Review comments

Reviewer 1

We thank the reviewer for his/her very thoughtful and constructive review. Please find our responses to all questions and comments below.

Page 2318, L17: The south to north gradient is not clear from Fig. 2. You may want to revise the color scale to make this feature evident in Fig. 2.

We have now revised the colour scale in Fig. 2, however, it is not possible to maintain a linear colour scale and make the gradient much clearer owing to the large range of values.

Page 2319, L4-6: Is this classification of lower and upper stratosphere also valid for the tropics where tropopause could be located above the 380 K isentrope. I would suggest adding a black line showing the tropopause height in Fig. 2 so that readers can better recognize lower and upper stratospheres.

We have now included a dotted black line indicating the position of the annual mean tropopause.

Page 2319, L16: Reichler et al. (2003) also suggested that average lapse rate between the level (at which lapse rate becomes less than 2 K km-1) and all higher levels within 2 Km should not exceed 2 K km-1. This condition removes the probability of false tropopause detection. Did you check this condition as well? If yes, please add this information to the manuscript.

Yes, we included this condition as well, i.e. following the method of Reichler et al. (2003). We have now included this extra information in section 3.1.1, second paragraph.

Page 2320, L13-14: Why is N2O sink larger in ACTMt42l32 while it was stated in section 2.1 (Page 2313, L16-19) that loss rates in all models are scaled such that global annual total loss of N2O was about 12.5 Tg N?

It was suggested in the experiment protocol that the global annual loss of N2O should be approximately 12.5 TgN, while most models were able to achieve this, it was not the case in ACTMt42l32. The reason for the larger sink is most likely due to a combination of the strength of the photolysis rate and how it is distributed with altitude. To achieve the recommended total loss requires tuning this.

Page 2320, L18-19: Is N2O lifetime calculated as (column 4/ column 5) of Table 7? If yes, N2O lifetime in TOMCAT comes around 108 years. In addition, the lifetimes shown in Table 7 and Fig. 3 are different (e.g. blue cross in Fig. 3 says lifetime of N2O is less than 90 y while Table 7 says it is 92 y). Please check these calculations again and accordingly revise the discussion.

We apologise for this mistake. We had updated our calculation in Table 7 but had forgotten to update Fig. 3. We have now corrected Fig. 3 for the updated values and also corrected the mistake for TOMCAT in Table 7.

Page 2325, L23-24, Why does model show such a larger amplitude compared to observations at MLO?

The models all show a much larger seasonal amplitude at MLO than is observed. This is owing to an overestimate of the influence of stratosphere to troposphere transport (STT) at this site and to the unrealistic later summer maximum in soil emissions (i.e. in OCN) in the northern mid latitudes. The minimum in the modeled seasonal cycle, i.e. circa May, is consistent with modeled STT, in which stratospheric air, depleted in N2O with respect to the troposphere, is transported across the tropopause with a maximum occurring in winter (December to February, in the northern hemisphere) and approximately a 3-month delay for transport to the lower troposphere. When the BWM soil emissions are used instead of OCN (BWM has no seasonality) the seasonal cycle is still overestimated by the models at MLO, albeit to a lesser extent as with OCN. We have now added this to section 3.3.1.1, end of the first paragraph.

Page 2328, L13-14: Is not this the case for PFA and ULB too? Can you please elaborate how did you conclude that modeled STT influence is stronger only at Hawaii?

This is related to the answer to the above question. Since the amplitude of the modelled seasonal cycle at MLO is much larger than observed, and considering that the seasonal cycle in the model is strongly determined by STT, this together indicates an overestimate of the importance of STT at MLO (and similarly at HAA, which is also in Hawaii) in the models.

Section 3.3.2: It appears that the emission scenario BWMN04 leads to best agreement between the models and the observations. This information may be included in the abstract. Given this, please justify your choice of OCNPIC as control simulation over BWMN04. How the results presented in the previous section would have differed if BWMN04 were used as the control simulation?

We chose OCNPIC as the control scenario as at the outset we thought that the ecosystem model, OCN, would better reproduce the real N2O emissions from soils, considering that this model is driven by climate data and resolves the emissions seasonally. Had we instead chosen to use BWM for soil emissions, the result that would have changed in Section 3.3.1, is that we would have seen an improved agreement in the phases of the model and observed seasonal cycles in the northern mid to high latitudes. It would have also improved the fit to the seasonal cycle at MLO somewhat, owing to the absence of the late summer peak in emissions in the northern mid-latitudes, which also influences the modeled mixing ratio at MLO. Moreover, we could not have known this before having done the model runs and analysis.

Reviewer 2

We thank the reviewer for his/her very thoughtful and constructive review. Please find our responses to all questions and comments below.

