data showed a slight negative bias at the higher end of observed concentration levels, but this appears to be driven by a small number of data points.

Inter-sensor variability is of interest if these sensors are to be widely deployed. Low variability could allow us to calibrate fewer sensors and apply those calibrations to other sensors in a large network. Inter-sensor variability for CO was generally low, with median correlation coefficients among the M-Pods 0.70 (range 0.62–0.78). The median signal to noise (S/N) ratio, defined as the median observed value over the standard error, was 1.13 (range 1.00–1.26). This compares with the reference instrument S/N of 10.0, calculated using the median standard error from multiple days of zero and span-check data as noise. The S/N ratio provides straightforward comparison of instruments, and shows us how often the measurements are above the noise.

For the O\textsubscript{3} and NO\textsubscript{2} sensors, the model in Eq. (2) fit well, with evenly distributed residuals and median standard errors of 6.1 ppb (range 4.2–15.4 ppb), and 8.4 ppb.
(range 6.9–9.5 ppb), respectively. As shown in Table 2, the linear model from Eq. (2) was found to fit the data nearly as well for NO₂ as the non-linear model from Eq. (1), and is much less computationally intensive to use. An NO₂ example time series using the linear model from Eq. (2) is shown in Fig. 3. The non-linear model was not able to fit the O₃ data with any success, also shown in Table 2. The reason for this was not determined despite repeated testing. Lab calibrations were not performed for O₃ and NO₂. Median inter-sensor correlation for O₃ was 0.83 (range 0.46–0.99), and 0.96 (range 0.94–0.99) for NO₂. The median NO₂ S/N was 3.6 (range 3.3–4.4), compared with the median reference instrument S/N of 23.4. For O₃, the median S/N ratio for the M-Pods was 1.4 (range 0.5–2.0), while the reference instrument had S/N of 1.6.

Figure 3 NO₂ data from M-Pod 23 from the December co-location.

3.1.2 NDIR CO₂ sensor results

CO₂ values quantified with lab calibrations showed bias in some M-Pods (see Table 2), while others showed a high degree of accuracy. With co-location calibration, we also found a previously unseen temperature effect, described by

\[ v = p_1 + p_2 C + p_3 (T - p_4)^2, \]  

where \( v \) is the raw sensor signal. This model fit better than a linear model in concentration, and an example is shown in Fig. 4.

As shown in Table 2, using linear models in concentration only, the median standard error for M-Pod CO₂ measurements using the lab calibrations was 68.4 ppm (range 15.2–138.7 ppm), and was 16.8 ppm (range 10.1–48.8 ppm) using the co-location calibration. Median standard error was 10.9 ppm (range 7.2–24.8 ppm) using the co-location calibration model from Eq. (6), and adding a linear time correction to this model further improved the fit, dropping the median standard error to 6.9 ppm. This drift term was statistically significant. The improvement in fit with the more complex model may be due to a temperature effect of the semiconductor infrared sensor, or an unidentified confounding variable. Adding humidity as a variable was not found to improve the
fit significantly. Using the co-location calibration approach, the median correlation between CO\textsubscript{2} sensors in different M-Pods was 0.88 (range 0.58–0.98). The median signal to noise ratio was 42.2 (range 18.5–63.4), as compared with a reported 300–500 from the reference instrument used (LICOR, 1996).

3.2 User-study results

Based on initial lab and co-location calibration results, calibrations for the user-study were performed only with co-location calibrations. Co-location calibrations were carried out before and after the 3-week measurement period. Calibration fits were comparable to the prior co-location calibrations for CO (median standard error of 0.3 ppm), NO\textsubscript{2} (median standard error of 8.8 ppb), and O\textsubscript{3} (median standard error of 9.7 ppb). For CO\textsubscript{2}, the median standard error was high (36.9 ppm), likely because we were unable to co-locate a reference monitor with the M-Pods at these times. Instead, a calibration curve was generated using data from a nearby ambient monitor operated by NCAR and a lab calibration. Correlations among paired M-Pods during the user study ranged between 0.88–0.90 for NO\textsubscript{2}, 0.48–0.76 for CO, 0.33–0.92 for CO\textsubscript{2}, and 0.04–0.35 for O\textsubscript{3}. The range of correlations for CO\textsubscript{2} was due to power supply issues, which will be discussed later. We expect reliable CO\textsubscript{2} sensor performance to be easily achievable in future work. Despite the low standard error from O\textsubscript{3} sensor calibrations, we found low correlations among the paired M-Pods during the user study, which is also likely due to a power supply issue.

