

Reply to the Editor's comments

We thank the Editor for providing useful comments on this manuscript. We firstly address the Editor's comments regarding modifications to the manuscript in response to comments from Reviewer 2 (R2) and then address the Editor's own comments.

Editor's comments regarding to modifications to the manuscript is response to R2's comments

Comment 1:

Overall I see that you might overcome some of the criticism being more explicit what you actually want to achieve with the presented analysis. It seems that you aim to conduct for the first time such a detailed multi-model analysis of global O₃ dry deposition, without aiming to also already at this stage analyze in quite detail the sources of some of the main discrepancies/differences between the models and between the models and measurements.

Author's reply

We have now more explicitly stated the aims of this study in the introduction (Introduction, paragraph 7) to emphasize that we are conducting an initial assessment of dry deposition in global scale models. We aim to identify key sources of difference between models and to highlight what out puts are required in future model intercomparison studies for a more detailed assessment of dry deposition. However, at this stage we do not seek to identify in detail the reasons for these differences.

Comment 2:

Editor's comment regarding the author's reply to R2's comment 1

"I would suggest you to specifically add here "long-term average O₃ dry deposition fluxes" and then also indicating what you mean with long-term (e.g., a full seasonal cycle where you expect that the model should at least resolve the large contrasts between summer and winter)."

Author's reply:

We have now modified the text as suggested by the Editor (Methods, paragraph 6).

Comment 3:

The Editor (in agreement with R2) strongly recommended that we include a comparison of the model data to ozone dry deposition velocity data and surface ozone data from CASTNET data.

Author's reply:

We have included a comparison between the CASTNET data and the modelled data. We have described the data that we used and how we processed it in the Methods section (paragraph 8). The comparison between the model data and the CASTNET data is plotted in Figure 11 and discussed in Section 4.4.

Comment 4:

Editor's comments regarding the author's reply to Reviewer 2, comment 7 regarding the effect of chemistry on dry deposition fluxes.

“This is not completely correct. There has already been some studies conducted with a global chemistry-climate modelling system including an explicit canopy exchange model system to considers the role of chemical reactions in O₃ deposition (and NO_x, BVOC) bi-directional exchange fluxes (Ganzeveld et al., JGR, 2002 and 2010). These studies did not directly discuss the role of chemistry in O₃ dry deposition fluxes but using a model that calculates in-canopy chemistry implies that, for example, the role of chemistry in explaining some of the non-stomatal destruction of O₃ besides the essential stomatal component, has been considered. On the other hand, the statement you added seems to sufficiently express the potential relevance of this feature but also that it is not considered in the presented study”.

Author's reply:

We thank the Editor for this clarification and we acknowledge that there are global models that do include in canopy chemistry schemes. We have modified our statement at Section 4.3 (paragraph 6) to emphasize that the models we used in this study do not include canopy chemistry.

Comment 5:

Editor's comments regarding our initial use of normalised O₃ dry deposition fluxes rather than O₃ dry deposition velocities (R2 comment 12).

“I would therefore suggest to also state this explicitly in the explanation of these results, e.g., “In Figures 1 (df), 2 (eh), 3, 4 (cd), 5 (cd), 6(b, e, h), 7 (b, e) and 8 (b, e) normalised O₃ dry deposition fluxes have been used to infer the deposition velocity by dividing this flux by this 30 ppbv mixing ratio”

“In showing the dry deposition velocities I think it is essential to mention explicitly, e.g. in the figure captions that you are showing the monthly mean dry deposition velocities. In addition, where you introduce this in the text for the first time I would suggest to add a statement that the monthly mean dry deposition velocities reflect generally the mean of generally large daytime V_d, especially for dense forest sites with high radiation and wind speed conditions, and generally small nocturnal V_d's also due to limited surface uptake and suppressed turbulent mixing.”

Author's reply:

Following comments from the reviewers we have changed our analysis and now present O₃ dry deposition velocities rather than normalised O₃ dry deposition fluxes.

