

RESEARCH ARTICLE

Tower measurement network of in-situ CO₂, CH₄, and CO in support of the Indianapolis FLUX (INFLUX) Experiment

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A twelve-station tower-based observation network measuring CO₂, CH₄, and CO was deployed in and around the Indianapolis, IN metropolitan area as part of the Indianapolis Flux Experiment (INFLUX). Measurements began in 2010 and the full network was deployed by 2013. Observations were made at heights ranging from 39 to 136 m above ground level using existing communication towers. Several towers in the network had multiple measurement levels. Cavity ring-down spectrometers (CRDS) were used at all twelve sites and at least one calibrated reference tank was sampled daily at each site. Real time data communications were performed via cellular modems and data were transmitted daily for processing and quality assurance checks. Data quality control procedures were utilized to ensure compatibility within the INFLUX tower network and with global standards. For example, field target/calibration tanks were used to detect long-term instrument drift and instrument failure. Network-wide round robin tests were performed every 1–2 years to detect possible target tank drift and ensure network-wide comparability between measurements. NOAA flask packages were deployed at six of the INFLUX towers to provide a flask to in-situ direct comparison of the atmospheric samples. Results from these activities demonstrate that the compatibility of the CO₂, CH₄, and CO INFLUX in-situ tower-based measurements are less than or equal to 0.18 ppm CO₂, 1.0 ppb for CH₄, and 6 ppb for CO.

Keywords: Network; CRDS; INFLUX

1. Introduction

Continuous measurements of atmospheric carbon dioxide (CO₂) mole fraction have been traditionally accomplished using infrared-based instruments (e.g., Keeling et al., 1960; Peterson et al., 1986; Bakwin et al., 1995; Stephens et al., 2011; Andrews et al., 2014). In 2007, cavity ring-down spectroscopy (CRDS; Picarro, Inc.) and off-axis integrated cavity output spectroscopy (Los Gatos Research, Inc.) instruments became commercially available. These instruments have been field tested (e.g., Winderlich et al., 2010; Chen et al., 2010; Richardson et al., 2012; Rella et al., 2013; Yver Kwok et al., 2015) and demonstrated to have

improved stability, and consequently require reduced calibration frequency, compared to infrared instruments.

Emissions from urban areas contribute about 70% of the total global CO₂ fluxes (IEA, 2008) and thus understanding the spatial and temporal variability of those emissions is crucial toward informing effective greenhouse gas mitigation strategies. Towards that end, several cities have recently been instrumented with multiple continuous CO₂ measurements including Paris (Breon et al., 2014), Boston (McKain et al., 2014), Los Angeles (Kort et al., 2013), and Indianapolis (Miles et al., 2017).

Methane (CH₄) is a potent greenhouse gas (GHG), with a global warming potential over 100 years that is approximately 25 times greater than CO₂ (Myhre et al., 2013). The global CH₄ mole fraction has increased by approximately 150% since preindustrial times, and is responsible for nearly 20% of anthropogenic radiative forcing of the climate system (Myhre et al., 2013). Additionally, carbon monoxide (CO) is emitted during incomplete combustion and is correlated with the fossil fuel component of CO₂ emissions (Meijer et al., 1996; Turnbull et al., 2015a).

The urban signals of greenhouse gas mole fractions in this flat Midwest US city have maximum observed dormant season enhancements of 2.9 ppm CO₂, 21 ppb

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CH₄ and 29 ppb CO (Miles et al., 2017). Thus, to achieve compatibility within 10% of the mean dormant-season urban signal in Indianapolis during the winter, the compatibility requirements are 0.29 ppm CO₂, 2.1 ppb CH₄ and 2.9 ppb CO (Miles et al., 2017).

This paper describes continuous in-situ measurements at twelve tower locations in and around the city of Indianapolis, IN, as part of the Indianapolis Flux (INFLUX) project (Davis et al., 2017). We describe the instrument calibration procedures used prior to deployment in the field, the air sampling strategy used at each site, and the use of the on-site calibrated reference tank in data post-processing. Long-term instrument stability and total data availability are examined. Finally, flask to in-situ comparisons are presented, along with results from two round robin style tests in which three or four NOAA calibrated tanks were sampled at all twelve sites over the period of a week.

2. Methods

2.1 Network installation

Installation of the first two INFLUX sites occurred in 2010 (Sites 01 and 02, **Figure 1**) to measure CO₂, CH₄, and CO. Additional site installations began in March 2012, and by July 2013, all twelve sites were operational. The new sites consisted of three CO₂ and CH₄ instruments (Picarro Inc. model CFADS, G2000 series), three CO₂ and CO instruments (Picarro Inc. model CKADS, G2000 series), and four early version CO₂ only systems (Picarro Inc. model CADS, G1000 series; Richardson et al., 2012). In November 2014, the CADS systems were replaced with CFADS (G2000 series; CO₂ and CH₄) systems. Site 12 was used for a short period and then decommissioned and the instrumentation was moved to Site 13.

Each CRDS instrument was calibrated in the laboratory prior to deployment (Richardson et al., 2012) using three to five standards traceable to the WMO X2007 for CO₂, WMO X2004 for CH₄, and WMO X2014 for CO scales. These NOAA tertiary standards ranged between 360 and 450 ppm CO₂, 1790 and 2350 ppb CH₄, and 140 and 310 ppb CO.

2.2 In-situ sampling procedure

The in-situ sampling method closely follows the procedures described in Richardson et al. (2012). Shown in **Figure 2A** is a schematic of the air sampling system for a site with multiple measurement levels. Sites 01, 02, 03, and 09 had multiple sampling levels for the CRDS systems to examine the vertical gradients and used a pump to continuously purge the sampling lines at ~1 lpm. The sites with only one sample line did not have a purge pump. Collocated at the top sampling level of each tower were two ¼ in (0.64 cm) OD Synflex 1300 (Eaton Corp.) tubes for the CRDS instruments and one or two ¾ in (0.95 cm) OD Synflex 1300 tubes for the flask sampling packages; all other levels had a single ¼ in (0.64 cm) OD Synflex tube for the CRDS system. The top end of each tube was equipped with a rain shield to prevent liquid water from entering the sampling line. Separate lines were used for the CRDS and flask sampling lines because of the large flow rates required for the one-hour integrated samples collected into the flask packages (Turnbull et al., 2012) and to ensure independence of the samples. Switching between sample and calibration gas(es) (and between different measurement heights where applicable) was accomplished using 3-way solenoid valves (part number 091-0094-900, Parker Hannifin Corp.) and 1/8 in (0.32 cm) OD stainless steel tubing,

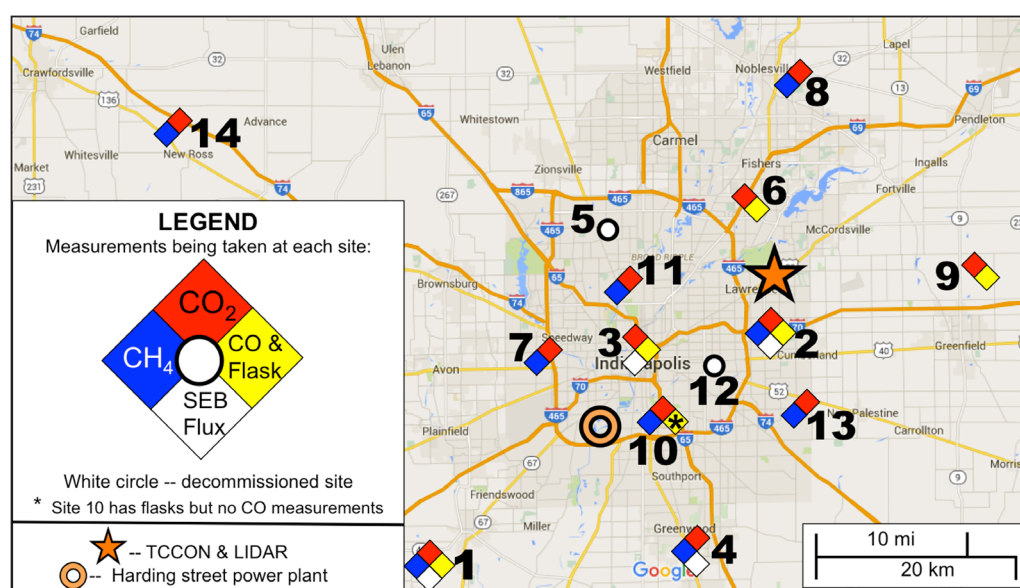


Figure 1: Locations of INFLUX tower in-situ sites as of June 2017. Note Site 12 was decommissioned in 2013 and the instrumentation was moved to Site 13. Site 05 was decommissioned in 2016 and Site 14 was installed in April 2017 to serve as an additional background site. Legend indicates sites that measured CO₂, CH₄, and/or CO. Surface Energy Balance (SEB) sites were located at Sites 01, 02, 03, and 04. The star indicates the location of a Total Carbon Column Observing Network (TCCON) site. The Harding Street power plant location is shown by the open circle. DOI: <https://doi.org/10.1525/elementa.140.f1>