Personal exposure measurements using the M-Pods were generally low for all users, but clear trends and effects of behavior were seen. Table 3 shows summary statistics. Median CO and CO\textsubscript{2} exposure were 0.58 ppm and 949.0 ppm, respectively. Median O\textsubscript{3} exposure was 14.0 ppb, and Fig. 5 has an example probability density plot and time of day trend plot for a user’s O\textsubscript{3} exposure.

Measurement uncertainty calculated with the method of propagation and the duplicate M-Pod statistics, ARPD and SD\textsubscript{diff}, defined in Eqs. (4) and (5), are compared in Table 4. The results show moderate agreement among the methods for most pollutants.
For CO and O₃, the propagated uncertainty is lower than the SD_{diff} and ARPD, roughly 50–75% of it, confirming that there are sources of error that are not accounted for in the uncertainty propagation. For NO₂, the propagated measurement uncertainty seems to capture most of the uncertainty observed in the pairs. The RMSE values from the sensor calibrations were found to account for the majority of the propagated error. Figure 6 compares the CO measurements from M-Pods 23 and 25, along with their 95% confidence interval, the ARPD, and SD_{diff}.

Drift was seen to affect the measurement results, as described in detail in the Supplement. We compensated for drift using multiple co-location calibrations with linear time corrections (Haugen et al., 2000), and observed improved calibration fits. Average daily drift during the user study is shown in Table 3. For CO, all M-Pods experienced drift under −0.05 ppm day⁻¹, apart from M-Pod 15, which showed behavior we cannot explain. O₃ sensors experienced between −2.6 ppb day⁻¹ and 2.0 ppb day⁻¹ drift. CO₂ drift ranged from −4.2 ppm day⁻¹ to 3.1 ppm day⁻¹, excluding the bad results from M-Pod 9. NO₂ generally showed a slight positive drift over time, with a range of −1.56 to 0.51 ppb day⁻¹.

4 Discussion

The M-Pods performed well given the relatively low ambient concentration environments encountered in the region. For CO, NO₂, and CO₂, the reference instruments exhibited S/N ratios 8–10 times higher than the M-Pod measurements. For O₃, the reference monitor S/N ratio was only slightly higher than the median M-Pod value. The user study demonstrated the utility of the M-Pod in real time continuous monitoring applications, in terms of user behavior changes. For example, one user experienced elevated nighttime CO₂ and NO₂. CO₂ levels often increase at night in rooms with little ventilation, and can lead to restlessness. NO₂ levels increase due to combustion, and in this case the use of the furnace on these cold nights appears to have increased
personal NO₂ exposure substantially. With this kind of readily available concentration data, users can then adjust their behavior to reduce night time exposures.

4.1 Lab calibration

Lab calibrations had higher measurement error than co-location calibrations, likely because the field data covered a wider range of environmental parameter space than the lab calibration. The poor performance of lab calibrations may also be due to the difference between zero-grade air cylinders and ambient air. Conducting co-location calibrations in the region of interest helps to account for confounding factors and meteorological variability.

CO₂ lab-calibration results showed accurate results in some cases, while in other M-Pods we found significant bias. Some CO₂ sensors consistently showed worse performance than others. Strangely, the worse performing ones were usually in good agreement with each other. We have no explanation for this behavior, apart from a potential power supply issue.

