Reply to Anonymous Referee #2 (Received and published: 14 April 2012)

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

In this manuscript, the authors have simulated three tracers – CO_2, CH_4 and N_2O – 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 CO_2 are strongest in the lower troposphere, whereas those of N_2O 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. Therefore, they conclude that the significant TCCON-model mismatches seen for the third tracer, CH_4, must stem from errors in its surface fluxes and oxidation of CH_4 by the OH radical.

Thank you very much for providing critical and useful comments and suggestions.

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 CH_4 that could be attributed to transport model errors, which can perhaps be estimated at different layers by looking at N_2O and CO_2 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.

We partially agree with you on quantitative conclusions reached in this manuscript. We are still in a learning phase how to treat the total column values in model-observation comparison. As shown here, sometimes the stratosphere contributes significantly to the total column seasonal variations. There are several issues, in our opinion, before we go on to talk about what the model-observation differences imply for surface fluxes. For example, what differences we would see between the retrievals and model simulations, if the a priori vertical profiles from the same chemistry-transport model (CTM) were used in TCCON retrievals. How much it is to our benefit, if we cannot separate/understand the tropospheric and tropospheric columns first – this probably is the first study attempting to bring up this issue. The layer above the tropopause constitutes ~10% of air mass; any variability of 1% or more will disturb the signals we have in the total column measurement for surface flux inversions (in addition there is role of averaging kernel profiles). To add further to this, we already know the vertical and horizontal gradients in CTM simulations differ by ~1% in the upper troposphere region and increases with altitudes in the stratosphere (TransCom-CH_4 study; Patra et al., 2011). The state-of-the-art CTMs handles troposphere better than the stratosphere. Our study thus recommends serious efforts are needed to

overcome these limitations by both the measurement and modeling communities.

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.
Such comparisons for ACTM simulations have already been done in Patra et al. (ACP, 2011) and Niwa et al. (ACP, 2011) using CONTRAIL measurement of CO_2 vertical profiles over a variety of sites, and we found ACTM performance for chemical tracer simulations is generally satisfactory in the free troposphere. Thus, the transport errors in the PBL and the stratosphere are stronger than that in the free troposphere. Niwa et al., ACP (2011) is added to the reference.

2. Page 5682, line 5: The mention of mid-IR total column retrievals seems irrelevant here.
Since this data stream is very similar to results discussed here, we mentioned this. We have deleted following your suggestion.

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.
We do not think that Chevallier et al. have shown the estimate fluxes in details at continental scales in the paper. They have discussed mainly the northern hemisphere mean seasonal cycle.
As you note later, the ACTM total columns at LAM, LEF and BRE are about 1-2 ppm higher than the TCCON values. A model-observation mismatch of 1-2 ppm is considered large signal in total column CO_2 variability in space and time. A measure of goodness of model-observation agreement is now included following the first of the general comments of Reviewer#1 (RSD in Table 1). This means, "the XCO_2 distributions at TCCON measurement sites contain significant new information on the local-regional surface fluxes" (as stated in p.5690, line15). Our next aim will be to use the TCCON data in source/sink inversion.

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.
The tropospheric OH fields are taken from Spivakovsky et al. (2000), and global total is scaled to reproduce CH_3CCl_3 growth rates in the 1990s and 2000s (Patra et al., 2011b). Our simulations of CH_3CCl_3 in TransCom-CH_4 experiment suggest that OH

IAV is very small, if we assume the CH3CCl3 surface are decreasing exponentially in the decade of 2000s (Patra et al., ACP, 2011b).

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.
The soil emissions from EDGAR2 are scaled by factor of 1.2 for producing the observed N_2O growth rates, approximately. A sentence is added in the main text.

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?
The impact of the water vapor pressure is small, mostly less than a few tenths of a ppm for CO_2, less than a ppb for CH_4 and a few tenths of a ppb for N_2O. We have used Ps and Pt from ACTM surface dry pressure and tropopause height.

