

## ***Interactive comment on “Calibration of the total carbon column observing network using aircraft profile data” by D. Wunch et al.***

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Received and published: 16 September 2010

The authors wish to thank the two anonymous reviewers for helpful and thoughtful comments and suggestions. Each comment is addressed individually below.

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### **1 Referee #1**

#### **1.1 Specific Comments**

**Sections 3.1, 3.2, 3.3 please provide demonstrated precision and accuracy of aircraft instrumentation or include in Table 2.**

The precisions and accuracies are now listed in Table 2.

**p2612, L7 is it intended here that the lowest measured aircraft value was assumed to be the surface value?**

Yes. This has been clarified in the text:

“In the event that there were no surface or tower measurements available, and the aircraft did not measure to the surface, the lowest measured aircraft value was assumed to be the surface value (e.g. Park Falls on 14 July 2004).”

**p2612, L11-13 in order to determine the entire integrated column, it was necessary to extrapolate the aircraft in-situ profile to the lower-most part of the planetary boundary layer and to the uppermost part of the troposphere and the stratosphere. What percentage of the profile does this typically constitute?**

The aircraft profiles typically extend from just above the surface to the ~200 hPa level (the altitude ranges are now listed in Table 3). For a uniformly mixed gas like CO<sub>2</sub>, this altitude range encompasses ~ 80% of the total column. For CH<sub>4</sub>, N<sub>2</sub>O, CO and H<sub>2</sub>O, it will be more than 80%.

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**How much does this contribute to the error assigned to the integrated aircraft column?**

The error budgets, broken down by the stratospheric, surface and aircraft contributions are in new Table 4.

**Has the the Network for the Detection of Stratospheric Change (NDSC) or Network for Detection of Atmospheric Composition Change (NDACC) been investigated as a supplementary source to fill in UT/LS data gaps since the assumed profile above the aircraft ceiling contributes the greatest uncertainty to the integration e.g. Raman Lidar for water vapor at Lauder?**

The water vapor profiles are generally measured by radiosondes, which reach well above the UTLS, so errors from the stratospheric water profile are not a major concern. The largest errors from a lack of stratospheric or UTLS knowledge are for CH<sub>4</sub>, N<sub>2</sub>O, CO and CO<sub>2</sub>. The mid-infrared NDACC Fourier transform spectrometers do measure those molecules, but not with sufficient accuracy and precision to be helpful here.

**Find discussion on pages 2612-2613 somewhat disjointed. Starts out discussing CO<sub>2</sub>, moves briefly to CH<sub>4</sub> and N<sub>2</sub>O, back to CO<sub>2</sub>, then GFIT of HF is introduced. Suggest leading off with the statement on p2612, L11 "In general, etc.; then discussing CO<sub>2</sub>; beginning the N<sub>2</sub>O and CH<sub>4</sub> discussion with L9-11 on p2612 followed by why it is necessary to invoke HF for N<sub>2</sub>O and CH<sub>4</sub>; then discussing GFIT, Fig 3., etc.**

Done.

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**There is mention of an air mass dependent artifact for CO<sub>2</sub> at noon vs sunrise/sunset common to all the TCCON sites that is attributable to spectroscopic inadequacies. Is work on-going beyond Rothman et al. (2009) and Toth et al. (2008) to reduce this systematic error or is the 1% absolute accuracy in XCO<sub>2</sub> considered the best that can be achieved for CO<sub>2</sub>?**

There is ongoing work in determining the parameters required to describe non-voigt lineshapes (e.g. line mixing), which may reduce our airmass dependence. Ongoing efforts are in place to implement and test these new spectroscopic models, but the work is not far enough along to include in this paper.

