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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Latitude-time variations of atmospheric column-average dry air mole fractions of CO₂, CH₄ and N₂O" *by* R. Saito et al.

Anonymous Referee #2

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******** General comments ********

In this manuscript, the authors have simulated three tracers – CO2, CH4 and N2O – in the atmosphere using a global atmospheric transport model with chemistry (ACTM), and compared their simulated total column concentrations of the three tracers with measurements at a network of TCCON stations. Seasonal variations of CO2 are strongest in the lower troposphere, whereas those of N2O are strongest in the stratosphere. Since their model simulates the variations of both these tracers accurately – as evident from their model-TCCON comparison – they conclude that their transport model does not have major flaws in either the troposphere or the stratosphere. There-

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fore, they conclude that the significant TCCON-model mismatches seen for the third tracer, CH4, must stem from errors in its surface fluxes and oxidation of CH4 by the OH radical.

The authors' idea of separating transport errors from flux errors by using multiple tracers is innovative and interesting. My main concern, however, is that the authors have stopped short of drawing any quantitative conclusions. In its present form, the manuscript reads like a performance evaluation of the ACTM model, which in itself would be of interest to a limited audience. What would be far more useful to the community would be a method to estimate the fraction of a total-column model-TCCON mismatch coming from flux errors as opposed to the fraction coming from transport errors. For example, could the authors predict the percentage of TCCON-model mismatch in total column CH4 that could be attributed to transport model errors, which can perhaps be estimated at different layers by looking at N2O and CO2 measurements, as the authors seem to think? The authors could possibly use tracer measurements from aircrafts to estimate the performance of ACTM at different altitudes. In the revised manuscript, I would like to see some quantitative conclusions about the sources of a model-TCCON mismatch of a tracer, the methodology behind which could be used by other groups to quantify the performance of their own tracer transport models, with a view towards separating – quantitatively – flux-related uncertainties from transport-related ones.

The above is my only reason for recommending major revisions, since otherwise the manuscript is well-written and needs only a few minor revisions as detailed below.

******** Specific comments ********

1. Page 5682, line 9: The weaker seasonal cycles can also be a result of transport errors in the free troposphere (and not just the PBL or the stratosphere), which can be estimated by comparing simulated CO2 fields with aircraft measurements.

2. Page 5682, line 5: The mention of mid-IR total column retrievals seems irrelevant

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here.

3. Page 5683, lines 23-26: If I understand correctly, the “optimized” CO2 fluxes are only optimized for 2008, and then replicated across all years. Given the considerable interannual variability of the terrestrial CO2 flux, I am surprised that this method yields a reasonable match to TCCON total column CO2 across multiple years, especially at continental sites such as LEF and LAM. Does this mean that assimilating the TCCON total column CO2 in an actual multi-year inversion would add very little information and put very few additional constraints on the surface fluxes? I would like the authors to clarify this point, since this effectively means that inversions a la Chevallier et al, GRL 38, L24810 (2011) should not improve the quality of surface fluxes compared to inversions using only surface point measurements.

4. Page 5684, line 3: Is the interannual variation of OH included in the simulation? How are the OH fields generated? Since the interannual variation of OH is significant, I would like this information to be added to the manuscript.

5. Is the N2O flux scenario used in this simulation optimized against N2O measurements, or is it an inventory estimate? Please mention that in the manuscript.

6. Page 5685, line 14: What is the impact of using a wet-air pressure on equations 2 and 3, given that the water fraction of the total pressure has a seasonal cycle as well? Since ACTM also simulates the dry air mass – suggested by the first paragraph of the page – why is that not used to calculate the partial column ratios?

7. Figure 4: It seems that applying the averaging kernel decreases the N2O total column at all sites, which is consistent with the averaging kernel being higher at layers with lower N2O concentration, i.e., the stratosphere. However, at Wollongong the averaging kernel seems to increase the total column N2O mixing ratio. Why is that? Please clarify.

8. Page 5689, line 15: Can the seasonal biases at Sodankyla and Darwin be ex-

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plained?

9. Page 5690, lines 8-10: The authors seem to suggest that the inverted CO2 flux is influenced more by Park Falls than by Lamont. Is this because their inversion did not have any surface layer data at Lamont? Also, Lamont is in an area known to suffer from droughts in the summer, leading to a shallower trough in CO2 compared to what coarse-resolution models would predict. Could this be a factor behind the modeled overestimation of the seasonal cycle depth? I would like the authors to discuss/clarify this point in the revised manuscript, since the two sites (LEF and LAM) are more similar than different as far as measurements there go.

10. Page 5691, line 22: Please provide a citation for "age of air", e.g., Jones et al, JGR 106, 32295-32314 (2001), or Andrews et al, JGR 106, 10257-10274 (2001).

11. Figure 2: If tropopause dynamics is the main reason for the seasonal variation of total column N2O, then why does the N2O peak (dark red band over the equator in subfigures c and f) not follow the ITCZ? Also, why is the N2O peak over the equator more "flattened" in July than in January (total column N2O seems to fall off faster away from the equator in January compared to July)? Please add information about this in the manuscript.

12. Page 5692: The authors mention that most of the seasonal variation of total column N2O comes from tropopause dynamics. This does not explain, however, why the N2O seasonal cycle is so much higher at Park Falls than at Lauder, given that they're roughly the same distance away from the equator. Nor does it explain why the season cycles over these two places have the same phase, since one would expect them to be six months out of phase. Could the authors explain?

13. Figure 6: While on the topic of the N2O seasonal cycle, there are several factors that influence its amplitude and phase, and its relative contribution from the tropo/stratosphere: (a) the tropopause height, (b) the Brewer-Dobson circulation which injects stratospheric air into the troposphere, (c) the seasonal variation in the averag-

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ing kernel owing to the variation in the solar zenith angle, (d) the change in photolysis rate owing to change in insolation, and (e) change in agricultural emission of N2O over the landmass. I would like the authors to spend some time explaining the reasons behind their observed seasonal variation of total column N2O, since "getting it right" could be due to getting some – but not all – of these factors right. Since the authors make a strong point about ACTM accurately simulating the total column N2O, I would like some discussion on which of the aforementioned factors are accurately simulated by their model and which are not, and the relative importance of the different factors.

14. Figure 3: At LAM, LEF and BRE, TCCON total column CO2 seem to be higher than ACTM simulated columns in 2010, but not in 2009. Where do these mismatches come from?

15. Figure 4: Why does the averaging kernel make a greater impact on the N2O total column over Darwin compared to other sites?

******** Technical corrections ********

1. Page 5684, line 17: "time series at 15TCCON" -> "time series at 15 TCCON"

2. Page 5688, line 14: "daily variability" -> "seasonal variability", perhaps?

3. Page 5691, lines 18-20: Expand UT, LS and STE the first time these abbreviations are used.

4. Page 5691, line 20: "conservative quantity" – perhaps the authors mean "conserved quantity"?

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 5679, 2012.