

Interactive comment on “A multi-year record of airborne CO₂ observations in the US Southern Great Plains” by S. C. Biraud et al.

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Reviewers' comment: Page 8: The authors claim an accuracy of 0.1 ppm for more than 329 flights. I think this value remains quite optimistic, and may not take into account all bias.

Authors' response: The reviewer does not suggest specific bias that we might not have accounted for, so to address the comment we have listed the different bias we try to account for in our study:

1. Bias of the analyzer sub-systems. Each analyzer uses a set of three cylinders for calibrations (one responsivity cylinder, one differential zero cylinder, and one target

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cylinder). These calibrations tanks are re-calibrated against NOAA primary cylinders every 4 months (more frequently than what we ultimately aim for). There are no moving parts of the analyzer sub-systems, and they have negligible sensitivity to platform motion. Pressure is controlled to 0.1%. Temperature difference between the reference and sample cells is made zero by passive design. The one place where there would be a concern is the hermetic seals of the collimating and concentrating optics. Each has a chemical getter for CO₂, and any slow drift in the sealing mechanism should be compensated by the frequent calibrations of differential zero and responsivity. Further, the value of responsivity has remained the same aside from seasonal dependence of ~15% from the changes in ambient temperature (17 versus 20 ppm/count). It is worth noting that temperature control of the photo-detectors attenuates the effect of ambient thermal drift. It is not absolute. Colder photo-detectors will give smaller responsivity. Decrease in sample temperature, also during the colder months, will tend to decrease responsivity further. Behavior of the responsivity remains consistent with good hermetic seals. Steps in pressure do not show corresponding steps in CO₂ response as would be expected of a rupture in hermetic seals. There is no evidence of bias of the analyzer sub-module.

2. Storage fidelity of the on-board calibration cylinders. We have studied several types of cylinders that have size appropriate for inclusion into our continuous high precision analyzer systems. Not all cylinders show a significant drift in CO₂ concentrations, and we have been able to identify a few types that can be used for our missions considering our targeted half year maintenance cycle. For smaller cylinders (500 cc and 200 cc), appropriate for responsivity and target cylinders, drift is steady and no more than ~0.2 ppm per half year. Cylinders preparation is being improved to reduce the already small drift to lower values. For the larger differential zero cylinder (3000 cc), drift is about 0.10 ppm as demonstrated by a great many calibrations of RM0 in the field and its flight reference tank. It is expected that this drift will be made negligible at the 0.10 ppm level by curing and preparation practices that are yet to be tested for this size cylinder.

3. More about Storage fidelity of system references. The on-board calibration cylinders, integrated into the analyzer systems, are exchanged out of phase. In this manner, it becomes possible to use the performance of the unchanged analyzer as reference for the changed one containing the new calibration cylinders.

4. Bias associated with gas processor. As already explained, calibration of system references is done by the introduction of WMO reference gases at the system inlet at ambient pressure. This protocol cancels any small bias associated with the gas processor consisting of the compressors and driers. The bias itself has been shown to be small to negligible by teeing sample from a 30 liter tank of drier reference gas, one line going to the differential zero and the other to the system inlet by buffer vented to ambient pressure. There is no evidence of substantial bias being associated with the drier. Early in the development cycle, we were encouraged to send reference gas through the same drier used for the wet atmospheric sample. This proved to be an unnecessary, inefficient strategy. AOS built a custom membrane drier, and testing showed that it has negligible bias.

5. More about bias of the gas processor. The chemical drier cartridges are replaced out of phase for the broadband pair of analyzers. In this manner, it is possible to use one analyzer, the unchanged one, as reference for the one that was changed.

6. Bias from introduction of WMO reference gases. We used fourteen calibration cylinders (concentrations ranging from 350 ppm to 450 ppm) to calibrate the on-board calibration cylinders of both analyzers at their inlets. Unless all fourteen of the original ones, half of which were purchased from NOAA and the other half referenced as secondary standards to NOAA tanks, have significant bias relative to the WMO scale, the error in absolute scale should be negligible.