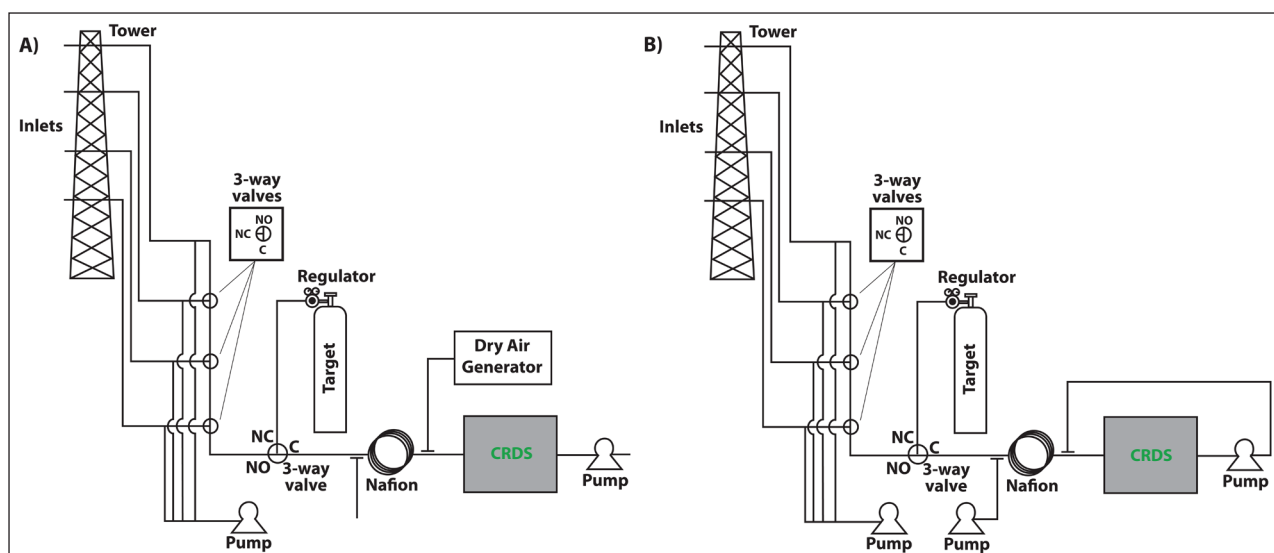


Figure 2: A) Schematic of air sampling system used at sites with multiple measurement levels and dry air as the Nafion purge gas. Air was drawn down from the inlet on the tower, through the Nafion drier, into the CRDS instrument for analysis. A water guard, which consisted of two nested aluminum funnels with wire mesh over the opening of the inner funnel to prevent bugs from entering, was located at the top of each sample line. All sample lines were continuously purged at ~1 lpm using a pump, and solenoid valves were used to switch in different sample levels. A dry air generator provided the purge gas for the Nafion drier with a flow rate at least twice the volume of the air sample. Target tank gas was introduced upstream of the drier to detect leaks. All equipment was housed either in an existing building at the tower site or in a shed installed for the project. **B) Schematic of air sampling system used at sites with multiple measurement levels and the reflux method for sample drying.** Air is drawn down from the inlet on the tower, through the Nafion drier, into the CRDS instrument for analysis, and then used as the purge gas in the Nafion drier (i.e., re-flux method). As per the manufacturer recommendations, a vacuum was pulled through the purge gas path and was maintained at at least $\frac{1}{2}$ atmosphere (380 Torr) to maintain the desired 2:1 purge-to-sample volumetric flow ratio. A water guard, which consisted of two nested aluminum funnels with wire mesh over the opening of the inner funnel to prevent bugs from entering, was located at the top of each sample line. All sample lines were continuously purged at ~1 lpm using a pump, and solenoid valves were used to switch in different sample levels. Target tank gas was introduced upstream of the drier to detect leaks. All equipment was housed either in an existing building at the tower site or in a shed installed for the project. DOI: <https://doi.org/10.1525/elementa.140.f2>

Air Liquide (formerly Scott Specialty Gas) regulators (part number 51-14 A-590) were used for sampling the field calibration tanks.

At sites measuring more than one level, the lower two or three levels were sampled for ten minutes each, and the top level was sampled for the remainder of the hour. Flask samples were always collected from the highest level at each tower.

The samples at all sites measuring CO were dried using Nafion dryers (PermaPure, part number MD-110-24S-2 at Site 02 and MD-070-96S-2 at the remainder of the sites), and dry air generators (Twin Tower Engineering, part number MW200) for counter flow. Gradually, as maintenance issues with the dry air generators arose, the reflux method was implemented using an additional pump (part number ME1, Vacuubrand, Inc.) on the outlet of the Nafion dryer as shown in **Figure 2B**.

As of late May 2013, the incoming sample air at all INFLUX sites was dried. Water vapor values in Indianapolis, measured at Site 10 prior to the installation of the drying system, showed that the water vapor mole fraction was over 2% during about one-third of the afternoon hours

(1700–2100 UTC) in the summer (June–August). The sample air stream was dried at all sites in order to avoid the annual water vapor calibrations necessary for water vapor values over 2% (Rella et al., 2013). However, co-located instruments at an INFLUX site for 3 months (May–July 2011), one with drying and without drying, indicated comparable results for CO₂ (0.01 ± 0.09 ppm, Rella et al., 2013).

This drying system resulted in water vapor levels below 0.2% for sites utilizing a 96 in (244 cm) Nafion drier. Only Site 02 used a 24 in (61 cm) Nafion which resulted in water vapor levels less than 0.6%, depending on ambient conditions. The field calibration tanks went through the Nafion drier and were humidified to about 0.15% and 0.4%, for the long and short Nafion driers, respectively, minimizing the humidity difference between the sample airstream and calibration gases, as in Andrews et al. (2014). Manufacturer-supplied correction factors were applied to adjust the CO₂ and CH₄ values for the effects of the remaining water vapor (Rella et al., 2013); no such correction was available for CO. Leaks in the Nafion driers could be detected via the target tanks (Section 2.3).

2.3 Target tanks

A comprehensive field calibration method to detect changes in instrument response would include sampling at least three known tanks to develop a field calibration curve, and one tank to be measured as an independent unknown. Yver Kwok et al. (2015) showed that, for CO₂, CH₄, and CO, the observed instrument drift was most likely due to drift in the calibration scale and not in the linearity of the instruments. Thus a linear calibration curve can be used. In addition, the time rate of change of the slope of the instrument calibration curve is very slow (see Section 3.1) and sampling one tank and applying a daily offset correction produces sufficient measurement compatibility for the INFLUX project. Thus, one (or two at Sites 01 and 02) NOAA-calibrated tank(s) were sampled at each of the twelve INFLUX sites every 23 hours for ten minutes as a data quality assurance measure and to correct for instrument drift. This tank is referred to here as a “target” tank. A sampling frequency of 23 hours was chosen such that the sampling time progressed throughout the day to capture any time-of-day measurement variability; none was detected. The first four minutes of the ten-minute sample were discarded to allow the instrument to equilibrate. Allan deviation results of Yver Kwok et al. (2015), as well as laboratory tests performed (not shown), showed that the Allan deviation at six minutes was less than 0.02 ppm for CO₂, less than 0.1 ppb for CH₄ and less than 1 ppb for CO. The difference between the measured tank value and the known tank value, the “target tank offset”, was used as an offset correction to the day’s data. The average target tank offset was used at Sites 01 and 02 where two tank measurements were available. The variability in the measured daily target tank value contained both instrument drift and instrument noise, and ideally only instrument drift would be part of the offset correction. Instead of applying the offset correction independently for each day’s data, a correction smoothed over several days, for example, could have been applied. However, this method added error when there were sudden shifts in instrument response. The standard deviation of the differences between these two techniques in hourly-calibrated values over the period January–March 2017 was 0.02 ppm CO₂, 0.1 ppb CH₄, and 0.9 ppb CO. Thus, applying the offset daily did add noise to the data, but it was negligible.