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.
Which way the averaging kernels (AVKs) change the total columns depends on the location of tropopause. For instance, we can see at Lamont the ACTM values after applying AVK also increase compared to those without treating AVK during the summer months. When the tropopause is lower in the winter, the ACTM XN_2O are corrected downwards. It worthwhile to reiterate here that we add offset of 3.2 ppb to the ACTM XN_2O (blue dots) after applying the AVK to account for the overall reduction in ACTM results (brown line). Such an offset is not required for the ACTM N2O simulations for comparison with surface sites.

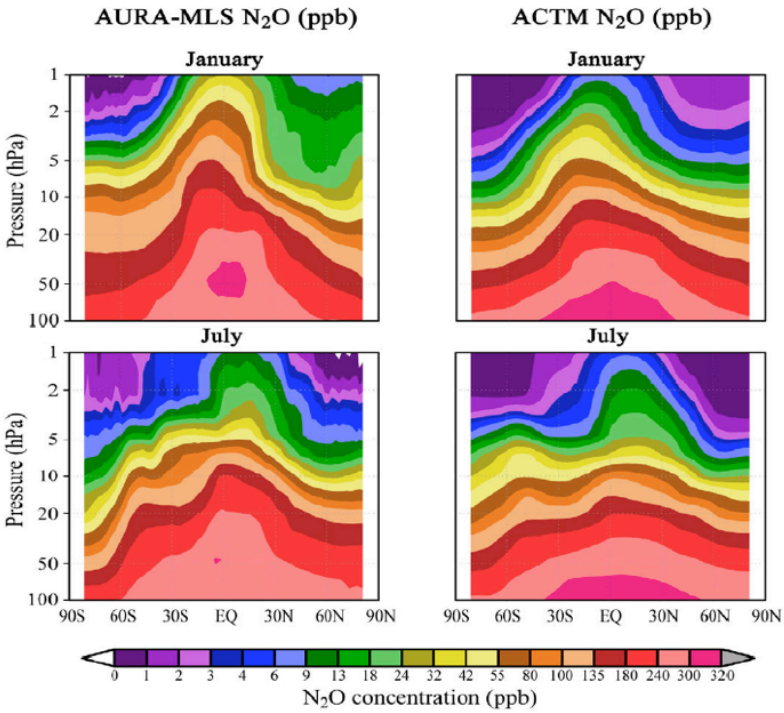
8. Page 5689, line 15: Can the seasonal biases at Sodankyla and Darwin be explained?
We have not stressed on the possible reasons of the large bias in Sodankyla (also Eureka), because the period of time series are too short less than 1 year to explain the seasonal cycle. For the model-observation differences seen at Darwin, we discuss possible reasons in section 3.3.

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.
Our analysis does not suggest that the TCCON measurements at LEF and LAM are more similar than different (Fig. S5). The seasonal cycle amplitudes are much different at the two sites, ~9 ppm at LEF and 6 ppm at LAM. The model clearly overestimates the seasonal amplitude at LAM by about 2 ppm, which we attribute to the coarse inverse model resolution (only 4 regions over the temperate North America). Unfortunately, in our inversion setup does not contain many measurements

from the southern USA. Not much could be said here, but using measurements from LAM could be useful for the inverse models to constrain CO_2 flux from the continental USA (this is what we have mentioned by "Thus, the XCO_2 distributions at TCCON measurement sites contain significant new information on the local-regional surface fluxes").
We add this sentence here "Also note that the continental CO2 measurement sites in the 4 temperate North America regions are located north of 40^oN"

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).
To the best of our knowledge the age of air concept was not introduced in these papers. The age of air is a relatively well known concept in the research community of long-lived atmospheric species (first by Kida (1983) in a model world and then Bischof et al. (1985) derived using measured CO_2 in stratosphere). Here we cite a paper, which discusses in detailed how the 'age' of air can be used for constructing stratospheric profiles/column of the chemical tracers (Saito et al., 2011), likewise the one for HF tracer (Washenfelder et al., 2003). Saito et al. have shown how to prepare fairly accurate 3-dimensional age distributions at monthly intervals by combining ACTM simulations and balloon-borne measurements of SF_6, and are freely available from the lead authors.