We have now included text in the figure captions (Figs, 1, 3, 6, 7, 8, 9, 10 and 11) as suggested by the Editor to emphasize that were are comparing monthly mean O₃ dry deposition velocities fluxes and surface O₃ in this study.

We have also clarified that the monthly mean deposition velocities may result from large day time O₃ dry deposition and small night time O₃ dry deposition, as suggested by the Editor.

Editor's comments

Comment 6 – Part 1:

Comparing actually the dry deposition fluxes at a monthly resolution implies that your analysis show the simulation of seasonal cycles in dry deposition and which, for gases such as O₃ and SO₂ with a strong leaf uptake component, are sensitive to the representation of the seasonal cycle in LAI (e.g., Ganzeveld et al., JGR, 1998). Thus I guess it would be worthwhile to at least indicate in the Table if the model calculations use a seasonally resolved LAI/biomass estimate or that they simply use fixed LAI (for each PFT??), which used to be quite common until not so long ago (surprisingly recognizing the availability of remote sensing LAI products).

Comment 6 – Part 2:

Section 3.2: “This peak is driven by higher surface O₃ and possibly LAI in this region, as the deposition velocities are fairly evenly distributed between 30N--30S”. This expresses that there is not a clear understanding what explains the differences in dry deposition. This statement could be partly corroborated by indeed analyzing the LAI fields from the models. I guess that this should be standard output fields of the model. O₃ dry deposition indeed depends to some extent on LAI but it is also stomatal uptake that plays a key role (as long as this is also indeed considered in the models dry deposition algorithms). Stating this I realize that this is actually essential information that should be potentially added to Table 1. Most of the models use the original Wesely scheme with “fixed” vegetation uptake rates but some of the models might use an explicitly calculated stomatal resistance (e.g. provided by the land surface scheme of the meteorological driver model) and that is used to calculate vegetation uptake (I know that the TM5-JRC model should use that feature) and which could then to a large extent explain relative high uptake in tropical regions due to high radiation regimes.

Author's reply:

We have included a statement in the supplementary material indicating that the models use either satellite derived LAI, an LAI data set or LAI that is calculated as a function of ecosystem type. We have also included a statement that stomatal uptake plays a key role in determining the seasonal cycle in O₃ dry deposition flux (Section 23.2, paragraph 3) as suggested by the Editor. The HTAP study did not include any LAI output that could be compare with the O₃ dry deposition fluxes. Therefore, at this stage we can only suggest that LAI (and stomatal conductance) are driving seasonality in O₃ dry deposition as the monthly mean data do not support a more detailed analysis. We recognize the importance of a more detailed comparison and analysis of O₃ dry deposition and LAI (and stomatal conductance) and we hope that this study provides a strong argument for producing more detailed deposition diagnostics in future studies. (In fact, we have already requested this information for HTAP Phase 2 and for the ongoing CCMI model intercomparison).

Comment 7:

In the introduction you state that “This uncertainty arises from the complexity and heterogeneity in dry deposition processes which depend on both meteorological conditions near the surface and the characteristics of the surface.” I would also definitely add here the lack of observations of long-term dry deposition fluxes for many surface types including the oceans (although there is now a very nice dataset), desert areas, etc.

Author's reply:

We have modified the text in the Introduction (paragraph 5) to indicate that uncertainty in modelled dry deposition also arises from a lack of measurement data. We fully agree with this statement and emphasize the importance of measurement data, especially long term data sets elsewhere in this manuscript.

Comment 8:

“Calculating dry deposition velocity does not account for second order variation in the dry deposition flux which might arise, for example, from the feedback associated with the decrease in deposition velocity as O₃ is removed from the atmosphere.”

I am reading this in a way that you want to express that more deposition results in small concentrations which would result in a decrease in the dry deposition flux (and not dry deposition velocity as stated above). But this should already be expressed by using the monthly mean dry deposition flux isn't it?

Author's reply:

This is correct. Our analysis does not take into account that greater deposition flux reduces surface O₃ and in turn reduces deposition flux. The Editor is also correct in that this feed back has little effect on the monthly mean O₃ dry deposition fluxes used in this study. We have altered the wording to try and make this clearer.

