

Interactive comment on “An evaluation of ozone dry deposition in global scale chemistry climate models” by C. Hardacre et al.

L. Ganzeveld (Editor)

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Dear author, co-authors,

I have read the two reviews that have been provided on your paper on an evaluation of simulated global ozone deposition as well as your response to the raised comments and the revised version of your manuscript. One of the reviewers has mainly some minor comments and that have been all tackled. However, reviewer #2 has some substantial critical remarks on your manuscript and suggested major revisions. 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 aim-

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ing 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. However, the comments that were raised by reviewer #2 has resulted in that this latter component has been become a more explicit aim of your study. Reviewer #2 is also correctly pointing you at the fact that there have been already extensive evaluations of particular model analyses regarding O₃ dry deposition and also where more detailed analysis on the role of different drivers of this deposition has been included.

Because of the quite substantial comments raised by reviewer #2 I have invited this reviewer to check your revision and replies to the raised comments. In addition, in my reading of these replies and the revised version I also came across issues that I think need to be further addressed.

Below you can these additional observations/comments from my side in response to your reply to the reviews and the revised ms.

The main criticism was that your evaluation was mainly limited to European ozone deposition data ignoring the observations available for other regions including the US but also observations collected in the tropics and other sites. You address this issue in your reply indicating that you deem it more appropriate for the evaluation of these global scale models to compare long-term average fluxes. “To compare measured fluxes with the modelled average monthly O₃ dry deposition fluxes we focussed on studies that reported an average O₃ dry deposition flux or where measurement data was independently made available for this study.” 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). This use of long-term data poses a limitation on the number of available observations where many have been collected in field campaigns covering relative short periods. On the other hand, you also indicate that you didn’t use the US CASTNET data since this is according to you mainly a model product combined with observed O₃ concentrations.

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However, the model that is used to calculate dry deposition is based on the inferential method that is driven by the locally measured parameters that mainly control the dry deposition process and these models have been quite extensively evaluated/calibrated with field campaign observations. Consequently, one can anticipate that these flux estimates are pretty accurate and for sure a good reference for a comparison of large-scale models that might not include all the processes and detail on drivers compared the inferential method calculations.

I read in your reply that you have tackled some of the reviewers comments on the missing information on parameters that are quite essential to appreciate the simulated dry deposition, e.g., the land cover types but also including some more details on what kind of dry deposition scheme has been implemented in the different models. I think this is a very useful modification of your manuscript that will more optimally inform the readers and how some of the differences among the models can be explained by differences in dry deposition drivers. Another comment that has been raised by reviewer #2 is about a missing discussion about the role of chemistry in O₃ dry deposition fluxes. In your reply you included the following statement “The importance of these chemical processes is being explored in process-based models in a number of current projects, but the results from these are not yet suitable for implementation in global-scale models. This development work is clearly valuable, and it is hoped that the present study provides a first benchmark against which the improvements from more advanced deposition schemes can be assessed. We have now added a statement highlighting the potential importance of chemistry for dry deposition, but indicating that these factors are not yet included here.” 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

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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.

Another essential modification has been in response to the raised comments has been that, rather to discuss O₃ dry deposition fluxes, you also present some of the dry deposition fields by dividing the normalized flux (using 30 ppbv as a reference value) with this mixing ratio to infer V_d. I would therefore suggest to also state this explicitly in the explanation of these results, e.g., “In Figures 1 (d–f), 2 (e–h), 3, 4 (c–d), 5 (c–d), 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.

In your reply you also indicate the added information in Table 1 about the dry deposition models to address the comment from reviewer #2 about the need for more information on the detail of the dry deposition simulations. You reply by stating that “While a more detailed characterization of model schemes including sources of LAI, LCC mappings and other land surface and meteorological interactions would be interesting, we do not feel that it would aid in interpretation of the results presented in the paper given the very limited monthly model data available”. 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

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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)

In addition, I already indicated in the first stage of the review process that I planned myself to also provide feedback on your feedback, in particular on the oceanic ozone deposition component. However, due to the usual reasons I had so far not yet submitted my editors comments which I already put together having read version of the manuscript submitted last year. I am now still providing this feedback since this might be still useful to consider for a final version of the paper.

Further editors comments

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.

“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?

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

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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??

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 30± N–30± S”. 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.

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”.

Helmig, D., E. Lang, L. Bariteau, P. Boylan, C. Fairall, L. Ganzeveld, J. Hare, J. Hueber, M. Pallandt, Atmosphere-ocean ozone fluxes during the TexAQS 2006, STRATUS 2006, GOMECC 2007, GasEX 2008, and AMMA 2008 cruises, *J. Geophys. Res.*, 117, D4, doi:10.1029/2011JD015955, 2012.

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“It is important to include a well constrained O₃ dry deposition velocity and global area for tropical forests as day time observations of between 3.8–40 cm s⁻¹ (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.

“..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.

Page 16: “although there is less”

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?

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

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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)

Line 108-109: “However, these significant developments have not been generally implemented in global scale models.”

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.

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

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”

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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.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 22793, 2014.

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