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.
It is somewhat following ITCZ, and also the location of the tropical upwelling branch of Brewer-Dobson circulation (as seen at Darwin site). When the ITCZ/upwelling branch is in the SH (NH) during austral (boreal) summer, high N_2O columns are measured or simulated. The flatness or sharpness of column N_2O decrease depends on the N_2O distributions around the tropopause height. These features can be further clear from the plots below:

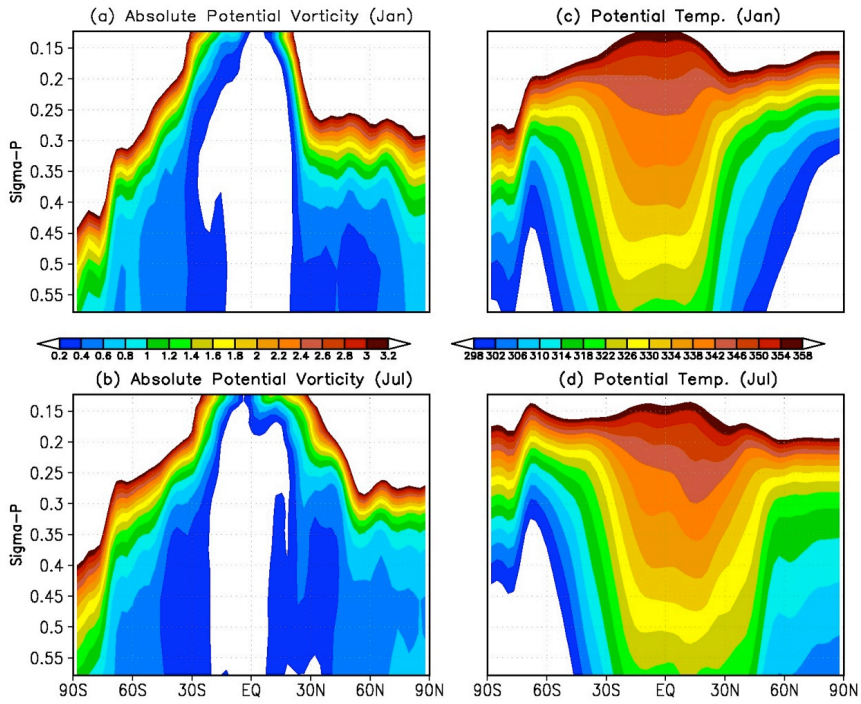


Modified from Ishijima et al. (2010, doi:10.1029/2009JD013322)
Figure (2) Monthly-zonal mean N_2O concentration observed by Aura-MLS (left column) and simulated by ACTM (right column) in January and July of 2006.

Here we add "During January the tropical upwelling branch of the Brewer-Dobson circulation is narrow and N_2O-rich air is transported deep in to the stratosphere (240 ppb isopleths reach beyond 10 mb), while during July similar N_2O concentrations reside below 10 mb but over a wider tropical latitudes. Thus the peak in N2O column over the equator is more flattened in July than in January."

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?
Good question. The seasonality of gravity wave generation in the troposphere and their breaking in the stratosphere are higher in the northern hemisphere than in the southern hemisphere. Thus the tropopause layer is less well defined in the NH than in the SH, allowing greater exchange of air across the tropopause in the NH.

The figure below shows seasonal variations in latitude-sigma (pressure) distributions of absolute potential vorticity (in PVU; left column) and potential temperature (in ^oK; right column). While both the potential vorticity and temperature do not show significant differences in meridional and vertical gradients between January (top row) and July (bottom row) in the SH, large differences in meridional (e.g., the location of the surf zone) and vertical gradients are seen for both these parameters. When the potential temperature contours are closely spaced around the tropopause (commonly defined as the 2 PVU line), the stratosphere-troposphere exchange (STE) will be weaker, as is the case of January, compared to the time of July in the NH.