Comment 9:

Reading through the section on the analysis of the LCC related O₃ dry deposition fluxes I am getting confused. You state that “The land cover schemes from individual models were not available for this study, so we apply two common schemes to all models”. It reads to me that you used a global land cover map and then determined for that land cover map and the resembling location in the model simulated O₃ deposition flux data that should be representative for that land cover type as analyzed. But so it might be that the actual land cover distribution in the model itself, and which has been used to calculate the dry deposition flux, might be very different from the map that you applied. Or is this a misunderstanding??

Author's reply:

To partition dry deposition fluxes to land cover class we used two global land cover maps to identify the location of different land cover classes. As the land cover data sets used in the models were not available we used what was available. We acknowledge that there are likely to be discrepancies between the land cover distributions in the data sets we used and those used in the individual models. We have altered the wording to make this clearer

However, we used fairly broad land cover classes that were in many cases similar to those used by the models and the locations of land cover classes are broadly known at 3x3 resolution. In order to account for some of this variation we analyzed fluxes at grid cells with 100% coverage (according to our land cover data set) as there are less likely to be discrepancies in these areas where the land cover is relatively uniform over a wide area.

Comment 10:

Section 4.2: “Improved characterization of deposition velocities over the ocean, which has been the focus of studies by Ganzeveld et al. (2009) and Helmig et al. (2012), would therefore make a substantial contribution to reducing the uncertainty in total global O₃ dry deposition”.

Author's reply:

We have modified this sentence to include the additional references (Ganzeveld et al. (2009) and Helmig et al. (2012)) as suggested by the Editor.

Comment 11:

“It is important to include a well constrained O₃ dry deposition velocity and global area for tropical forests as day time observations of between 3.840 cm s (Rummel et al., 2007) suggest that they are an effective O₃ sink.” I didn’t check myself the Rummel et al. statement but here I am pretty sure that you misinterpreted the units. Rather than having here 40 cm s⁻¹ it should be 40 mm s⁻¹ (and thus 4 cm s⁻¹). The typical maximum O₃ dry deposition velocity for tropical forest is around 2 cm s⁻¹ and this is already really high and mainly due to very efficient stomatal uptake.

Author's reply:

We thank for Editor for noticing this. We have now replaced this value with the more up to date figure of 2.3 cm s⁻¹ from Rummel 2007.

Comment 12:

Section 4.3: “..and in addition to that, more coherent representation of land cover and LAI across the models would contribute to a better representation of dry deposition in them”.

Here you should definitely add the explicit consideration of the role of stomatal uptake (as a function of radiation, moisture and other drivers) since this term is to a large extent determining the seasonal cycle in V_d.

Author's reply:

We have added a statement in Section 4.3 (paragraph 4) as suggested by the Editor.

Comment 13:

Section 5.3: “There was also less variation in O₃ dry deposition fluxes across the model ensemble at the Malaysian Borneo sites compared with the Amazonian sites possibly due to the contribution of water to the average deposition flux at the corresponding grid cell”.

This statement is really raising a lot of questions regarding the remark about the contribution of water. I initially thought you wanted to indicate here something about the role of soil moisture but there realized that you are referring to the fact that the grid box results used for this comparison contains a substantial fraction of ocean surface for the Borneo evaluation?

Author's reply:

The Editor is correct in that we are referring to the fact the relevant grid cell has a substantial fraction of it's area covered by water. We have modified our text here (Section 5.3, para) to clarify this statement.

Comment 14:

Conclusions: “While global scale O₃ dry deposition has not previously been reported at this level of detail, we recommend that future model comparisons request these additional flux diagnostics to allow deposition processes to be tested more thoroughly. In this study we make the first assessment of the multi-model simulations results on O₃ dry deposition fluxes against observations.”