2.4 Round robin style testing

In November 2013 and September 2015 round robin style tests, hereafter referred to as simply “round robin tests”, were performed in which three (in 2013) and four (in 2015) NOAA-calibrated tanks were sampled at all sites over a one-week period. This procedure was designed to ensure the compatibility of the INFLUX tower network, both within the network and to the global scale, using an independent set of standards that was common to all sites. Also, since it was not feasible to return all CRDS instrument to the laboratory for recalibration, this testing accounted for long-term changes in the slope of the instrument calibration curve (see Section 3.1; Yver Kwok et al., 2015) that were not accounted for using the target tanks at each site.

For these tests, the round robin tanks were treated as unknowns, with the daily target tank offset applied as for atmospheric samples (as described in Section 2.3). Each round robin tank was sampled for ten minutes, passing through the plumbing as an atmospheric sample. After the conclusion of the round robin tests, the results were used to field-calibrate (apply a slope and offset correction) the instruments, and the new calibration was used from the time of the test until the next calibration. Note that this method can result in a shift in the target tank offset correction (Section 3.1) when the new calibration is applied, and this could be smoothed out by allowing the calibration to change linearly between calibrations. However, doing so would mean data collected after one round robin test, but before the next one, would not be available.

2.5 INFLUX flask sampling: comparison to in-situ

Flask samples were collected at six of the INFLUX tower sites as described by Turnbull et al. (2012). These samples were collected in the mid-afternoon (1300–1500 LST, 1900–2000 GMT) when atmospheric mixing was most vigorous and were collected only on days when the wind was from the western sector (180–360°) so that Site 01 represented background conditions (Turnbull et al., 2015a). Flasks were measured for a variety of species (Sweeney et al., 2015) including CO₂, CH₄, N₂O, SF₆, CO, H₂ (Conway et al., 2011), ¹⁴CO₂ (Turnbull et al., 2015a), a suite of halo- and hydrocarbons (Montzka et al., 1993) and stable isotopes of CO₂, CH₄ and/or CO (Vaughn et al., 2004). The flask samples allowed partitioning of CO₂ into fossil and biogenic components (Turnbull et al., 2015b) and potentially among more detailed sectors or processes using the multiple species measured.

The flask samples for INFLUX were collected as one-hour integrated averages, primarily to average out the short-term atmospheric variability that was difficult to interpret (Turnbull et al., 2012). A secondary benefit of the one-hour integrated samples, as opposed to grab samples, was that comparison with one-hour averages of in-situ CO₂, CH₄ and CO data was straightforward and the comparison was less susceptible to timing errors which would complicate grab flask comparisons with in-situ data. Turnbull et al. (2012) showed that while the weighting function of the one-hour integrated sample was not perfectly linear, it was close enough to linear that the flask results could be compared with the simple mean of the in-situ measurements over the same hour. For towers which sample multiple levels, we only averaged the in-situ measurements made at the same level as the flask samples, which were typically 40 minutes out of the hour, and note that this could introduce bias in the comparison. The flask systems were located at sites with G2000 series Picarro instruments that measured CO.

3. Results

3.1 Long-term instrument drift

Figures 3, 4 and 5 show the daily target tank offset (difference between the CRDS measured value for CO₂, CH₄, and CO and the target tank known value)

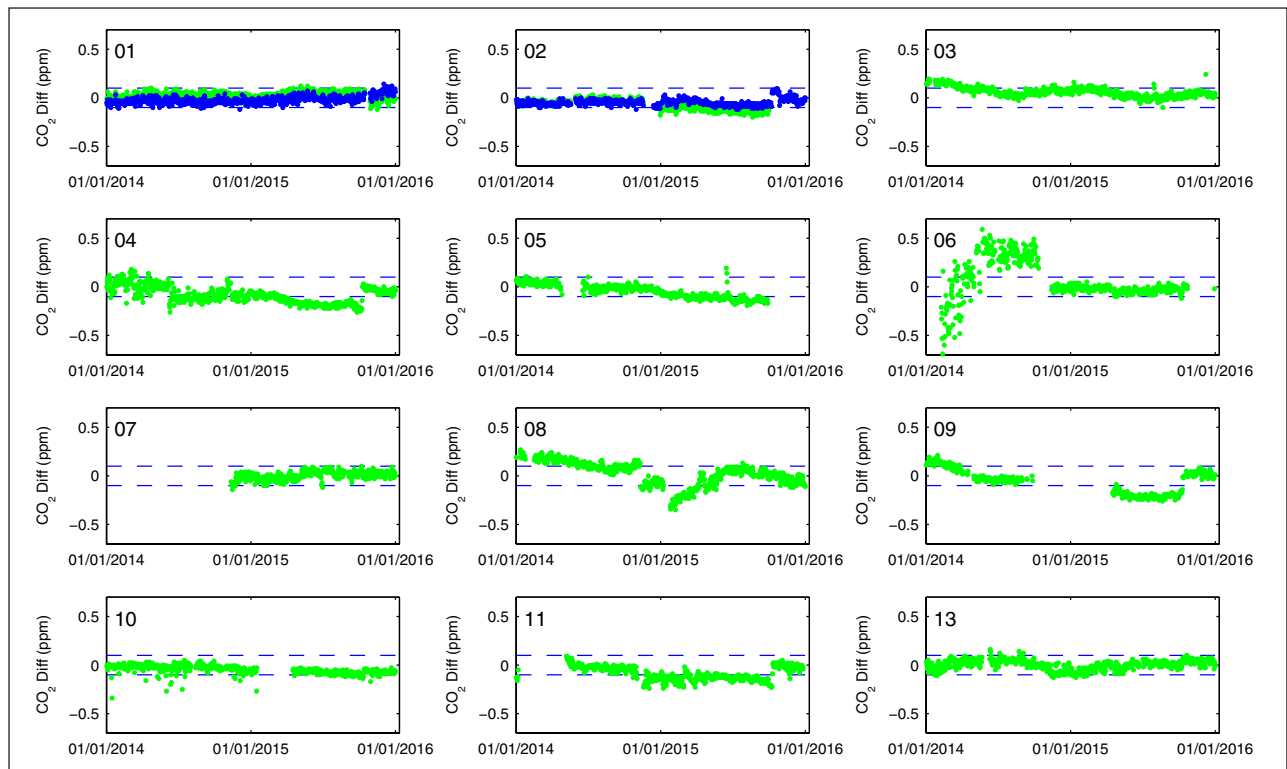


Figure 3: Daily offset correction for CO₂. The difference between the uncorrected measured CO₂ value of the daily target tank and the known tank value for each site, for 2014 and 2015. Site numbers are indicated on each graph. This difference was used as an offset correction to the atmospheric data. Dashed lines indicate ± 0.1 ppm CO₂. Sites 01 and 02 had an additional tank with known CO₂ as shown by the blue line. DOI: <https://doi.org/10.1525/elementa.140.f3>