7. Bias of ground-based calibration versus in-flight calibration. Unlike the situation with the existing flask technology, it is possible to calibrate the response of the continuous analyzers by the introduction of reference gas on the platform at ambient values of

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pressure and temperature for actual mission profiles. This procedure is still being perfected, but already it has shown that there are no fluctuations to explain the substantial atmospheric fluctuations, some of which are being observed in the FT. Values of the reference gas are being predicted to ± 0.1 ppm so far. More calibrations of this kind are planned, and a new generation of flask technology should be invented that enables the same kind of in-flight calibration with and without drying of atmospheric sample during acquisition.

Reviewers' comment: Figure 5 shows indeed a good precision and accuracy of RM0 during one hour. There is not much detail about the condition of this experiment. What is performed on the ground? at a constant elevation ? Was the reference gas passed through the drier?

Authors' response: A small cylinder (3000 cc) was added to the aircraft's payload for a ground-based measurement of system noise. In this manner, the pair of analyzer systems could draw dried sample gas at ambient pressure for an extended period of time as the means to do a full test of the integrated systems. The reference gas from the small tank was flushed through the drier of each system. This test was performed on the ground with both instruments integrated in the aircraft as they were used for the airborne missions reported in the paper. Figure 5 shows an on-platform test of the response of the analyzer systems to a common stream of CO₂ concentration. It was not done during a flight as stated in the text. A similar experiment, but this time during a typical mission profile, was done on September 2012, after the paper was submitted.

Caption for Figure 5 was changed to: "Accuracy and precision in the aircraft on the ground for the two continuous CO₂ systems (black=RM0 and blue =RM12) estimated from the measurement of CO₂ concentrations delivered by a cylinder maintained at ambient pressure and flushed continuously by a stream of reference gas calibrated earlier in the laboratory. Precision of RM0 and RM12 as shown on top panel was 0.10 ppm (standard deviation of N=2814 observations) and 0.25 ppm (standard deviation of N=2937 observations), respectively. Accuracy as shown on bottom panel, including

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the specific mission calibration and accuracy of delivery of reference gas, was 0.13 ppm and -0.06 ppm for RM0 and RM12, respectively.”

Reviewers’ comment: The comparison of RM0 and RM12 shown on figure 4 has a mean difference of 0.04 ppm. Still this difference doesn’t has a very strong meaning considering the structure of the differences being -0.3 ppm during most of the ascent, and +0.3 ppm during most of the descent. Have you plot the differences versus altitude?

Authors’ response: This is a good question. To address the reviewer’s request, we made a graph of the differences between RM0 and RM12 as a function of altitude. The graph (see attached Figure 1, response to reviewer #2 comments) does not show altitude (pressure) dependence of the (RM0 - RM12) difference. We do note that a small error in phasing between the two continuous analyzers may add to the difference, especially where the atmosphere has fast, substantial fluctuations. Typically, the lowest levels of a vertical profile are in the PBL and have the largest fluctuations.

For the observations in our more than 300 missions, errors of the type illustrated in Figures 4 and 7 average down to high precision (See Figure 6d.) We want to emphasize that fluctuations in the atmosphere are substantially larger than the instrumental errors of the continuous analyzers. That is the actual comparison that should be made for the performance of the analyzers. Indeed, the analyzers could be noisier and still provide almost as much information as long as they have negligible dynamical and static biases over long integration times. Æ Reviewers’ comment: Regarding the flask/in-situ comparison, the storage drift described in page 8 (and on the ESRL website), corresponding to a CO₂ depletion over time, is not applied in the figure 6. Using a mean correction of -0.2ppm for a 3 weeks storage time, the mean difference would be about -0.2 ppm overall and -0.4 ppm in the free troposphere. By the way how do you explain the larger bias in the free troposphere compare to lower altitude? Could it be related to a pressure effect on in-situ measurements?