General comments

This study investigates the influence of surface emissions, tropospheric transport, and transport from stratosphere to troposphere (STT) on the variability of atmospheric N2O concentrations through observations (NO2, SF6, CFC-12) and their equivalents from 6 different transport models and two model variants. All the models underestimate the inter-hemispheric (IH) (i.e., south to north) gradient of N2O concentrations, while models that have provided results for SF6 reasonably capture this gradient. Focusing on the seasonality and the inter-annual variabilities of the studied species, the authors show that the surface emissions and/or the STT in the Northern Hemisphere (NH) are the causes of the underestimate of the observed inter-hemispheric gradient by the models. Indeed, the authors show that the seasonality of CFC-12 (which has emissions reasonably known in NH and has its sink in the stratosphere) is well captured by the models that provided results on CCF-12, but the STT seems to be more vigorous in the models. Regarding the Southern Hemisphere (SH), all the models fail to simulate the seasonality of both N2O and CFC-12 concentrations. The authors conclude that the STT is not well reproduced by the models. Interestingly, the authors point out the potential deficiency in the Brewer-Dobson model in explaining the seasonality of STT in SH based on the observations. Most of these results are consistent with some recent studies reported in literature. The study clearly shows that there is still a room for such work since as inherent in the model intercomparison exercise, the identified issues of each of the studied models cannot be investigated in details. Hence, as a future work, I suggest to the authors to focus on each of these models to quantify the contribution of each of the identified issues (when possible). The authors have fully considered the main comments of my first review relevant for ACPD. The paper is now clear and the results are clearly explained. Hence, I recommend it to be published in ACP after considering the few minor comments reported below:

Specific comments
Page 2311, line 5: *recent studies instead of* ***a*** *recent studies?*

We have corrected this.

Page 2311, line 20: *... spatial and temporal variabilities?*

We consider that "variability" is correct and thus have not changed this.

Page 2316, line 2: *... forward? I understand what the authors mean, but it seems for me that this word does not add anything here. If they want to use this word, they need to explain it*

We have now removed "forward" and written the following: "Six models and two of their variants participated in the inter-comparison of modelled N_2O mixing ratios.."

Page 2316, line 11: *You say that the models LMDZ4 and TOMCAT provide concentrations relevant for the closest model time-step to the observations. Since the* *temporal resolution of these two models is not reported, it is hard to appreciate how far the model data are from the observations in time. Please, clarify*

We have now included the model timesteps for LMDZ4 and TOMCAT at p2316, l12.

***Page 2317, line 5**: GC-MS. Please define the acronym MS*

We have now defined this on line 5.

***Page 2317, lines 12-15**: The computed mean biases (i.e., calibration offsets) are subtracted to the observations of the relevant sites? Please clarify*

We forgot to mention that the calculated offsets were added to the observations for the model-observation comparison. This has now been clarified on p2317, l15.

***Page 2318, lines 12-13**: 'This is a particularly simplification for species such N2O and CFC-12, which have a source to the troposphere and stratosphere sink". The sentence is not clear for me. Please clarify*

What is meant by "This is a particularly useful simplification for species such as N_2O and CFC-12, which have a source to the troposphere and stratospheric sink" is that the N2O (or CFC-12) budget can be simplified for the troposphere/stratosphere by considering only the source to/loss from the troposphere/stratosphere and the flux across the 380K isentrope. We have tried to make this clearer in the following formulation:

"This is a particularly useful simplification when considering the budgets of species such as N_2O and CFC-12, which have a source in the troposphere and sink in the stratosphere"

***Page 2319, lines 25-28**: Since all models use the same prior fluxes (OCNPIC), differences in the modelled growth rates are due directly to differences in the net cross-tropopause N2O flux, which depend (not **s** here) on the upward and downward mass fluxes and the above and below tropopause N2O mixing ratios. The authors can add that these differences are linked to the meteorology used in each model and also the vertical definition of the models.*

We have changed this to the following:

"Since all models use the same prior fluxes (OCNPIC), differences in the modelled growth rates are due directly to differences in the net cross-tropopause N_2O flux, which depend on the upward and downward mass fluxes and on the above and below tropopause N_2O mixing ratios; factors that are determined by the meteorological data used as well as on the vertical definition of the models."

***Page 2321, lines 5-8**: The authors should put these acronyms on Figure A2. This help to easily follow their demonstration in the text.*

We have added these site acronyms to Fig. A2.

Page 2321, line 17 *and elsewhere when relevant: The authors should fix the use of CTM or ACTM.*

We have corrected this now and use “CTM” throughout the manuscript.

Page 2329, line 17*: .. to* ***a*** *lack …*

We have corrected this.

Tables and Figures

Table *7: The period of study is 2006-2009 instead 2007-2009 as specified in the text? Please clarify*

This was a mistake in the caption of Table 7. It should be 2006 – 2009. This has been corrected.

Figure 1*: As already mentioned above, please fix the use of CTM or ACTM*

This has been corrected to “CTM”.

Figure A2*: Legend. It is the map that shows the locations of the observational sites. Also, you should put the acronyms of the sites*

We have now added the site acronyms.

Figure A3*: You state that you subtract the mean mixing ratio (model/obs). Are mean values computed at the global scale? Please clarify.*

Yes, we subtracted the global mean. This is now stated in the caption.