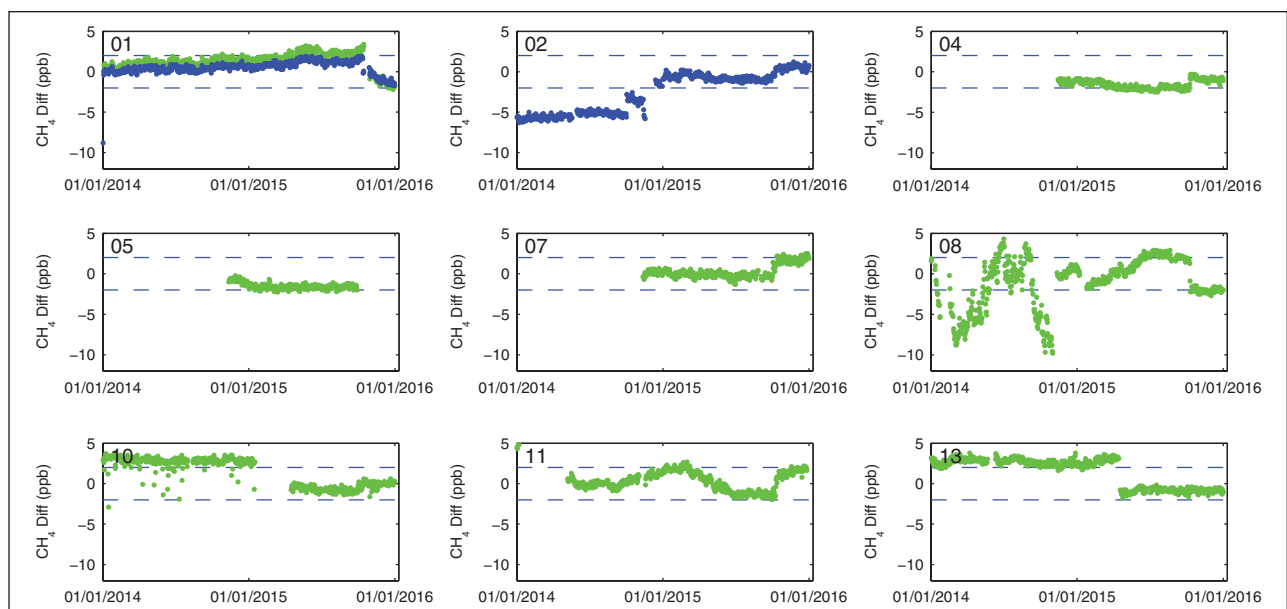


Figure 4: Daily offset correction for CH₄. Difference between the uncorrected measured CH₄ value of the daily target tank and the known tank value for each site, for 2014 and 2015. Site numbers are indicated on each graph. This difference was used as an offset correction to the atmospheric data. Dashed lines indicate ± 2 ppb CH₄. Site 01 had an additional tank with known CH₄ as shown by the blue line, the extra tank at Site 02 was only characterized for CO₂ so there is only one target tank for CH₄ at site 02. DOI: <https://doi.org/10.1525/elementa.140.f4>

at all sites for 2014 and 2015. Recall that these daily (23 hour) target tank measurements were used as a bias correction for the atmospheric data. While many

sites were stable over time with slowly varying target tank offsets over the two years, numerous exceptions are evident. Looking first at the CO₂ (**Figure 3**), the

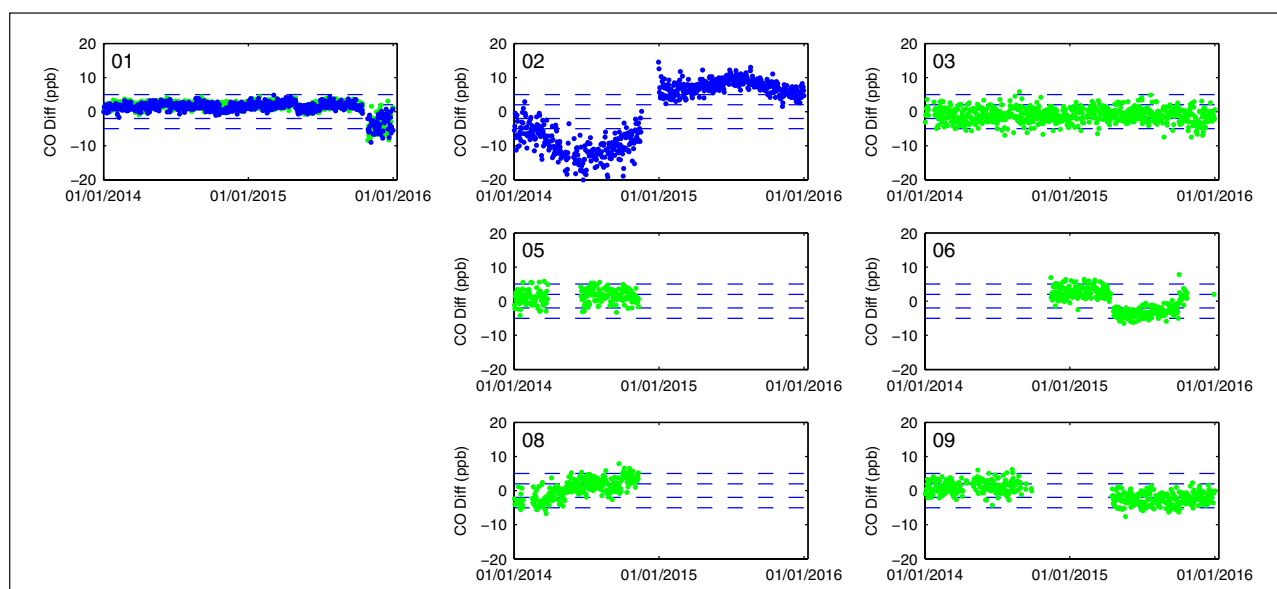


Figure 5: Daily offset correction for CO. Difference between the uncorrected measured CO value of the daily target tank and the known tank value for each site, for 2014 and 2015. Site numbers are indicated on each graph. This value was used as an offset correction to the atmospheric data. Dashed lines are ± 2 ppb and ± 5 ppb CO. Site 01 had an additional tank tank with known CH_4 as shown by the blue line, the extra tank at Site 02 was only characterized for CO_2 so there is only one target tank for CO at site 02. DOI: <https://doi.org/10.1525/elementa.140.f5>

instruments at Sites 04, 06, and 07 (Site 07 was not operational in 2014) were G1000 series instruments (CADS) that were eventually replaced with G2000 series systems (CFADS) in November 2014. The decreased noise in the CO_2 measurement is evident at Sites 04 and 06 and is consistent with the findings of Yver Kwok et al. (2015) which found improved performance in later model instruments. During 2014, site 10 showed target tank offsets in excess of 0.2 ppm and eventually this instrument failed and was returned to the manufacturer for repair. After installing the repaired instrument several months later, there was less noise in the target tank measurement for the remainder of the period. Several sites (02, 04, 09, and 11) showed a small discontinuity in the target tank offset near the end of 2015, which was the result of applying the calibration derived from the 2015 round robin testing. The abrupt change at Site 08 in November 2014 was attributable to an instrument replacement; the other discontinuity in early 2015 occurred for unknown reasons.

Methane target tank offsets (Figure 4) occurred for many of the same reasons as CO_2 . In particular, several sites (01, 04, 07, 08, and 11) showed a shift in target tank value in late 2015 when the round robin calibration was applied. For example, the daily bias correction at Site 04 prior to the round robin calibration is ~ 2 ppb, afterward it is ~ 1 ppb. As was the case for the CO_2 target tank offset, Site 10 was noisy until it was repaired. Site 08 showed large daily changes in target tank offset until the instrument was replaced in late 2014 and also showed an abrupt change in early 2015 as was observed in the CO_2 measurements. Although Site 02 did not show any significant changes in CO_2 target tank offsets, three distinct periods are evident in the CH_4 target tank offsets; each

was associated with a different instrument. Site 11 had no abrupt changes in CH_4 target tank offsets but did show a long-period sinusoidal change.

In Figure 4, drift in the methane calibration is apparent at Site 01 from early 2014 to late 2015. The offset increased in time and the offsets of the two tanks were correlated, thus the increasing offset cannot be attributed to tank drift (unless both tanks drifted identically). This drift in instrument calibration represented a real change in the instrument response and could be due to many different factors, e.g., laser aging. It is this change in the calibration curve, more specifically the slope of the calibration curve, that was corrected in the round robin tests using multiple field calibration tanks every ~ 2 years, and the offset was corrected daily using a single target tank (Section 2.3). This use of tanks provided the required measurement accuracy.

Figure 5 shows the CO target tank offset at seven measurements sites in the INFLUX network. There was seven instead of five CO measurement sites shown because the Site 05 instrument was moved to Site 06 in late 2014 and because Site 08 had a temporary instrument with CO while the primary instrument for that site was repaired. As is evident in the figure, the noise inherent in the CRDS (Series 2000) measurement of CO varies by instrument. For example, Site 01 had the least amount of day-to-day CO target tank offset of any site until the instrument failed and was replaced in late 2015. Site 02 showed a varying CO target tank offset until failing in 2014 and being replaced with a different instrument in early 2015. Site 06 had an operating system upgrade in early 2015 with a subsequent recalibration; a small shift in the target tank offset was evident without an appreciable change in the noise of the measurement.