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Authors' response: The reviewer is correct that a storage correction term was not applied. It was decided by NOAA that flask-based observations should be presented without correction. This potential issue has to be investigated and resolved by NOAA. (Note that the average storage bias of 0.2 ppm is described on the NOAA/ESRL website <http://www.esrl.noaa.gov/gmd/ccgg/aircraft/qc.html>). Both the flask technology and the continuous measurements can have errors, and that is why we integrated a second continuous analyzer on the airborne platform. This provides an expanded set of validations consisting of the double-blind comparison between each analyzer and the flask technology. As shown in the paper, we have developed a strategy to better quantify measurements errors, and a follow-up paper describing and quantify these errors is in preparation. We do not have an answer as to why the bias was larger in the free troposphere. We evaluated a possible dependence on altitude (pressure) of the mean values and standard deviation of the RM0 - flask difference. This is shown in the attached Figure 2. The same altitude dependence described in Figure 6b and 6c is shown, indicating that large fluctuations of CO₂ concentration in the PBL are not well captured by the flask technology.

Reviewers' comment: Lines 23-25/page 10 seem in contradiction with the results of figure 6.

Authors' response: Figure 6 gives the histograms of observations. Figure 6b shows RM0 analyzer minus flask values for higher altitudes, while Figure 6c shows RM0 analyzer minus flask values for lower altitudes. I assume that the reviewer is referring to the mean values shown on the Figures 6b and 6c, while the text is referring to the standard deviation of the mean values We added the following sentence to the text page 10, line 23-25: "Figures 6b and 6c show that the standard deviation of the mean difference between RM0 and flask samples is smaller in the FT (0.43 ppm) than in the PBL (0.81 ppm).".

Reviewers' comment: You should also explain more precisely which part of the in-situ measurements you are using to compare with the flask samples. Are you using the

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whole average at a given level, or do you extract the closest time period +/- 30sec?

Authors' response: We updated the text in the paper to explain what part of the continuous measurements is used to compare with flask samples. The last sentence of Figure 6's caption has been changed to: " Each point refers to the mean difference between a flask sample and the 1-minute average of the continuous observations centered around the time that the flask was filling, one for each of the 12 steps during descent."

Reviewers' comment: There is a recent publication describing a similar program with the same type of in-situ analyzer: 'Variation of CO₂ mole fraction in the lower free troposphere, in the boundary layer and at the surface', Haszpra et al., JGR 2012. The uncertainty of the in-situ measurements estimated for this program is higher than 0.1ppm. Is the setup different from SGP flights?

Authors' response: Although the continuous CO₂ analyzers deployed by Haszpra and by LBNL were manufactured by the same company (AOS Inc.), we suspect that the observational setup of the two programs is fairly different. Our instrument was modified in various ways while we were integrating and testing on the airborne platform. Below is a list of factors affecting airborne observations that we have focused on:

- Maintain long term integrity of the analyzers (including hermetic seals, observational protocols and data processing program).
- Maintain calibration traceability to WMO scale.
- Define calibration protocol for in-flight calibration.
- Test fidelity of storage for the system reference tanks for long maintenance cycles of half year.
- Perform regular reference-tank flights where a target cylinder is substituted to atmospheric sample during actual mission profiles.

- Use phased broadband technology to detect problems at an early stage and staggered maintenance of continuous analyzers.
- Use and test custom built membrane drier and test against the commercial ones. - Test maintenance schedule of the expendable component of drier.
- Control and verify performance of the flask technology.
- Use a platform dedicated to the monitoring program alone that houses the payload permanently.
- Test effect of gas storage in the analyzer's manifold and gas processor (this led to the elaborate pre-flight flushing protocol).
- Make observations available to researchers so that they may evaluate self-consistency of the observations.

