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Atmospheric Measurement Techniques Discussions

Interactive comment on "Greenhouse gas measurements over a 144 km open path in the Canary Islands" *by* J. S. A. Brooke et al.

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We would like to thank both reviewers for their time and effort and for their valuable comments. We have carefully considered everything, and our point-by-point response is outlined below. Reviewers' comments are repeated in normal text, and our responses are in *italics*. The page and line numbers quoted refer to the draft revised manuscript, which is attached as a supplement.

REVIEW 1

However, I have a problem with the overall conclusions. For example with the last sentence given in the abstract stating that from the results presented one can conclude

that the new method has a sound basis for monitoring CO2 and other greenhouse gases in the free atmosphere. Or with the first sentence of the coclusions (Sect. 4) starting with "We have successfully demonstrated ...". These statement are too strong and need to be re-formulated. This is because the reported uncertainty (+/- 14.7 ppm) appears to be quite large and a discussion is missing what the required accuracy and precision is and if the instrument is able to meet these requirements. I therefore recommend to add a discussion on what the requirements (and related applications) are for monitoring CO2 and other greenhouse gases in the free atmosphere and to that extent they are met or not.

We have revised the "Discussion and Conclusion" section overall to account for these comments, as well as for similar concerns of Reviewer 2. We also changed the final sentence of the abstract accordingly, to align it with the new conclusion text. We avoided a detailed discussion of requirements, as we see this as beyond the scope of what is needed for context in this first demonstration paper, but added brief text and two additional citations, Rayner and O'Brien (2001), and Larsen et al. (2009); the latter was the basic publication of ACCURATE-related requirements in ESA context.

The full revised text is as follows (pages 7-9):

"We have successfully demonstrated that atmospheric carbon dioxide concentrations can be determined from SWIR absorption measurements over very long path lengths, with relatively low power diode lasers (4 to 10 mW). The accuracy of these demonstration measurements ($\pm$15 ppm for CO_2) is limited by errors in determining the temperature and pressure along the atmospheric path length, uncertainties in the least-squares fitting procedure (partly due to low signal-to-noise ratio and errors in spectral line parameters – see below), and problems in the field associated with a detector offset error. The static link between the Islands also does not demonstrate the scanning of the atmosphere which occurs between LEO satellites, and the experiment was too short to monitor trace gas variability over time. A detailed description of how the monitoring with the LEO system works is given by Kirchengast et al. (2010).

In general, the desired precision for remote sensing of CO_2 for carbon cycle science is about 1 ppm (Rayner and O'Brien, 2001); a detailed discussion of the observational requirements for the ACCURATE concept is available in Larsen et al. (2009), adopted by the mission proposal of Kirchengast et al. (2010). While the accuracy of this first demonstration experiment is not ideal, previous studies (Kirchengast and Schweitzer, 2011; Proschek et al., 2011) indicate that greenhouse gas profiles for an ACCURATE mission are obtainable with <1 to 4% r.m.s. error (outside clouds; above 5 km; the goal for CO_2 is <1%). The sources of error contributing to the value of $\pm$15 ppm are expected to be significantly smaller for an ACCURATE mission than in this least-cost demonstration. The detector offset error is a fixable issue (see Sect. A5) and significantly more accurate frequency knowledge and higher signal-to-noise ratios will be available from advanced instrumentation. Furthermore, accurate temperature, pressure and humidity will be determined from simultaneous microwave occultation measurements (Kirchengast and Schweitzer, 2011; Schweitzer et al., 2011b), and a more accurate retrieval algorithm (Proschek et al., 2011) will be used to extract greenhouse gas concentrations from the infrared occultation measurements.

Implicit in the high accuracy of the ACCURATE mission is as well the requirement for accurate spectroscopic line parameters. Unfortunately, the accuracy of the line parameters presently available in the HITRAN database limits the accuracy of the demonstration measurements. For example, the CO_2 line intensities in the SWIR spectral region have reported errors in the range >10% and <20%. It is necessary to improve the HITRAN line parameters for the targeted absorption lines substantially in order for the ACCURATE mission to meet its accuracy goals (Harrison et al., 2011).

In summary, we conclude from this first experimental analysis that infrared laser occultation between LEO satellites (Kirchengast and Schweitzer, 2011) has a sound basis for monitoring CO_2 in the free atmosphere; other greenhouse gases such as methane, nitrous oxide and water vapour can be monitored in the same way. Further refined analysis of the Canary Islands campaign data is currently on-going, including for methane

and water vapour in addition to CO_2, as well as preparations for more advanced instrumentation. This will enable a more stringent demonstration as a next step."