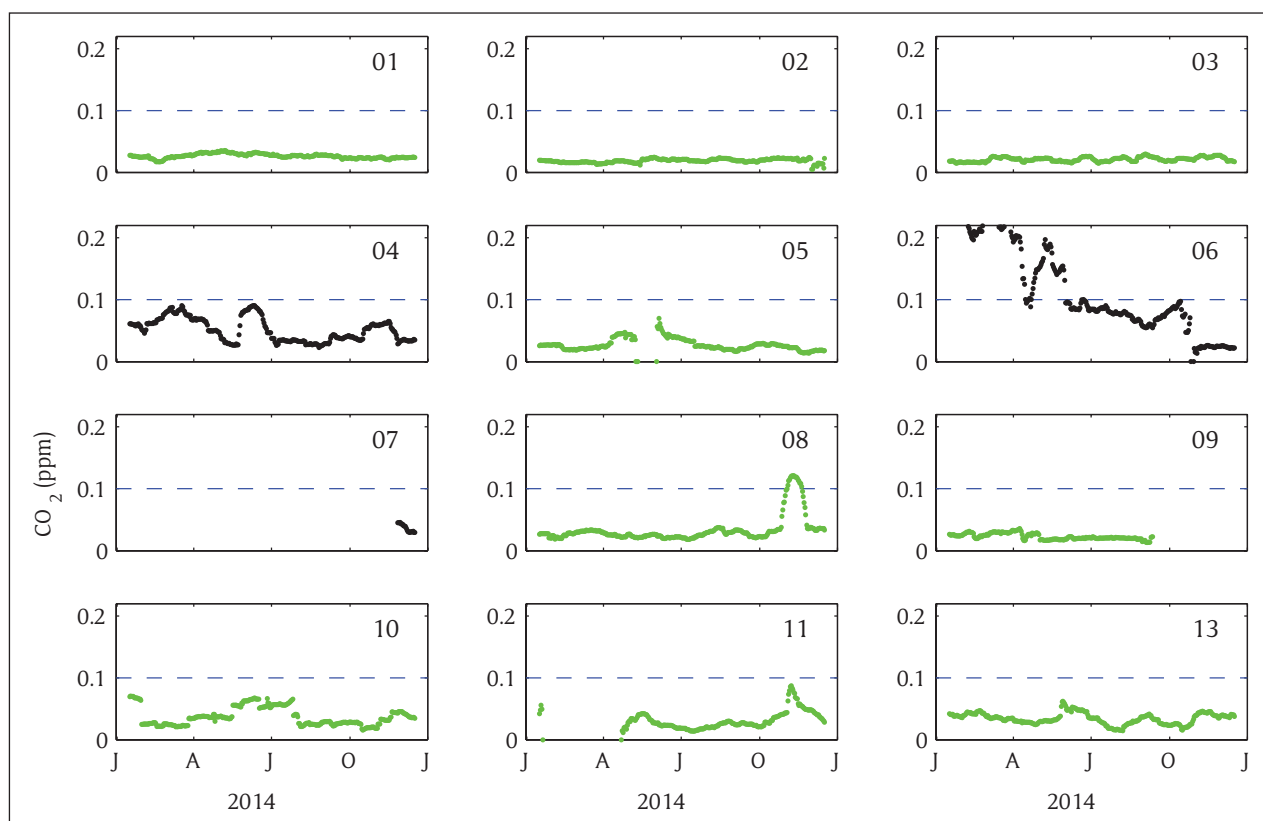


Figure 6: Instrument uncertainty for CO₂ for 2014 only. Instrument uncertainty for 2014, site numbers are indicated on each graph. Data shown in black are G1000 series instruments; green data indicate G2000 series instruments. DOI: <https://doi.org/10.1525/elementa.140.f6>

We emphasize that the data shown in **Figures 3, 4 and 5** are not estimates of the error in the reported atmospheric mole fractions. Instead, these data indicate the offset errors that would be present if no field calibrations were performed using the target tank at each site. Daily (23-hour) target tank sampling can be used to diagnose problems and document instrument performance. While less frequent sampling could have been employed without inducing significant differences in the final corrected values, at the flow rates of the G2000 series instruments (typically 250 cc/min), the target tanks (size 150 A) last 3–4 years. Applying a daily offset correction based on a single tank cannot detect tank drift or errors in the assigned values of the tank. For that reason, it is preferable to utilize two or more tanks. However, this was cost-prohibitive, and we instead relied on round robin testing to detect tank drift and/or tank assignment errors.

The target tank offsets were used as a measure of the instrument uncertainty by calculating a centered 30-day running standard deviation of the target tank offset. For example, **Figure 6** shows instrument uncertainty for CO₂ in 2014. Comparing this with the target tank offsets shown in **Figure 3**, one can see that instrument uncertainty is higher when daily target tank offsets are more variable (recall **Figure 3** shows data for 2014 and 2015). In addition, when there was an abrupt change in the target tank offset (e.g., **Figure 3**, Site 08 in late 2014), there was a corresponding large instrument uncertainty that was an artifact of the running mean. It should be

noted that events not associated with instrument problems (e.g., instrument swap) also caused a temporary increase in instrument uncertainty as quantified through this method. Sites 04, 06, and 07 had older CRDS instruments during much of 2014 and an increased instrument uncertainty (shown in black) is evident at Sites 04 and 06. Note the significant decrease in instrument uncertainty at Site 06 when a new instrument was installed at the end of 2014. The uncertainty attributable to variability in the instrument response was on average, about 0.03 ppm CO₂ for G2000 series instruments and generally less than 0.1 ppm CO₂ for G1000 series (except for during 2014 for Site 06). For CH₄, the instrument-based uncertainty was 0.3–0.6 ppb and for CO, 1–3 ppb (not shown). Andrews et al. (2014) described other components of the total uncertainty, including calibration scale uncertainty and atmospheric variability.

3.2 Round robin testing

Shown in **Figures 7 and 8** are results for the round robin tests, indicating the departure from assigned calibration values for each tank, for each species, and for each site. For the 2013 round robin, the deviations from known values at each site varied between –0.3 and 0.3 ppm CO₂. The mean CO₂ error (averaged over the three round robin tanks, **Figure 9A**), depended upon time since the last calibration, varying from –0.08 to 0.05 ppm CO₂ for calibrations performed less than one year prior, to –0.25 to 0.1 ppm CO₂ for calibrations performed greater than 2

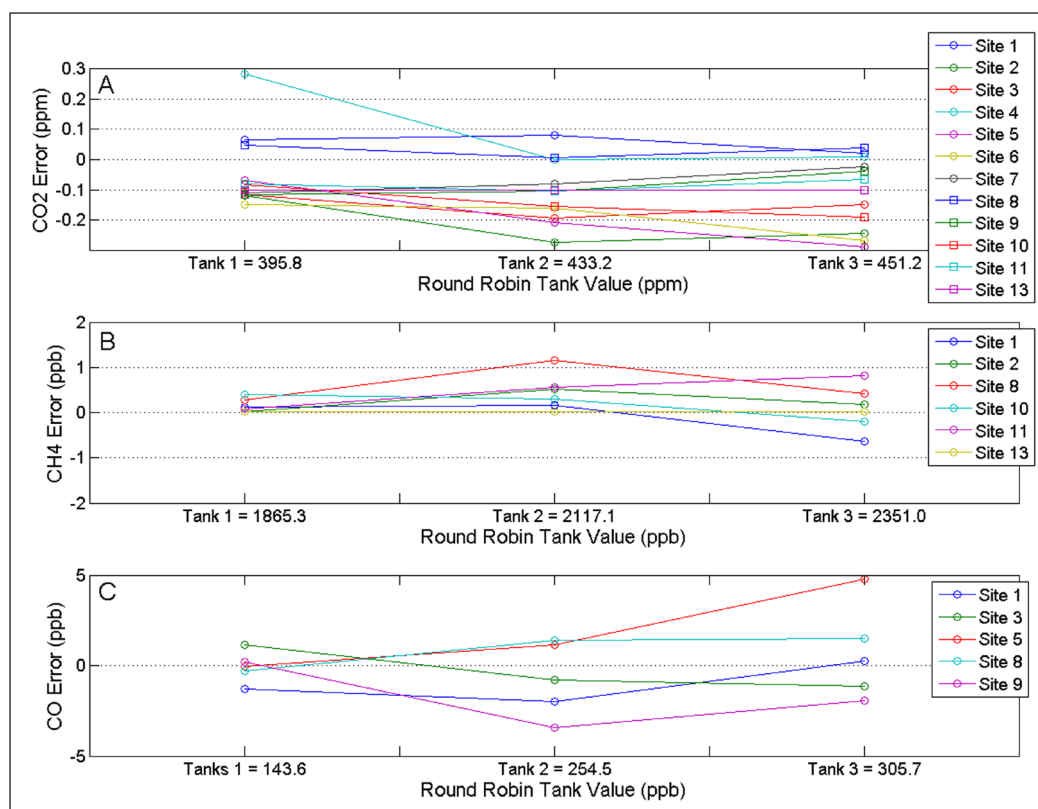


Figure 7: Round robin tank error (measured – known) for all sites, all species for 2013 test. Shown in **A)** is CO₂ (ppm), **B)** CH₄ (ppb), and **C)** CO (ppb) for the tests performed in November 2013. DOI: <https://doi.org/10.1525/elementa.140.f7>