We have added a citation to the Haszpa paper (page 7193, line 28). The following sentence was added: "This type of analyzer has been used by other research groups located in Germany, Spain, and Hungary (Haszpa et al., 2012)."

Reviewers' comment: Page 5: Number of levels: the last paragraph gives the impression that only two levels are sampled, whereas the first paragraph is stating 12 sampled levels.

Authors' response: The typical flight pattern always had 12 levels. However, the number of flasks available for air sample collected changed with time. Starting in 2002, we had only a 2-flask sampler (page 7192, line 23) and thus sampled at two altitudes, and in 2006 we added a 12-flask sampler (page 7193, line 8).

Reviewers' comment: Page 10: Detection of problems (line 7-10): could you give more details on the kind of problems which were detected thanks to the multiple technologies program?

Authors' response: (see answer to reviewer #1) During early phases of our program

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(prior 2007) when RM0 was the only continuous analyzer on board the platform, there were discrepancies continuous and flask-based observations. We determined that the commercial membrane drier of the analyzer had developed leaks and was not robust enough for the airborne platform. A custom membrane drier was constructed that solved the problem. Later, we found that the original second stage of drying by chemical cartridge (Magnesium perchlorate) could not support the targeted maintenance schedule of 80 flight missions, each lasting at least three hours and sometimes originating in hot humid conditions. The remedy was to develop a small reusable cartridge of the same material that could be replaced by the pilot in minutes without significant manipulation of the analyzer's enclosure. Maintenance is scheduled every 10 missions during the hot humid months and 15 missions otherwise. After this upgrade, the flask and analyzer technologies gave comparable mean concentrations. Early in our program, RM0 observations displayed substantial fluctuations in CO₂ concentrations even in the free troposphere at constant altitude. Because the analyzer has solid state cells and has negligible sensitivity to motion of the platform, we predicted that these fluctuations were real. This hypothesis was verified by the addition of a second continuous analyzer (RM12) that had an intentional smaller lag of 15 sec. It was shown that the fluctuations did indeed have the proper difference in lag expected for an atmospheric origin. Various kinds of artifacts (e.g., platform power supply fluctuations, and motion sensitivity) should happen simultaneously with differential lag of zero between the analyzers. No such correlations were observed. The integration of the RM12 provided an expanded set of validations consisting of the double-blind comparison between each analyzer and the flask technology and a new form, phased Broadband Validation. It can be used to validate the fast fluctuations and transients originating from the atmosphere, not just the mean absolute levels provided by the comparison with the flasks.

In response to the reviewer comment, we have added the following text to section 2.5 of the paper: “The use of multiple technologies on the ACME platform (i.e., broadband validation) is important, because of the large changes in ambient humidity, pressure, and temperature that the platform experiences during a flight. Mean absolute

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concentrations measured by a continuous analyzer are validated by comparison with flask observations. An additional level of validation is made by comparing continuous observations to each other, one analyzer having an intentional lag of 15 seconds with respect to the other one. Atmospheric fluctuations must be detected by both analyzers, one analyzer's response to these fluctuations lagging the other analyzer's response by 15 seconds. Any fluctuations happening simultaneously or with some other differential lags in both analyzers must be viewed as artifacts. This approach has improved objectivity of the airborne platform substantially by allowing detection and diagnostics of problems in all parts of the system: leaks in the flask sampler compressor package, drift in calibration cylinders used by the continuous analyzers, and aging of the inlet tubing."

Reviewers' comment: Page 12/line 20-21: The profile observed in Figure 12 doesn't look like a 'typical' one, as written on line20, since the FT values are lower than PBL ones.

Authors' response: The claim of a typical profile depends on the season as mentioned in the text. The FT – PBL difference in Figure 12 is not in contradiction with the text as shown by Figure 11 (panel JAS). During the summer months, atmospheric concentrations in the PBL are extremely variable and concentrations can be either higher in the PBL than in the FT or vice versa.