I fully support this recommendation also since in your study, the lack of this extra diagnostics results in that you can at some point only guess what explains some of the differences among the models and between the models and the observations. But I also think this is really an overstatement. Yes, your study is unique in that it shows for the first time such a multi-model evaluation of global O₃ dry deposition but there have been previous studies, that you also list in

your paper (e.g., EMEP evaluations, Mike Sanderson's work, my papers on deposition and canopy processes) where quite detailed analysis on the deposition process, its drivers and evaluation by comparison of simulated and observed fluxes have been made. (comments that I already noted reading the initial version of submitted manuscript).

Author's reply:

We acknowledge that individual regional modelling studies have explored these processes in more detail. However, they have not previously been explored in global scale model intercomparison projects, e.g. HTAP, ACCMIP, ACCENT. More detailed information on dry deposition, e.g. stomatal/non-stomatal fluxes, land cover specific fluxes and individual resistance terms will be needed to be explicitly diagnosed to allow this to be done in future. However, we accept the point made here, and have reworded our statement to clarify this issue.

Comment 15:

"I am aware that you are using the term here "generally" but wanted to let you know that in the implementation of the dry deposition schemes in the chemistry-climate model system ECHAM3/4 (Ganzeveld et al., 1995, 1998), and I guess also in the implementation of such a scheme by Mike Sanderson, that the role of soil moisture in stomatal exchange was actually included through the use of the climate models stomatal resistance in the dry deposition scheme instead of using the commonly applied Wesely 1989 scheme. I am raising this point since in the recent more frequent discussions/collaborations between the chemistry-climate and air quality community to also work on some of these topics, it appeared that these facts were not known to the colleagues more active in the air quality community regarding the role of emissions/deposition."

Author's reply:

We thank the Editor for drawing attention to the fact that some models do take account of the role soil moisture in determining O₃ dry deposition. We note that ECHAM was unfortunately not used in this study as this might have provided an interesting comparison.

Comment 16:

Line 169: correct the following line "fluxes were normalized (see Eqn. ??)"

Author's reply:

We thank the author for noticing this mistake. We have now removed this text as we are no longer analyzing normalized O₃ dry deposition fluxes.

Comment 17:

Line 172: "This does not account for second order variation in the dry deposition flux which might arise, for example, from the feedback associated with the decrease in deposition velocity as O₃ is removed from the atmosphere."

I would suggest to change this to "feedback associated with the impact of O₃ uptake resulting in a decrease".

Author's reply:

We have simplified this statement and rephrased it to avoid any potential confusion.

Comment 18:

Line 434: “The sensitivity of surface O₃ to small variations in dry deposition velocity over the oceans was also reported by Ganzeveld et al. (2009), who found that surface O₃ differed by up to 60% when the O₃ dry deposition velocity was varied between 0.01 and 0.05 cm s⁻¹. Being the author of that paper I obviously appreciate the discussion on the relevance on oceanic O₃ deposition in the presented study. You are correct in that the 2009 paper mentions the fact that there appears to be a large sensitivity of the Marine boundary layer O₃ budget to changes in the small oceanic O₃ deposition velocity. That initial finding formed the main motivation to further explore this issue on oceanic O₃ deposition. However, the main conclusion then of the 2009 study was that ultimately changes in the MBL O₃ budget through the explicit simulation of VdO₃ as a function of ocean biogeochemistry and mixing conditions, compared to the commonly applied constant VdO₃ schemes (as is the case in the global chemistry-climate models included in your study), were remarkably small. We explained this by indicating that there appear to be a number of compensating effects involved in MBL O₃ budget including the role of chemistry, exchange between the free troposphere and MBL and deposition. On the other hand, your paper focusses on the evaluation of the O₃ deposition fluxes in these global chemistry-climate models and where our analysis revealed changes in simulated O₃ deposition fluxes due to the implementation of more mechanistic deposition approach compared to the constant VdO₃ approach up to +/-25%. These large changes in fluxes are actually an important feature explaining the compensating effects but so also indicating significant changes in the fluxes to be considered in the evaluation of these models.

We thank the Editor for providing a detailed description of the findings from Ganzeveld et al., (2009) here. We have modified our text (Section 4.3, paragraph 4) to more accurately reflect the findings in Ganzeveld et al., (2009).