In this context it is also a clear limition that the measurements do not permit to judge to what extent temporal changes can be detected with the proposed measurement system. This is a key limitation of the presented data as this is a mandatory aspect for the envisaged monitoring application. This needs at least to be mentioned in the manuscript.

We have mentioned this in a sentence of the updated "Discussion and Conclusion" (page 8 line 5):

"The static link between the Islands also does not demonstrate the scanning of the atmosphere as occurs between LEO satellites, and the experiment was too short to monitor trace gas variability over time. A detailed description of how the LEO system works is given by Kirchengast et al. (2010)."

However, any details of temporal changes of trace gas concentrations are not within the scope of this first demonstration paper, and other referenced sources (Larsen et al. (2009), Kirchengast et al. 2010, Kirchengast and Schweitzer (2011) and Proschek et al. (2011)) describe the actual LEO mission design, including detailed measurement mode and retrieval descriptions.

I also find the following statement given in Section 4 too strong (page 3312 line 8 and following): "For an ACCURATE-type mission, the sources of error will be smaller ...". I recommend to modify this as follows: "will likely be smaller ..." or "are supposed to be smaller ...".

As well as generally improving this aspect of the "Discussion and Conclusion", providing clear rationale on why errors of the ACCURATE mission are expected to be significantly smaller, the statement has been changed to "are expected to be significantly smaller" (page 8 line 16), to make it weaker than the firm "will be", as suggested.

Specific: Page 3310, line 7: Please explain CRDS.

"cavity ring-down spectrometer" has been added (page 6 line 19), thank you.

Figures 4-5 and A1-A3: The figures would highly benefit from adding (transmission spectra or Jacobians) showing seperately the target gas absorption (CH4, CO2) and the absorption features of major interfering gases (e.g., H2O).

Figures A1-A3 have been reproduced based on this suggestion, and are attached as supplements to this comment. We no longer have access to the program used previously to calculate these example transmission spectra (GATS Spectralcalc), so these were recalculated with the same forward model as was used in the fitting procedure, and the respective sentence in Appendix section A6 "Conditions for Calculated Spectra" was changed to reflect this (page 14 line 3). It now reads "The spectra shown in Appendix Figures A1 to A3 were calculated with the same forward model as was used in the fitting procedure".

Figures 4 and 5 are present specifically to show the difference between the observed spectrum and the fit, and so extra information would not be appropriate here. These regions can be found in Figures A1 and A2, though, to see the absorptions of the interfering gases.

REVIEW 2

The abstract contains a motivation for the experiment, instead of providing more information about the experiment itself. A description like in the first line of the conclusion for example would have been more informative here. Also a sentence about what is called the "new technique" or the "new method" would have been good here.

The first sentence referred to is: "We have successfully demonstrated a new technique to directly determine atmospheric greenhouse gas concentrations from SWIR absorption measurements over long path lengths, with relatively low power diode lasers (4 to 10 mW)." The abstract already contains all of this information, except the part relating

to low power lasers. Therefore this information has been added to the abstract (page 1 line 28). A sentence about the "new method" is also already present; the first sentence of the abstract explains this ("A new technique. . ."; page 1 line 22). However, to make it more clear we now write "this new infrared-laser occultation method" (page 1 line 26), instead of just "this new method".

The full abstract now reads as follows:

"A new technique for the satellite remote sensing of greenhouse gases in the atmosphere via the absorption of short-wave infrared laser signals transmitted between counter-rotating satellites in low Earth orbit has recently been proposed; this would enable the acquisition of a long-term stable, global set of altitude-resolved concentration measurements. We present the first ground-based experimental demonstration of this new infrared-laser occultation method, in which the atmospheric absorption of CO_2 near 2.1 μm was measured over a 144 km path length between two peaks in the Canary Islands (at an altitude of 2.4 km), using relatively low power diode lasers (4 to 10 mW). The retrieved CO_2 volume mixing ratio of 400 ppm ($\pm$15 ppm) is consistent within experimental uncertainty with simultaneously recorded in situ validation measurements. We conclude that the new method has a sound basis for monitoring CO_2 in the free atmosphere; other greenhouse gases such as methane, nitrous oxide and water vapour can be monitored in the same way."