**As the O<sub>2</sub> dry-air mole fraction continues to decrease in the atmosphere due to the combustion of fossil fuels, will this lead to a larger error in XCO<sub>2</sub> with this technique/ approach?**

If left un-corrected, it would. Atmospheric O<sub>2</sub> is not, as assumed by GFIT, constant. The seasonal cycle in the O<sub>2</sub>/N<sub>2</sub> ratio ranges from ~50 per meg (at Samoa) to ~160 per meg (at Barrow), which is equivalent to ~10-30 ppm of O<sub>2</sub>. The atmospheric O<sub>2</sub> amounts peak in the summer, when the CO<sub>2</sub> amounts are at their minimum, and so using a constant O<sub>2</sub> amount overestimates the seasonal cycle in X<sub>CO<sub>2</sub></sub> by ~0.05 ppm.

The long-term depletion of O<sub>2</sub> is about twice the rate of the atmospheric CO<sub>2</sub> increase (~2 ppm CO<sub>2</sub>/year, giving a decrease in atmospheric O<sub>2</sub> of ~4 ppm/year). This causes GFIT to overestimate the secular increase by ~0.4%, or ~0.01 ppm X<sub>CO<sub>2</sub></sub>/year.

This small time-dependence will be built into a future version of our processing algorithm.

(Reference: The O<sub>2</sub> Balance of the Atmosphere: A Tool for Studying the Fate of Fossil-Fuel CO<sub>2</sub> Michael L. Bender, Mark Battle, Ralph F. Keeling. Annual Review of Energy

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**Recognize aircraft profiles are from flights of opportunity. Any plan for future profiles during different seasons to explore varying solar zenith angles and air mass values at each of the TCCON sites included in this analysis e.g. to calibrate Park Falls at higher air mass values?**

Yes. There are two more HIPPO campaigns planned for different seasons and including more TCCON sites. Profiles tend to be at local noon, but as you point out, in winter months, these angles are larger than in summer time. There was also a series of profiles measured over European sites during the IMECC campaign in 2009 (see Messerschmidt et al. 2010).

Messerschmidt, J., et al. 2010, EGU General Assembly 2010, held 2-7 May, 2010 in Vienna, Austria, p.2920, 12, 2920

**Presuming this analysis uses an average CO<sub>2</sub> from the two bands (6228, 6348 cm<sup>-1</sup>). It would be helpful to state this.**

Yes. A clarifying sentence has been added to the caption in Table 1.

**Fig 2 plot corresponding CO<sub>2</sub>, CO, CH<sub>4</sub>, and N<sub>2</sub>O SGP surface/tower data if available from NOAA ESRL or LBNL.**

The SGP profiles reach within 100 m of the ground, and there were, unfortunately, no surface flask measurements made at the SGP that day.

**Figures 4-8. Are INTEX-NA CO, CH<sub>4</sub>, N<sub>2</sub>O, H<sub>2</sub>O data available for inclusion in this study?**

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CO, CH<sub>4</sub> and H<sub>2</sub>O were measured on the INTEX-NA platform. N<sub>2</sub>O was not. On July 15, 2004, the aircraft flew through a fire plume, which made comparisons with the CO and CH<sub>4</sub> profile much more difficult, since the atmosphere was extremely variable. We decided not to include those profiles. On July 12, 2004, however, there was a CH<sub>4</sub> profile, but no CO profile, which is now included in the CH<sub>4</sub> calibration figure. The slope is unchanged as a result. The H<sub>2</sub>O profiles were used to compute the dry-air mole fractions.

## 1.2 Technical comments

**Fig 1 unable to see \*, + symbols**

Yes, they are unclear because they directly lie over the Tsukuba and Lamont sites. Text explaining this has been added to the figure caption.

**Table 2 Mentions NCAR Airborne Oxygen (AO2) Li-840 yet lists CO<sub>2</sub> as the measurement... does this instrument also provide an in-situ O<sub>2</sub> measurement?**

Yes, it does, but those measurements are not used in this study.

**provide lat, lon of TCCON sites used in this study**

Added to Table 3, along with the site altitudes.

**3.1 please provide seasons (months) in which START-08 and HIPPO-1 data were acquired to be consistent with sections 3.2 and 3.3; also**

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**clear up same inconsistency in Intro by either eliminating seasonal (monthly) mention or being fully inclusive. alternatively, refer the reader to Table 3 for mission dates.**

A reference to Table 3 was added in section 3, and months were added in section 3.1. References to specific seasons have been removed from the introduction.

