AMT Manuscript Title: Retrieval and validation of carbon dioxide, methane and water vapor for the Canary Islands IR-laser occultation experiment

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Response to Reviewer #4

We thank the Reviewer for his/her constructive and helpful comments. We carefully considered all comments and accounted for them in our paper as stated below.

The original comments of the Reviewer are cited in *italic font*, our response is put below each comment in standard font. The changes stated below are also **yellow-highlighted** in the Revised Manuscript.

General comments:

The manuscript is a thorough presentation of a small set of ground-based infrared-laser measurements of key greenhouse gases, and the necessary uncertain analysis in such measurements. Section 3 focuses on the data processing and the retrieval approach. While section 4 addresses the retrieval uncertainties. This approach gives a good understanding of the strengths and limitations of the observational technique.

Thank you for the positive general comment.

Specific comments:

- page 3, line 73 (in the AMTD online version page 11598 line 2): The statement is included without any further argumentation. What about the strong wind fields observed at the Tx and Rx site? Is the statement valid at all times and all latitudes? What about the changes in the volume mixing ratio of H2O during the first night of observations - as given in Figure 2?

Practically speaking, we could visually confirm that we were measuring above the top of the marine boundary layer (in those periods where we did the successful measurements reported on in this paper). It is true though that close to the Islands themselves there will be some boundary layer variability effects (e.g., indicated by wind changes) and also that the (free atmosphere) regime changed its moisture along the path of propagation. While we think we adequately discuss these aspects later in the paper we agree that one more qualifying sentence may be useful already here. We therefore added on page 11598 line 4: "We could visually confirm during the experiment that these free atmosphere and cloud-free conditions prevailed along the laser link; nevertheless we experienced variable meteorological conditions both near the Tx and Rx sites (e.g., changing wind conditions) and along the link (e.g., significant moisture changes), which we needed to take into account in the analysis and interpretation of results."

- page 7, line 217 (in the AMTD online version page 11603 line 29): The ECMWF resolution is given to be 14 km and 6 hours, leading to considerably variations in the volume mixing ratio of H_2O . Thus the coarse ECMWF input in the signal ray tracing ought to be more significant. The iterative retrieval seems to smear out some of these effects. Why is that? Scale sizes of H_2O above the boundary layer are often much smaller than the resolution of the ECMWF data.

The reason is that we can only get along-ray-integrated H_2O in the retrieval, so indeed any along-ray variations are no longer "visible" in the end result. That is, while the ray-tracing does a reasonably realistic job in capturing the along-ray variations (and this is useful to help accumulate a reasonable average), the end result is an along-ray average estimate; more cannot be obtained from this type of differential absorption technique. (In LEO-LEO geometry in space it is different: here the spherical geometry enables an Abelian transform, which can provide "local" profiles that we can attribute to the "tangent point region" of the ray paths.) In summary, as we discuss in the paper for H_2O , the relatively high uncertainty that we need to attribute to the H_2O volume mixing ratio results is indeed from the strong variability of H_2O .

- page 16, line 518 and line 536 (in the AMTD online version page 11616 line 19 and page 11617 line 7): The uncertainty due to water vapor is a limiting factor for the quality of the measurements of this greenhouse gas. So please expand a little on how 23 GHz measurements can mitigate the uncertainty estimates (see also your own remarks on page 19, line 643 (AMTD: page 11621 line 18), and onwards).

As we express on page 11617 lines 5-9, the 23 GHz water vapor measurements could be expected to observe (along-ray average) H_2O to 5 % accuracy, reducing the current conservative uncertainty of 50 % by an order of magnitude, and accordingly also the influence on the GHGs by about an order of magnitude (from order 1 % to order 0.1 %).

We added on page 11617 line 8 a reference "...23 GHz, as part of the LMIO method; Schweitzer et al. 2011b) would enable..." and we added a sentence in line 9: "This reduction of uncertainty in CO_2 and CH_4 by an order of magnitude (from order 1 % to order 0.1 %) would arise from the reduction of the H₂O uncertainty from 50 % to 5 %."

- page 21, line 709 (in the AMTD online version page 11624 line 3) + page 22, line 735 (in the AMTD online version page 11625 line 4): The statements about the ACCURATE/LMIO satellite mission concept, solely based on this ground-based proof-of-concept experiment, is too far fetched. Satellite occultation observations applying this technique are several orders more

complex than the presented measurements. Spatial scalability from this stationary ground-based experiment to space does not apply.

Yes, we are well aware of the different category of complexity to implement a full ACCURATE/LMIO mission. That is why in these statements we just carefully talk on "demonstrating the feasibility of...employing the differential transmission principle" and "...has a sound basis in this respect", "...is a vital step, among others, towards..." etc. We now further down-tuned the statement on page 11624 line 1 from "...was definitely successful to demonstrate..." We exercise this care in statements (and separately have a concise outlook to many next steps needed) exactly since we want to avoid any exaggerated claim that this IR-laser experiment would be demonstrating anything close to a complete LMIO mission.

Many thanks to Reviewer #4 again for his/her valuable comments that helped us to further improve our manuscript.