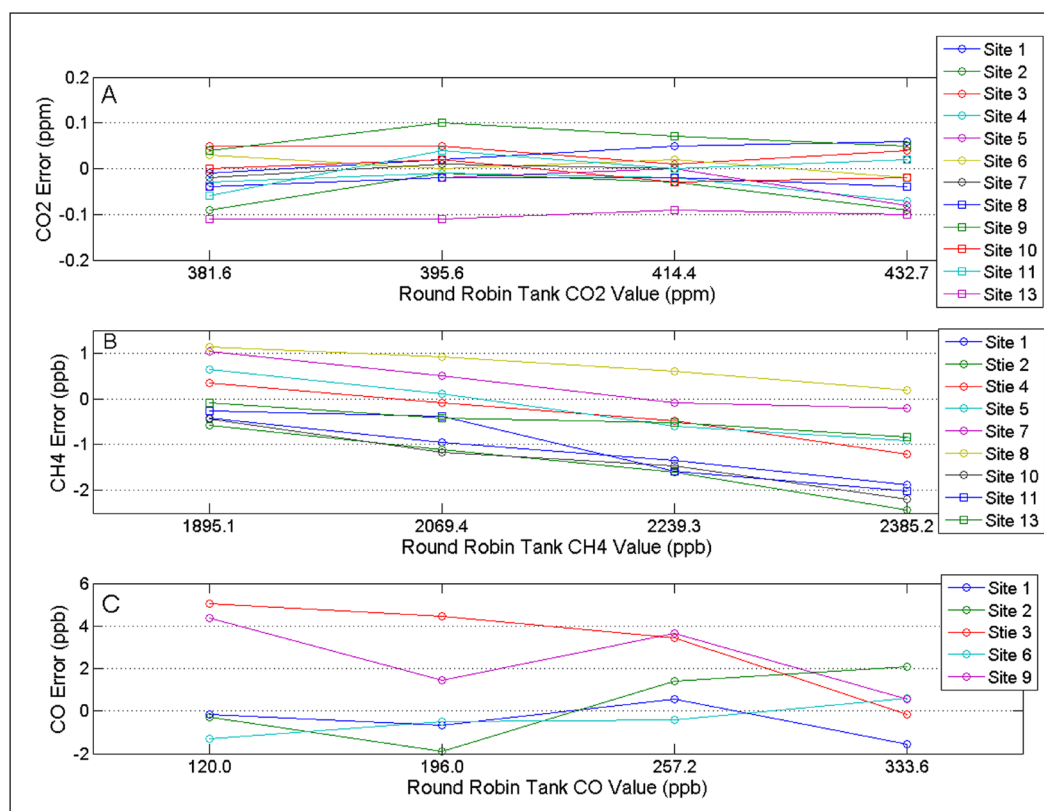


Figure 8: Round robin tank error (measured – known) for all sites, all species for 2015 test. Shown in **A)** is CO₂ (ppm), **B)** CH₄ (ppb), and **C)** CO (ppb) for the tests performed in September 2015. DOI: <https://doi.org/10.1525/elementa.140.f8>

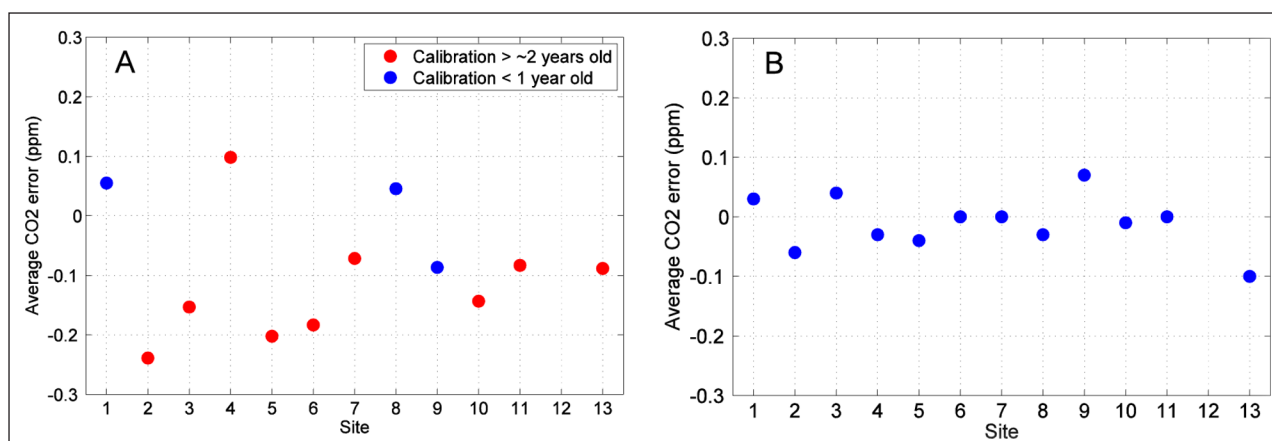


Figure 9: Average CO₂ Round robin tank error (measured – known) for all sites. Average round robin tank errors for CO₂. **A)** 2013 round robin CO₂ error at each site, averaged over the three round robin tanks. Sites calibrated less than one year prior to round robin are shown in blue, while those with the older calibrations (>2 years) are shown in red. (No calibrations occurred between 1 and 2 years prior to the round robin testing in 2013). **B)** 2015 round robin CO₂ error at each site averaged over the four round robin tanks. A slope and offset correction was applied to all instruments following the 2013 round robin testing using the results thereof. DOI: <https://doi.org/10.1525/elementa.140.f9>

years prior (in some cases, as long as 5–7 years prior). The network-averaged errors were -0.09 ± 0.11 ppm CO₂. The results of the 2013 round robin measurements were used to determine a slope and offset correction for each instrument and this was applied after the 2013 round robin was concluded. Thus, for the 2015 round robin, **Figure 9B**, all the instruments had been calibrated less than 2 years prior, and the results were significantly improved. The deviations from known values are all less than about 0.1 ppm CO₂ for the 2015 round robin test (**Figure 8A**), and the mean CO₂ error for each site vary between -0.10 and 0.7 ppm CO₂ (**Figure 9B**). For Site 13, all of the round robin tanks measured about 0.1 ppm CO₂ lower than the known values, indicating an incorrect value was assigned to the field calibration tank or a possible drift in the tank. The network-averaged error for the 2015 round robin were 0.00 ± 0.05 ppm CO₂.

For CH₄, the deviations from known values at each site varied between -0.6 and 1.0 ppb for the 2013 round robin (**Figure 7B**) and between -2.3 and 1.0 ppb in 2015 (**Figure 8B**). For CO, the deviations varied between -4 and 5 ppb in 2013 (**Figure 7C**) and -2 and 5 ppb in 2015 (**Figure 8C**). These deviations were within WMO recommendations for measurements in urban environments (± 2 ppb for CH₄, ± 5 ppb for CO; GAW Report No. 229, 2016). The network-averaged error was 0.2 ± 0.4 ppb CH₄ and 0 ± 2 ppb CO for the 2013 round robin and -1.0 ± 0.8 ppb CH₄ and 1 ± 2 ppb CO in 2015. Note the obvious, but small (~ 1 ppb difference over the tested range), slope error in the 2015 CH₄ measurements at all twelve sites. In July 2015 the NOAA CH₄ X2004 scale expanded from covering a range of 300 to 2600 ppb to cover a range from 300 to 5900 ppb and became the X2004 A scale. The round robin tanks used in the 2015 tests were on the X2004 A scale while the instruments were all on the X2004 scale as a result of the 2013 round robin tests.

The change in scale has a slight methane concentration dependence and this can be seen in **Figure 8B**. As stated in Section 2.4, the round robin results were used to field calibrate (slope and offset) the instruments, following the testing so this effect was removed.