At the end of the abstract and in the Conclusion it is written that the "new method" has a sound basis for monitoring carbon dioxide and other greenhouse gases. I agree with referee #1 that these are strong words. While I think that carbon dioxide is well covered, I feel that for other greenhouse gases there are still issues. It seems that for methane there were equipment problems or there was not enough time for the experiments and for water the spatial variability was to large. Therefore at most it could be said that based on the carbon dioxide experiments, other greenhouse gases are likely to be measured in the same way with a certain accuracy, too. The caveats for other greenhouse gas measurements are indicated.

Ok, the sentence section "has a sound basis for monitoring CO_2 and other greenhouse gases in the free atmosphere" has been changed to be restrictive to CO_2 in its direct meaning, and only to mention prospectively other greenhouse gases. It reads now "has a sound basis for monitoring CO_2 in the free atmosphere; other greenhouse gases such as methane, nitrous oxide and water vapour can be monitored in the same way". Note that we also revised the whole "Discussion and Conclusion" text carefully to better discuss limitations of this first demonstration (we cite this revised text as part of the Reviewer 1 response).

Was there no exploitable methane data from the measurements during night 7?

Unfortunately, those first data of night 7 were not exploitable towards retrievals.

In the conclusions I could not follow the statement that "for an ACCURATE type mission the sources of error will be smaller than in the demonstration." This statement should be better motivated.

This has been changed to "are expected to be significantly smaller" (page 8 line 16), as explained in the response to the first review.

The new full sentence is "The sources of error contributing to the value of $\pm$15 ppm are expected to be significantly smaller for an ACCURATE mission than in this least-cost demonstration."

Technical issues: 1. Introduction: it would be good to mention the experiments or instruments by name e.g. TCCON, IASI etc.

We carefully revised this part of the introduction and included all full names of the experiments or instruments (page 2 lines 12-26).

This section of the introduction now reads as follows:

"In situ measurements include numerous precise instruments on the ground, for example at Mauna Loa Observatory, Hawaii (Keeling et al., 1976; Komhyr et al., 1989); on

tall towers, for example in Europe for the CHIOTTO project (Continuous HIgh-precisiOn Tall Tower Observations of greenhouse gases; Vermeulen et al., 2011) as well as in the US (Bakwin et al., 1998); and in aircraft for the CARIBIC project (Civil Aircraft for the Regular Investigation of the atmosphere Based on an Instrument Container; Schuck et al., 2009), for calibration of the TCCON (Total Carbon Column Observing Network; Wunch et al., 2010) and as part of the NACP network (North American Carbon Program; Crevoisier et al., 2006).

Remote sensing of column CO2 is carried out from the ground using direct sunlight in the near-infrared in the TCCON (Wunch et al., 2010); recently from low Earth orbit (LEO) using reflected sunlight, for example by SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric ChartographY; Schneising et al., 2011) and GOSAT (Greenhouse gases Observing SATellite; Yoshida et al., 2011), and using thermal infrared emission by AIRS (Atmospheric InfraRed Sounder; Chahine et al., 2008) and IASI (Infrared Atmospheric Sounding Interferometer; Crevoisier et al., 2009)."

Experimental Is there a word missing or is the section called "Experimental"?

Yes, it was just called "Experimental". We have however changed it to "Experiment".

Fig. 1: The lat/lon indications are very rough! The actual lat/lon differences are less than 1° for both latitudes and longitudes.

These indicators are only present to denote the rough map area shown, so that someone could find the Canary's on a world map if they did not know where they were. However, in order to make it more clear, we now say explicitly in the caption "Illustration of the laser link... (map area 28° N to 29° N, 18° W to 16° W)".

Fig. 2: I could not find Label C-ARTEMIS beam collimator in the figure.

Thank you, this has been removed from the caption (was not part of our setup; can be considered part of OGS telescope so not worth mentioning as a dedicated unit).

OTHER CHANGES:

We have changed the final result and error from 400.1 ppm ($\pm$14.7 ppm) to 400 ppm ($\pm$15 ppm) to better reflect the level of accuracy observed, and the validation value and error from 386.7 ppm ($\pm$0.21 ppm) to 386.68 ppm ($\pm$0.21 ppm), to provide a consistent number of decimal places.

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Please also note the supplement to this comment:
http://www.atmos-meas-tech-discuss.net/5/C2065/2012/amtd-5-C2065-2012-supplement.zip

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Interactive comment on Atmos. Meas. Tech. Discuss., 5, 3303, 2012.