4.2 Co-location calibration

The time and resources required for lab calibration, and the difficulty reconciling the lab and ambient results, led us to rely more on co-location calibration. Co-location calibration performed well during two wintertime tests. However, during later co-location calibrations in warmer periods with rapidly changing weather, we found more interference from either reducing gases or humidity swings than we had previously seen. This effect, coupled with generally lower CO levels in the warmer months due to better atmospheric mixing and improved motor vehicle combustion (Neff et al., 1997), resulted in flatter and noisier calibration curves than previously seen. To minimize this effect, some portions of the calibration data set were removed for April and May user study calibrations. O₃ and NO₂ had slightly worse calibration fits than during the winter calibrations, likely also due to larger swings in atmospheric humidity.
4.3 User study discussion

The user study provided valuable insight and promoted changes in behavior of some participants. Users reported being more aware of “stuffiness”, and acted based on their data by opening windows to increase air exchange rates in their homes. They also noted increases in CO₂, CO, and NO₂ during driving, leading them to experiment with the best way to reduce exposures. S/N ratios during the user study were generally higher than during the co-locations. This suggests that during personal exposure measurement, when concentration peaks are often higher than background measurements, the M-Pod is able to detect those peaks above the noise. Analysis based on the propagated uncertainty, ARPD, and SD$_\text{diff}$ suggests that propagated uncertainty is capturing most sources of error, but it does require more testing to further validate uncertainty estimation approaches.

Temporal trends generally showed higher concentrations in the morning and evening for CO and NO₂, coinciding with commute-time increases in ambient concentrations. O₃ trends followed expected outdoor patterns, peaking in the afternoon in most cases. CO₂ trends generally showed nighttime increases. The distributions had a distinct “fresh air” mode, near ambient concentrations, and an indoor mode with a heavy tail. For other pollutants, exposure probability density function distributions varied substantially, likely due to exposure variability based on individual user behavior. One user’s overnight use of a wood-fired stove is evident with increased nighttime CO exposure and a wider distribution than that of other users. Most other CO distributions were quite narrow, though slightly right-skewed. The NO₂ distributions varied between right-skewed distributions for four M-Pods, and bi-modal distributions for the other five M-Pods. These modes appear to be driven by daily differences in NO₂ exposure rather than indoor/outdoor differences.

Some users did not carry and charge their M-Pods as fastidiously as others. This may have contributed to episodes of strange sensor behavior due to low power operation. Generally, the air quality encountered was very good, and this may have diminished
the perceived value of the M-Pod. Unfortunately, one M-Pod from each of two of the M-Pod pairs appears to have had persistent power issues, which would explain the low number of samples relative to their respective pairs, and worse sensor behavior during calibration.

A study shortcoming was the inability to wear reference monitors in addition to the M-Pods. This was illustrated with curiously high NO$_2$ concentrations, indicating that there may be problems we are not accounting for. Despite such potential issues, concentration changes near busy roadways are often apparent, showing the sensitivity and fast response of the sensors, valuable for source and trend identification.

5 Conclusions

Co-location and collaborative calibration will be a valuable tool in the next generation of air quality monitoring. With help from monitoring agencies and citizen scientists, detailed ground-level pollutant maps will one day help track sources, reduce the population’s exposure, and improve our knowledge of emissions as well as fate for each species. In this work, we have demonstrated a quantification system that can provide personal exposure measurements and uncertainties for CO$_2$, O$_3$, NO$_2$, and CO. This type of quantification approach provides access to air quality monitoring to a wider audience of scientists and citizens. It requires moderate investment to develop a calibration infrastructure, whether in a laboratory or near a monitoring station, but we believe it is worthwhile in applications like health and exposure, source identification, and leak detection.

Supplementary material related to this article is available online at http://www.atmos-meas-tech-discuss.net/7/2425/2014/amtd-7-2425-2014-supplement.pdf.
Acknowledgements. This work was funded by CNS awards O910995 & 0910816 from the National Science Foundation. Thank you to Bradley Rink and the CDPHE. Thank you to the Hannigan Lab: Tiffany Duhl, Lamar Blackwell, Evan Coffey, Joanna Gordon, Nicholas Clements, and Kyle Karber.

References


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Table 1. Co-location calibration summary statistics for December co-location using the linear model from Eq. (3).