3.3 Comparison with flask measurements

Since all flasks were measured on the same well-calibrated instrument at NOAA/ESRL, they allowed for an intercomparison with the in-situ tower measurements in order to evaluate them for possible errors. Unlike the round robin tank testing, the flask to in-situ comparison allowed comparison of the atmospheric samples of CO₂, CH₄, and CO, instead of sampling a known tank. The comparisons at the INFLUX towers yielded mean differences of 0.18 ± 0.55 ppm CO₂, 0.6 ± 5.0 ppb CH₄, and -6 ± 4 ppb CO for the period 2011–2016 (**Figure 10, Table 1**). The bias for CO₂ and CH₄ (in-situ – flask) were within one standard deviation of the differences. For CO, however, there was a clear bias towards lower values in the in-situ reported results. This bias may be attributable to inadequate drying of the in-situ samples or target tank drift (round robin results did not show any), but the exact reason is at this time unknown. Note that sample storage time (the amount of time between filling a flask and analyzing it) was examined and there was no correlation of the bias with storage time at any site. As discussed in Section 2.2, calibration gases (target tanks or round robin tanks) passed through the Nafion drier and were humidified to within approximately 0.05% H₂O (0.2% at Site 02) of the air sample. Manufacturer-supplied water vapor corrections for CO₂ and CH₄ were used to account for the remaining water vapor, but no correction was available for the CO measurements. Thus, the small amount of water vapor may have biased the CO measurements differently for each instrument because

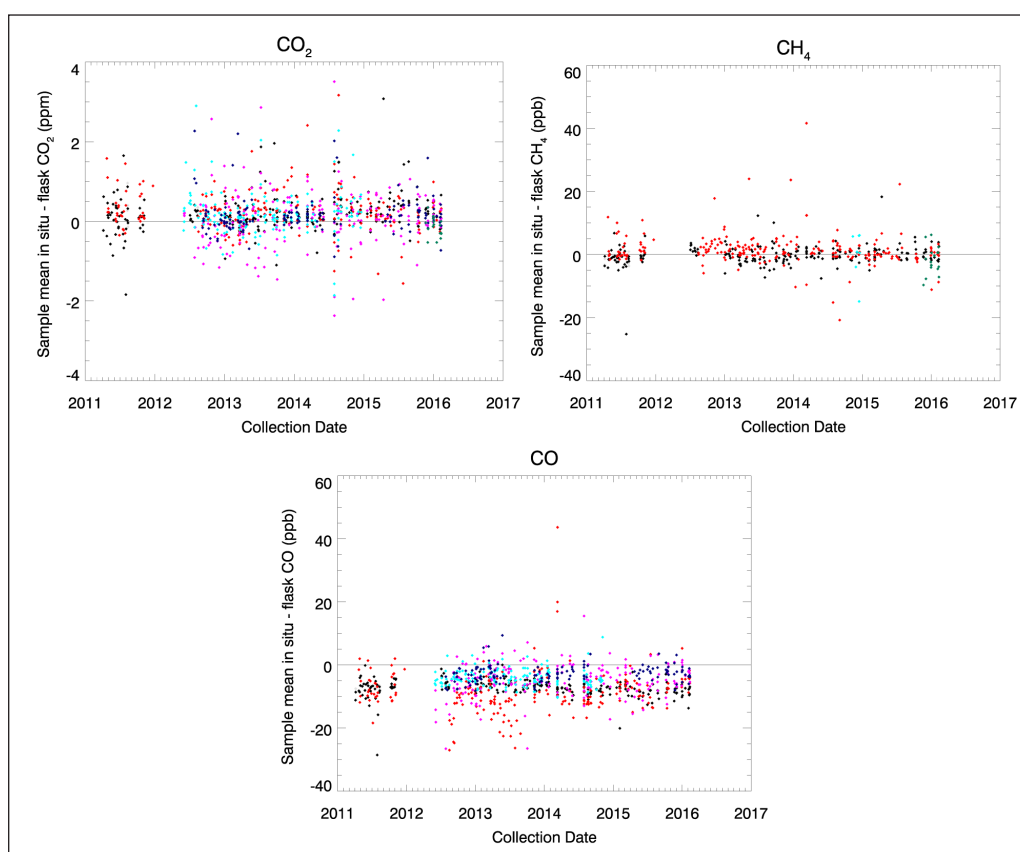


Figure 10: Hourly average in-situ – flask differences at six INFLUX sites from 2011–2016. In-situ – Flask for CO_2 , CH_4 and CO as a function of time. Each tower is indicated by a different color (Site 01 = black, Site 02 = red, Site 03 = pink, Site 05 = cyan, Site 09 = blue, Site 10 = green). DOI: <https://doi.org/10.1525/elementa.140.f10>

each instrument has a different water vapor dependence for CO (Chen, 2013). Plans are in place to implement more thorough drying at the six INFLUX sites measuring CO as well as add another target tank measurement that does not go through the Nafion drier. Note that tanks have been shown to drift in CO (NOAA-GMD, 2017) and this was a possible explanation for the bias in the CO comparison. However, no change in the bias with time is evident in **Figure 10**.

3.4 Total data availability

The number of CO_2 , CH_4 , and CO measurement sites increased throughout the INFLUX project as new sites were deployed. There were twelve measurement sites for CO_2 by early 2013. The number of CH_4 measurement sites increased gradually until late 2014 by which time nine measurement sites had been installed. The number of CO sites increased from two sites to five sites in June 2012. **Figure 11** shows the total number of sites reporting data for each species for each day from 1 January 2011 to 31 December 2015. Note the times in mid-2013 during which more sites were reporting measurements of CH_4 and CO than were “officially” deployed; this was due to the use of spare G2000 series instruments during repairs. Most, but not all, missing data is due to instrument failure (hardware or software); there were several brief (i.e., several days) power outages during the 5 years of measurements.

There were ten manufacturer repairs (most often requiring laser replacement) during 43 site-years of data collection. In addition, field repairs (most often replacement of logic board, motherboard, or hard drive) were successful six times during that period. Software problems, fixed by the manufacturer remotely or via a hard reboot, occurred approximately ten times. Minor software problems requiring a system reset are not included in this total. Early in the project, pump failures caused data outages as well, but replacement with Vacubrand MD1 and ME1 pumps alleviated this problem.

4. Conclusions/Discussion

To increase measurement accuracy, calibration (slope and offset) using three to five WMO-traceable tanks was performed prior to deployment and after each manufacturer repair or operating system upgrade. Air samples were dried to below 0.2% H_2O (0.6% H_2O at Site 02) to avoid the need for yearly water vapor calibrations (Chen et al., 2013). This level of drying was found to be adequate for CO_2 and CH_4 , but may have been insufficient for the CO measurements.

A daily target tank measurement was used to apply a offset correction, document instrument performance, and diagnose problems. Instrument failures were sometimes preceded by months of noisy target tank measurements, although this could not reliably be used as a predictor. Less frequent sampling of the target tank

Table 1: Mean difference between in-situ and flask dry air mole fraction, as measured at the INFLUX towers^a. DOI: <https://doi.org/10.1525/elementa.140.t1>

Species	ALL ALL (In-situ standard deviation < threshold*)	Tower 01 Tower 01 (In-situ std < threshold*)	Tower 02 Tower 02 (In-situ std < threshold*)	Tower 03 Tower 03 (In-situ std < threshold*)	Tower 05 Tower 05 (In-situ std < threshold*)	Tower 06 Tower 06 (In-situ std < threshold*)	Tower 09 Tower 09 (In-situ std < threshold*)	Tower 10 Tower 10 (In-situ std < threshold*)
CO ₂ (ppm)	0.18 ± 0.56 0.13 ± 0.36	0.22 ± 0.48 0.15 ± 0.34	0.25 ± 0.52 0.15 ± 0.37	0.10 ± 0.74 0.09 ± 0.45	0.19 ± 0.55 0.11 ± 0.26	0.20 ± 0.51 0.09 ± 0.17	0.18 ± 0.46 0.14 ± 0.36	-0.02 ± 0.42 -0.07 ± 0.45
CH ₄ (ppb)	0.62 ± 5.11 0.15 ± 3.19	-0.23 ± 3.11 -0.38 ± 2.04	1.49 ± 5.39 0.86 ± 4.23	N/A	N/A	N/A	N/A	0.92 ± 10.2 0.19 ± 1.31
CO (ppb)	-5.9 ± 5.0 -5.8 ± 4.0	-7.7 ± 2.9 -7.4 ± 2.4	-8.1 ± 6.7 -8.5 ± 5.1	-4.6 ± 5.3 -4.9 ± 4.2	-3.9 ± 2.8 -4.1 ± 2.5	-8.1 ± 3.5 -7.2 ± 3.6	-2.7 ± 2.6 -2.8 ± 2.4	N/A

^aTop row for each species includes all data. Bottom row excludes hours for which the in-situ standard deviation is larger than a threshold (1.0 ppm for CO₂, 7.0 ppb for CH₄, and 15.0 ppb for CO). CH₄ was not measured at Towers 03, 05, 06, or 09, and CO was not measured at Site 10.