Reviewers' comment: Figure 3: Do you pass the reference gases through the drier? Have you estimate the bias due to the drier by injecting reference gases alternatively through the drier or directly to the analyzer?

Authors' response: We do not run the on-board calibration gases through the drier in flight. But we do run reference gases through the drier when we perform calibrations of the on-board cylinders on the ground (using our primary standards to tie our calibration to WMO scale). The drier and the rest of the gas processor has been shown to have negligible bias by (1) teeing a single stream directly into the differential zero inlet of the

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analyzer, and (2) directly into the full system as substitute for atmospheric sample. Note that any small residual bias is cancelled by our calibration of the on-board calibration cylinders by running primary calibration gases into the full system as substitute for atmospheric sample.

Reviewers' comment: Figure 7: the mean difference is 0.06 ppm

Authors' response: Thank you. Figure 7 and the caption were corrected.

Reviewers' comment: Figure 9: It looks like the measurements start from 0m agl, which is not the case, except if you use surface data as combined to aircraft measurements.

Authors' response: Figure 9 was updated to reflect reviewer's comment.

Reviewers' comment: Figure 11: there is a large variability in the summer vertical profiles. Is this variability due to the diurnal cycle of the sampling (you could analyze the vertical profiles according to the timing of the flight), or rather to the origin of the air masse (you could use back-trajectories to estimate this factor).

Authors' response: We hope the paper aims at presenting observations collected over the US SGP and to start addressing a host of difficult sampling and technological issues. The kind of discussion that the reviewer is requesting (related to the origin of the certain attributes of the observations) will be addressed by a later publication. Nevertheless, we believe that the summer time vertical profile variability is associated with mixing of air masses from different origin (advection of air masses from the Gulf of Mexico region and strong convective events).

Reviewers' comment: Figure 13: it is not very clear for me how you came up with these profiles. Are you subtracting the mean vertical profile derived from flasks from the mean vertical profiles derived from in-situ measurements?

Authors' response: The graph does not show mean difference but rather standard deviation of the calculated difference. Caption was changed to: "Standard deviation of the differences in CO₂ concentration between RM0 and flask sample observations

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collected between 2007 and 2012. RMO observations were binned to one-minute averages. The mean and standard deviation of the difference between a flask sample observation and its associated 1-minute average continuous observations are calculated. Each quadrant of the graph corresponds to a 3-month average climatological vertical profile”.

Interactive comment on Atmos. Meas. Tech. Discuss., 5, 7187, 2012.

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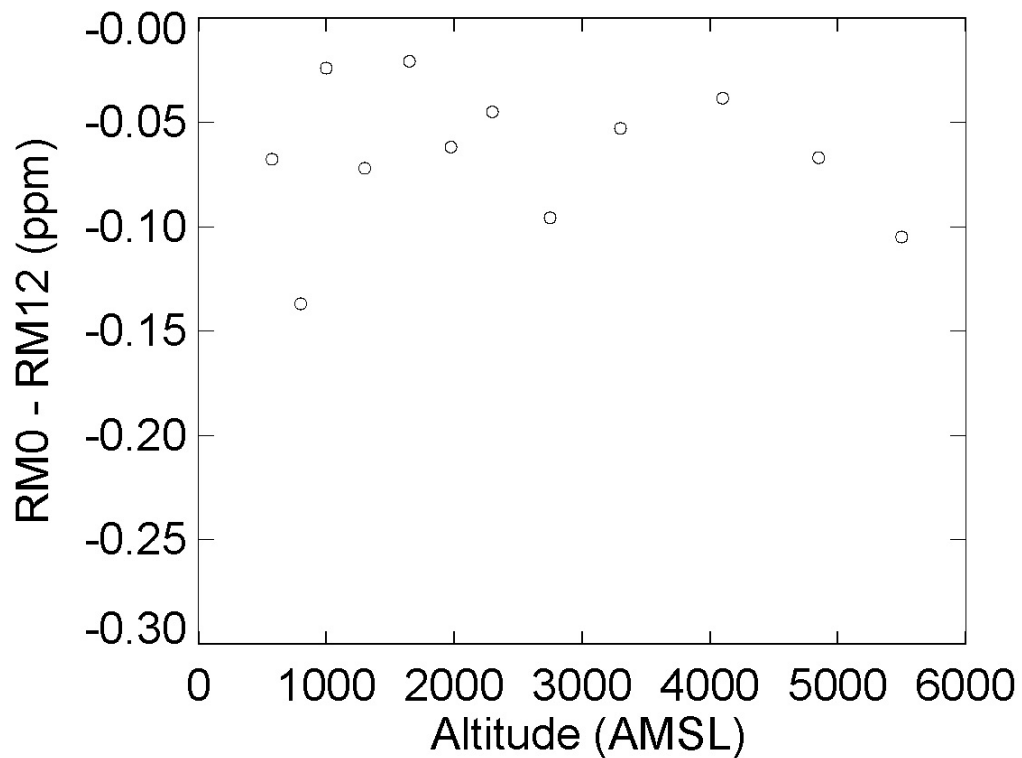