## 2 Referee #2

### 2.1 Specific Comments

**P2607 L14 The albeit small decrease in atmospheric O<sub>2</sub> is mentioned and although well below uncertainty values in gases of interest here its not stated whether it is taken into account.**

Please see comment above to Referee #1 about the O<sub>2</sub> time-dependence.

**P2608 L17 The airmass dependence more correctly is determined by solar zenith angle not time of day and its range is then dependent on latitude. There has been much recent work on these CO<sub>2</sub> line parameters and are arguably better than other gases why the issue with CO<sub>2</sub> not the other gases? (note this may be discussed in Wunch 2010 but that is not yet available.)**

The airmass dependence is determined as a function of the solar zenith angle in our analysis. The portion of the sentence: "which causes the retrievals to be ~1% larger at noon than at sunrise or sunset" has been replaced to clarify this point: "which causes

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the retrievals to be ~1% larger at low solar zenith angles than at high solar zenith angles."

A significant portion of the airmass dependence is due to that of the O<sub>2</sub>. The airmass dependence of the other molecules may partially compensate for that in O<sub>2</sub>, or it may be that the larger random errors on the other molecules masks any airmass dependence.

**Table 3 Since each aircraft flight maybe different expanding table 3 to include the altitude ranges of the profiles used helps the reader know from what altitude range the compared information is derived.**

Done.

**P2610 L5 The averaging kernels yield what the retrieval process does to a given state vector with all its errors and approximations built into it, so it may be an interpretation 'if it were measuring perfectly' but this may be better stated to say that eg 'this is what the retrieval should return given the in situ profile'.**

The sentence has been changed to: "The smoothed profile represents the profile that should be retrieved, if the FTS were measuring the true atmospheric profile without spectroscopic errors, given the GFIT a priori profile..."

**P2610 L6 'ie without spectroscopic errors' infers that this is the only error in the data, was 'eg' meant?**

This has been removed and replaced with the sentence above.

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**P2610 Eq 2 & 2613 L9** The importance and usefulness of the retrieval averaging kernels in comparing to the in situ profiles is well noted and used here as is their representation of the sensitivity of the measurement. These should be plotted for each gas noting the variation extent with SZA

A new figure (3) has been added showing the column averaging kernels and their solar zenith angle dependence.

**P2612 L8** Uncertainties are mentioned here and elsewhere (Sec 5) in particular the large contribution of the unknown stratospheric apriori profile. Although error bars are given in plots 4-8 a table of estimates of the known uncertainties should be used to pull this important information together.

A new Table 4 has been added, which breaks down the sources of the errors into stratospheric, surface, and aircraft components.

**P2614 L11** Its been shown recently that there is a strong correlation with the difference in total water vapor measured by two instruments and time lag between those measurements (Sussmann et al. *Atmos. Chem. Phys.*, 9, 8987–8999, 2009). What time lag do the measurements in F5 represent? Are they nearest at time of launch, after 30min?

All the  $H_2O$  FTS measurements in Figure 5 are 1-hour averages of individual retrievals centered on the time of the sonde launch. A reference to Sussmann et al. (2009) has been added to section 2.

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**More generally the dry air mole fraction can be viewed as a weighted mean VMR. Is this useful for water vapor which so rapidly falls off with altitude?**

We agree that dry-air mole fraction is a strange name for  $H_2O$ . We can also express the same quantity in the more well-accepted precipitable water (in cm). Converting to precipitable water involves adding more error, because the surface pressure at the time of the measurement at the location of the sonde launch and at the FTS becomes critical. When complete, this method gives a slope that is consistent within error of the current slope. Because this paper is meant to describe the calibration of the TCCON data product, we feel that the current plot should remain, to be consistent with the method described in the text.

**P2614 L11** What is meant by duration of the measurements? Do the points in Figs 4-8 represent one spectrum? Or several averaged? If not the same for all data points perhaps a table should be used to show this. It is correct to assume these are a standard data product?