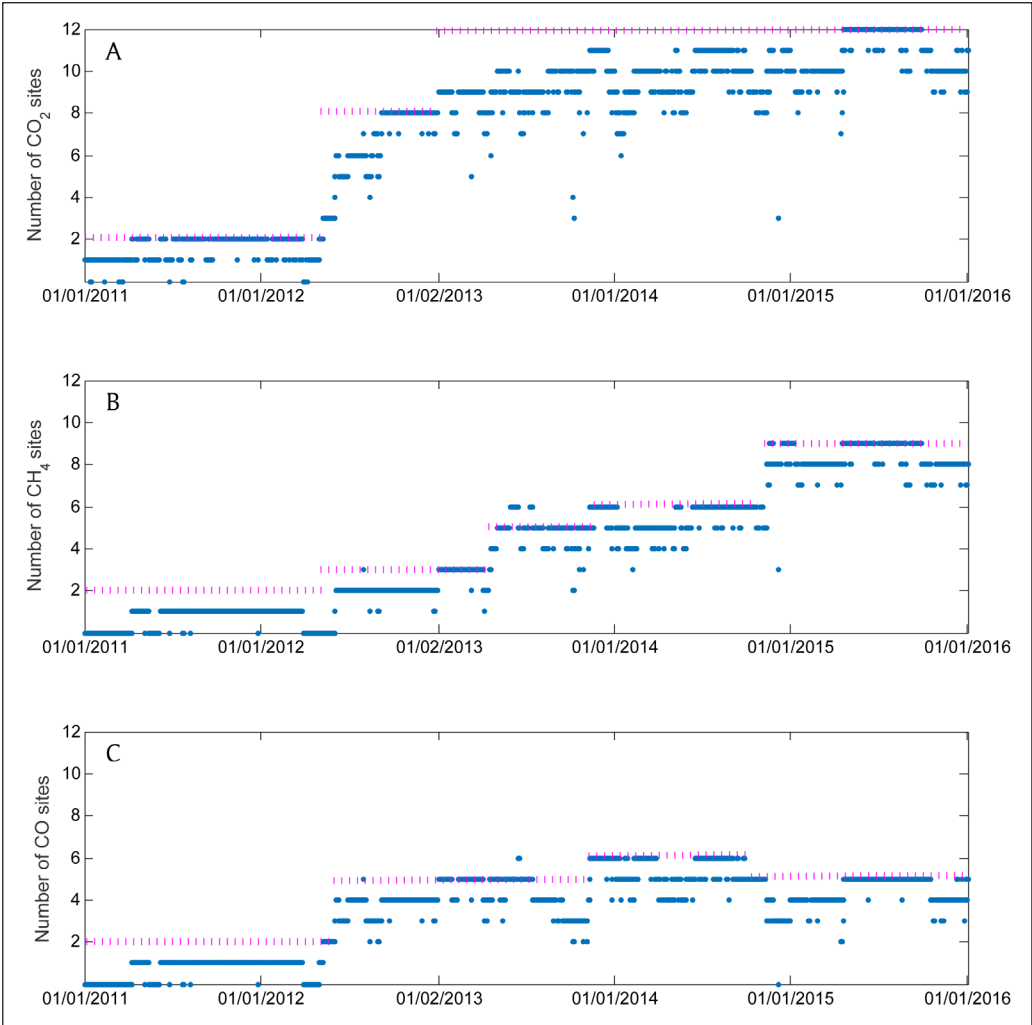


Figure 11: Total number of sites reporting data for each species from 1 January 2011 to 31 December 2015. Total number of sites reporting data. Pink markers indicate total number of deployed sites; blue dots indicate number of operational systems. A CRDS instrument with CO was used in mid-2013 and thus more sites were online than officially deployed. Also, a sensor with CO was deployed for much of 2014 while that site’s instrument was being repaired, thus the number of possible CO sites was 6 for most of 2014 instead of 5. DOI: <https://doi.org/10.1525/elementa.140.f11>

could be employed, but at the flow rates used here, the replacement frequency was 3–4 years. Two tanks would have been beneficial in order to detect possible tank drift, but round robin testing was used to detect target tank drift and tank assignment errors. The difference between **Figure 9A**, which included CRDS instruments that had

not been calibrated for several years, and **Figure 9B**, in which all instruments had been calibrated less than 2 years prior, demonstrates the usefulness of calibrations every 1–2 years. Yver Kwok et al. (2015) reported that, even after a simple instrument restart, there could be a shift in instrument response; this was not detected in instruments used in the INFLUX project, although some unexplained shifts in the target tank offsets occurred (**Figures 3, 4 and 5**).

One-hour integrated flask samples were collected at six INFLUX sites for a direct comparison with one-hour averages of in-situ CO_2 , CH_4 , and CO. The (in-situ – flask) difference for CO_2 and CH_4 were within uncertainty bounds of the comparison. However, CO showed a clear bias with in-situ being an average of 6 ppb lower than the flask samples. A possible cause was tank drift or insufficient drying of the CRDS air sample because, although the air samples were dried, a small water vapor correction was still required and applied to the CO_2 and CH_4 data; no water vapor correction was available for CO.

From 1 Jan. 2011 to 31 Dec. 2015, the INFLUX project represented 43 site-years of CRDS measurements at tower sites. Total data availability shown in Figure 12 demonstrates that spare sensors were required to replace faulty sensors to maintain high data availability. Typical manufacturer repair times were 8–12 weeks. INFLUX sites were deployed at communication towers with backup power so power outages were very infrequent (occurring less than 5 times since the project began in 2011) and brief when they did occur. Over the 43 site-years of the project, ten failures occurred that required manufacturer repair of the instrument, six sites were field repaired, and about ten software failures occurred that could be remotely repaired by the manufacturer. Other more frequent software-based problems were alleviated with a hard reset of the instrument, requiring a site visit unless a remote power switch was installed.

For the 2013 round robin testing, the error depended on instrument and length of time since calibration; the network-averaged errors were -0.09 ± 0.11 ppm CO_2 . The 2015 round robin testing showed improved results, with network-averaged errors of 0.00 ± 0.05 ppm CO_2 . The mean bias according to the flask to in-situ comparisons was $0.18 \text{ ppm} \pm 0.55$. Taking the magnitude of the largest of these can be used as a compatibility bound, the compatibility of the INFLUX CO_2 measurements is thus estimated to be 0.18 ppm.

For CH_4 , the round robin tests indicated network-averaged errors of 0.2 ± 0.4 ppb in 2013 and -1.0 ± 0.8 ppb in 2015. The flask to in-situ comparisons yielded a mean bias of 0.6 ± 5.0 ppb. Overall, the CH_4 compatibility bound is thus estimated to be 1.0 ppb.

For CO, the round robin tests resulted in network-averaged errors of 0 ± 2 ppb in 2013 and 1 ± 2 ppb in 2015. The flask to in-situ comparisons yielded a mean bias of -6 ± 4 ppb. Overall, the CO compatibility bound is thus estimated to be 6 ppb. The bias may be decreased by drying the air sample to less than 0.1% H_2O , rather than the 0.2 – 0.6% H_2O for the results presented here. It is also possible that tank drift in CO contributed to the

relatively large flask to in-situ differences and/or that this is a problem with the flask sampling or analysis.

In this paper, we have documented the calibration, air sampling methods, and performance of the twelve INFLUX tower-based in-situ CO_2 , CH_4 , and CO instruments. These data have been used to establish spatial and temporal patterns in urban greenhouse gas mole fractions (Miles et al., 2017) and to determine inversion-based urban emissions of CO_2 (Lauvaux et al., 2016) and CH_4 (Lamb et al., 2016).

Data Accessibility Statement

The INFLUX tower data are available at sites.psu.edu/INFLUX.

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Competing interests

The authors have no competing interests to declare.

Author contributions

- Contributed to conception and design: SJR, NLM, KJD, TL, JCT, CS
- Contributed to acquisition of in-situ data: SJR, NLM, DKM
- Contributed to acquisition of flask data: JCT, KM, CS
- Contributed to analysis and interpretation of data: NLM, SJR, JCT
- Drafted and/or revised article: NLM, SJR

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