Fig. 1. Difference between RM0 and RM12 observations a function of altitude.

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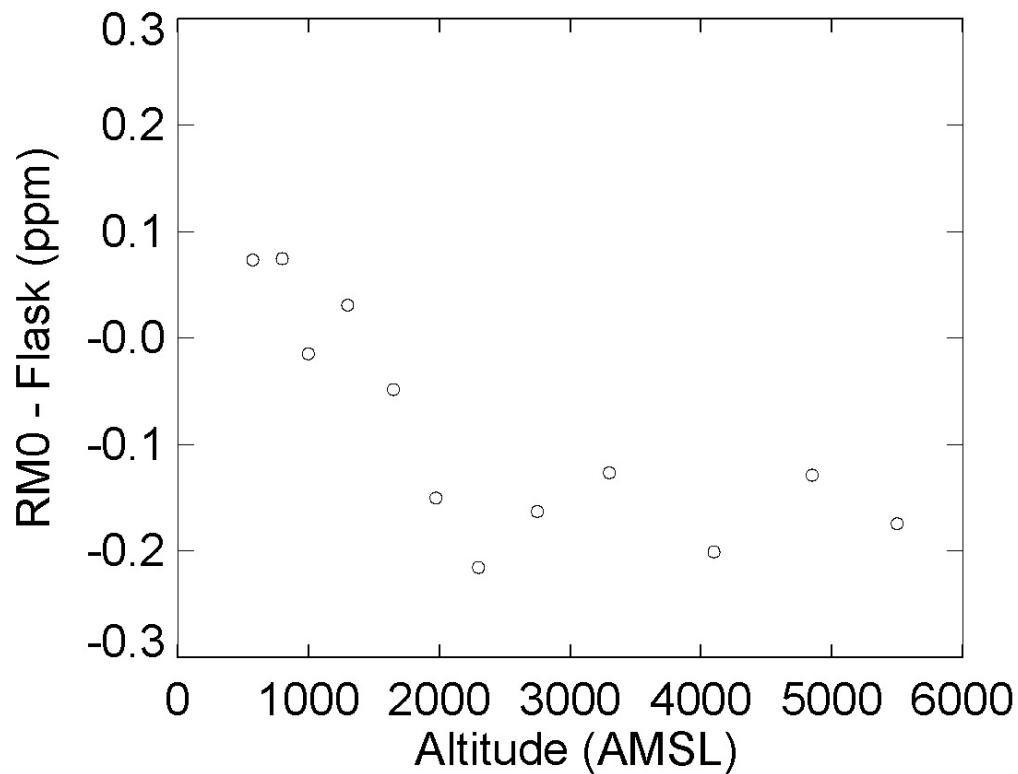


Fig. 2. Difference between RM0 observations and flask observations as a function of altitude.

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