This has been clarified in section 2:

"For comparison with the aircraft profiles, which are not measured instantaneously, averages are taken of TCCON  $X_{CO_2}$ ,  $X_{CH_4}$ ,  $X_{CO}$ , and  $X_{N_2O}$  columns retrieved while the aircraft measurements were taking place, weather permitting. Typical durations are between 0.5-4 hours. The standard deviations ( $1\sigma$ ) of the measurements are taken as the TCCON errors. One hour of TCCON measurements of  $X_{H_2O}$  are averaged to compare with each sonde profile, centered on the sonde launch time. Twice the standard deviations ( $2\sigma$ ) of the  $X_{H_2O}$  measurements are taken as the TCCON errors, because the atmospheric variability of  $H_2O$  can be much greater than for the other molecules (Sussmann et al., 2009)."

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**P2612 L13 – 2613 L13** Profiles of CH<sub>4</sub> and N<sub>2</sub>O are qualitatively similar but not identical and also not mirror images of HF. Accounting for ascent/descent in polar regions for instance using vertical shifts where the descent is the dominant dynamical feature, is a reasonable procedure. Inferring an ascent/descent because retrieved HF is not as expected seems to rely heavily on a prior certainty in the HF column as well as its relation to CH<sub>4</sub> and N<sub>2</sub>O and in the case of a network globally. This should be elaborated on.

This has been clarified in section 4:

"Stratospheric N<sub>2</sub>O and CH<sub>4</sub> mole fractions are more difficult to estimate than CO<sub>2</sub> because they decrease rapidly with altitude, causing transport-driven variations in the stratospheric column. To account for these transport-driven variations, columns of HF can be used, which are measured coincidentally with N<sub>2</sub>O and CH<sub>4</sub> by the TCCON FTS instruments. Due to a complete absence of HF in the troposphere, HF is a sensitive indicator of ascent and descent in the stratosphere. Indeed, a 1 km vertical shift in the HF profile produces a ~15% change in the total column, which is easily measurable. Furthermore, strong stratospheric CH<sub>4</sub>-HF and N<sub>2</sub>O-HF correlations have been observed globally by Luo et al., 1995 and Washenfelder et al., 2003, which we exploit in this analysis to determine the best stratospheric profile for a given overpass."

What was Figure 3 now has an extra panel showing the correlation between HF and CH<sub>4</sub> from the ACE-FTS and MkIV balloon flights over many years and latitudes and our a priori profiles are constrained to lie on that curve (black solid line). The correlation curves are very similar for all years and latitudes. A similar picture is true for N<sub>2</sub>O.

**Using this method what range of HF shifts are seen (one example gives .8km in Fig3)? Is there a dependence by station?**

The average adjustment is 0.4 km, and the maximum adjustment is 1.8 km. This is  
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now in the text. There does not appear to be a dependence by station.

**The uncertainty approximation determined by shifting by one km yields what column error?**

This is listed in new Table 4. It is on average 10 ppb for CH<sub>4</sub>, 4 ppb for N<sub>2</sub>O and 1.5 ppb for CO.

**Also at sites which have higher resolution NDACC FTS instruments which are sensitive in the lower stratosphere – can the be used to directly measure CH4, N2O and possibly CO in that region?**

This is an interesting suggestion. However, we see three main drawbacks to this approach:

1. The NDACC FTS instruments measure in the mid-infrared spectral region, and hence use different spectroscopic line lists.
2. The NDACC FTS instruments, to our knowledge, are not calibrated to the accuracy required here.
3. Many TCCON sites are not co-located with NDACC FTS instruments, and so could not benefit from this additional dataset.

## 2.2 Technical Comments

**P2606 L1 : should be e.g. <0.25%...**

Done.

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**P2610 Eq 3 when going from matrix multiplication in eq2 to the vector in eq3 A changes to aT. This is not mentioned in the text nor is it necessarily standard.**

This is mentioned in the text on P2610 L15 and now has the added phrase "a vector containing":

"a is a vector containing the FTS dry pressure-weighted column averaging kernel"

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Interactive comment on Atmos. Meas. Tech. Discuss., 3, 2603, 2010.

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