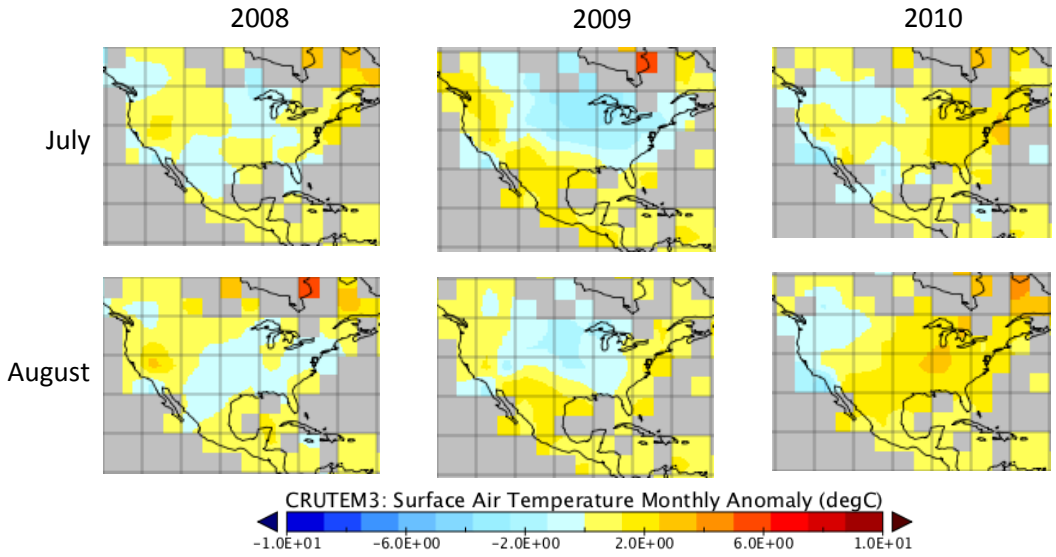
13. Figure 6: While on the topic of the N2O seasonal cycle, there are several factors that influence its amplitude and phase, and its relative contribution from the tropo/stratosphere: (a) the tropopause height, (b) the Brewer-Dobson circulation which injects stratospheric air into the troposphere, (c)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

It has been discussed that the N_2O total column, given the TCCON averaging kernels, contains weak signal from surface fluxes (including emissions from agricultural activities). The N_2O simulations by ACTM have been compared with Microwave Limb Sounder measurement in the stratosphere (Ishijima et al., 2010), and CONTRAIL (Ishijima et al., 2010) or CARIBIC (Patra et al., ACP, 2011) measurements in the upper troposphere and lower stratosphere. These provided check on the factors (a) to (d) you mentioned on realistic N_2O simulations by ACTM.

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?

As you have noted earlier, the terrestrial and oceanic CO_2 fluxes are used from a cyclostationary inversion for the year 2008. Thus such mismatches are caused by CO_2 flux error. Also it must be pointed out here that EDGAR4.0 emissions due to fossil fuel burning cover the period up to 2005, and global total emissions are extrapolated using the CDIAC for the years 2006 onwards. The CO_2 fluxes are not optimsed for simulating CO_2 growth rates at regional scale.

Figure 4 (panel g & j) suggests that the seasonal cycle minima were shallower for LEF and LAM in 2010 compared to 2009. The difference between 2009 and 2010 at BRE is not clear, except that there are differences in data coverage. We added these sentences here "The TCCON XCO_2 are higher by about 2 ppm than ACTM XCO_2 in 2010 at both LAM and LEF, while agreement between TCCON and ACTM were good in 2009. This mismatch in 2010 is produced by shallower seasonal cycle minimum for XCO_2 at the two sites (ref. Fig. 4). The summer high temperature anomaly over the North America in 2010 (Figure below based on CRU surface air temperature anomaly; source: Hadley Centre: http://hadobs.metoffice.com/crutem3/) may have led to weaker terrestrial ecosystem uptake in 2010. Further analysis is needed for quantification of the reduction in net carbon uptake."



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

Actually, the impact of averaging kernel is minimal at Darwin – seen as the difference between the brown line and light blue symbols (right column). Because of the tropical upwelling in the stratosphere, the N_2O concentration decrease rate with altitude is relative lower than in the extratropics or mid/high-latitudes in the lower stratosphere.

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

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

2. Page 5688, line 14: "daily variability" -> "seasonal variability", perhaps?
Both daily and seasonal - the text has been changed accordingly.

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

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