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<td>N mean std med 5th % 95 % drift S/N</td>
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<td>14 157 0.59 0.69 0.47 −0.18 1.87 0.02 1.22</td>
<td>12 919 11.8 18.4 9.7 −9.2 41.1 −0.6 0.7</td>
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<tr>
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<td>M-Pod 21</td>
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<td>Median</td>
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<td>13 366.5 12.2 12.9 8.7 −3.6 35.8 −0.3 1.4</td>
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<tr>
<th></th>
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<th>CO₂ (ppm)</th>
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<td>N mean std med 5th % 95 % drift S/N</td>
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<td>M-Pod 1</td>
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<tr>
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<td></td>
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<td>14 311 467.3 50.4 457.1 410.9 573.4 −1.5 18.5</td>
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Table 2. Standard errors for the various calibration models tested with the December co-location data set. Equation (1), the exponential model, was not able to fit O₃ satisfactorily for some unknown reason.

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<tr>
<th>Calibration type Model</th>
<th>( \text{CO}_2 ) (ppm)</th>
<th>( \text{CO} ) (ppm)</th>
<th>( \text{O}_3 ) (ppb)</th>
<th>( \text{NO}_2 ) (ppb)</th>
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<td></td>
<td>Co-loc Eq. (6)</td>
<td>Co-loc Eq. (6) w/time</td>
<td>Co-loc Linear</td>
<td>Lab Co-loc Eq. (1)</td>
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<tr>
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<td>37 558.7</td>
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<td>82 843.3</td>
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<td>0.39</td>
<td>23 064.1</td>
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Equation (1), the exponential model, was not able to fit \( \text{O}_3 \) satisfactorily for some unknown reason.
### Table 3. Personal exposure summary statistics using the model in Eq. (3). Rows in italic/bold indicate paired M-Pods.

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<th>CO (ppm)</th>
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<th>CO₂ (ppm)</th>
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<td></td>
<td>N</td>
<td>mean</td>
<td>std</td>
<td>med</td>
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<tr>
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<td>0.62</td>
<td>0.37</td>
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<tr>
<td>M-Pod 6</td>
<td>10595</td>
<td>0.47</td>
<td>1.30</td>
<td>0.48</td>
</tr>
<tr>
<td>M-Pod 9</td>
<td>7214</td>
<td>1.26</td>
<td>0.68</td>
<td>1.27</td>
</tr>
<tr>
<td>M-Pod 15</td>
<td>8710</td>
<td>8.83</td>
<td>1.27</td>
<td>8.90</td>
</tr>
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<td>10308</td>
<td>1.54</td>
<td>0.60</td>
<td>1.54</td>
</tr>
<tr>
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<td>11816</td>
<td>−0.15</td>
<td>0.53</td>
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<tr>
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<tr>
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**NO₂ (ppb)**

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<th>95%</th>
<th>R²</th>
<th>Drift</th>
<th>ppm/day</th>
<th>S/N</th>
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<th>mean</th>
<th>std</th>
<th>med</th>
<th>5%</th>
<th>95%</th>
<th>R²</th>
<th>Drift</th>
<th>ppm/day</th>
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<td>68.7</td>
<td>54.2</td>
<td>83.3</td>
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<td>544.5</td>
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<td>M-Pod 25</td>
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Table 4. Average pooled uncertainty calculations for user study duplicate measurements.

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<th>O₃ (ppb)</th>
<th>NO₂ (ppb)</th>
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<td>SD_{diff}</td>
<td>ARPD</td>
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<td>0.58</td>
<td>0.63 (66.9 %)</td>
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<tr>
<td>M-Pod 15, 16</td>
<td>0.92</td>
<td>3.8</td>
<td>4.57 (133 %)</td>
</tr>
<tr>
<td>M-Pod 23, 25</td>
<td>0.28</td>
<td>0.36</td>
<td>0.36 (55.5 %)</td>
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</table>
Fig. 1. The M-Pod and the accompanying MAQS3 phone application.
Fig. 2. MiCS-5525 CO sensor response to various CO concentrations while held at different ambient temperatures.
Fig. 3. NO$_2$ data from M-Pod 23 from the December colocation.
Fig. 4. Calibration surface (using Eq. 4) for a CO$_2$ co-location calibration performed from 3–12 December using M-Pod 1.
Fig. 5. Probability density function and time of day trend for O₃ from M-Pod 23.
Fig. 6. Personal CO measurement comparison between M-Pods 23 and 25, including 95% confidence intervals in light and dark gray, respectively.