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# Reviewer's comments, author's responses and changes to the manuscript

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## 1 Reviewer 1

### Comment 1:

“Page 22795, line 2: also add Tai, A.P.K., M. Val Martin, C.L. Heald (2014), Nature Climate Change, doi:10.1038/nclimate2317 C7657 ACPD 14, C7657–C7658, 2014”

### Author’s Reply:

We have added this reference as suggested.

### Changes to the manuscript:

Introduction, Paragraph 2:

The following text was added:

**“However, the role of O<sub>3</sub> in future climate scenarios is not straightforward. Tai et al., 2014 analysed the combined effect of O<sub>3</sub> and climate change on crop production between 2000 and 2050 finding that O<sub>3</sub> could either exacerbate or offset the effects climate change depending on scenario, crop type and region.”**

### Comment 2:

“Page 22797, line 21: what about the other models, which approach do they use? Identify in Table 1 which models use Wesely. Note that the CAM-Chem model has since HTAP been updated (Val Martin, M., C.L. Heald, S.R. Arnold (2014), Coupling dry deposition to vegetation phenology in the Community Earth System Model (CESM): Implications for the simulation of surface O<sub>3</sub>, Geophys. Res. Lett., 41, doi:10.1002/2014GL059651)”

### Author’s Reply:

We have amended Table 1 to include the dry deposition scheme that was used in the model and the number of land cover classes included in the model. We have included a more detailed table in the supplementary information that details the land cover classes and provides extra

references for models that have modified their dry deposition scheme from the original Wesely scheme. We acknowledge in the paper that, as for CAM-Chem, models may have undergone further development since this time (Introduction, para. 6). However, recent developments such as these do not influence the results contributed to the TF-HTAP study that we analyse here.

### **Changes to the manuscript:**

We have modified Table 1 as described in the author's reply. We have also included supplementary information with extra information about the deposition schemes and land cover classes used in the different models. See also Reviewer 2, Comment 3.

### **Comment 3:**

"Page 22798, lines 11-20: I don't quite understand why the normalization is done using a constant 3 ppbv instead of the model surface ozone (which is used in Figures 6 and 7). Please explain rationale."

### **Author's Reply:**

We "normalised" dry deposition fluxes using constant ozone of 30 ppbv (approximately the global average surface ozone concentration) to remove differences in dry deposition driven by model differences in surface ozone concentration. This method allowed us to compare dry deposition rates for different models and different geographical regions independent of modelled surface ozone concentrations. However, we have now modified this analysis to show dry deposition velocity rather than normalised fluxes 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). The deposition velocity is directly proportional to the normalized flux, so our conclusions are unaffected, and is a more physical quantity. Note that the deposition velocities simulated by the models are not available from the HTAP results, but are inferred from the dry deposition fluxes and surface ozone concentrations for each model.

### **Changes to the manuscript:**

The changes are described in detail at Reviewer 2, Comment 12.

**Comment 4:**

“Page 22800, line 10: could you identify how many models were similar with those studies?”

**Author’s Reply:**

We have included short statements (Section 3.1, Para2. Approx. Page 22800, L10-20) noting how many models were similar in the comparisons made by Wild et al., 2007 and Stevenson et al., 2006 and Young et al., 2013.

**Changes to the manuscript:**

Section 3.1, Paragraph 2:

We have indicated how many models participating in the ACCENT and the study by Wild et al., 2007 were similar to the model ensemble used in this study.

## 2 Reviewer 2

**Comment 1:**

“A major problem is that the comparison with measurements makes use mainly of very old data from a limited number of sites, almost all of them in Europe, and for short-term periods. The authors seem unaware of the large network of sites which are used to estimate O<sub>3</sub> deposition in both Canada and the USA over long-periods (e.g Schwede et al, 2011, Zhang et al. 2002a). Data also exist from South America and Asia Tropical forests - Ganzeveld and Lelieveld showed some back in 1995 and there have been many measurements since then”

**Author’s Reply:**

To make a useful comparison between modelled monthly ozone dry deposition fluxes and measurements we sourced long term data sets (generally of at least 1 year) from which a

monthly average value could sensibly be determined. Two of these data sets were obtained within the last 5 years, and they span four land cover classes (coniferous forest, deciduous forest, grass land and citrus orchard). We agree that the CASTNET data over the US is a useful resource for model-measurement comparisons, but we elected not to use them for this study because the ozone fluxes reported are model-derived based on measured surface ozone and meteorology. However, we have now expanded the number of short term data sets used in this study to include measurements from french maize crops, tropical forests and oil palm where either the raw data or a suitable literature value could be sourced. We acknowledge that there are good deposition measurements that have been made in many other locations. However, most of these are unsuitable for the present comparison where we only have monthly-averaged model fluxes to compare with. Depending on the focus of the measurements many studies only report stomatal conductance, maximum or minimum fluxes or deposition velocities. Given the uncertainties involved, we do not feel that it would be useful to compare inferred model deposition velocities with measured deposition velocities at some sites and fluxes at other sites.

### **Changes to the manuscript:**

Methods, Paragraph 5:

The following text was added.

**“To compare measured fluxes with the modelled average monthly O<sub>3</sub> dry deposition fluxes we focussed on studies that reported an average O<sub>3</sub> dry deposition flux or where measurement data was independantly made available for this study.”**

#### 1. Table 3:

New short term measurement sites have been included. These are three maize crops in France (Stella et al., 2011), tropical forest in Malaysian Borneo (Fowler et al., 2011) and the Amazon (Rummel et al., 2007), and an oil palm plantation in Malyasian Borneo (Fowler et al., 2011).

Figure 11 (previously Figure 10):

The new measurement data has been incorporated in to Fig. 11. The comparisons between modelled and measured O<sub>3</sub> fluxes have been grouped according to land cover class and plotted on separate sub-plots.

### Section 5.3, Paragraph 2:

We have extended the discussion of the comparison between the modelled and measured fluxes at the short term measurement sites to include the additional sites.

### **Comment 2:**

“The choice of land-cover categories used in this comparison is also difficult to understand. Why look at data from untypical LCCs such as onion fields, but not at major crop species such as wheat or maize, or of grasslands (outside Europe), which have been the subject of many deposition studies (eg Gerosa et al., 2003, Bassin et al., 2004, Finkelstein, 2001, Stella et al., 2011, Tuovinen et al., 2004, Val Martin et al. 2014). Or data for Tropical forests?”

### **Author’s Reply:**

As noted above, we have now expanded the number of measurement sites to include data from maize crops, tropical forests and oil palm. Although some of the measurement sites are from atypical LCCs, they do contribute to information about dry deposition to crop land cover in general. Further, most of the long term datasets which we focus our model-measurement comparison on are from sites in coniferous forest, deciduous forest and grass land which are major land cover classes.

### **Changes to the manuscript:**

Please see the changes made for Comment 1.

### **Comment 3:**

“One problem is the need to make many assumptions when looking at land-cover specific deposition velocities. Related to this, the paper provides few details that would help the reader understand model-to-model differences. There should be a table with some description of the basic features of deposition from each model.

Some examples: -do the models use constant or e.g. monthly LAI estimates? If so, where from? -Are Wesely schemes really used in all models? If not, what is used? -Since tropical forests are globally very important, and also turn out here to have very different deposition velocities, the treatment of these also deserves special mention.

I do not expect a detailed review, but a paper which compares 15 models for one specific model output (dry deposition) should take the time to catalog such details, and inform the reader about the main characteristics of the models.”

### **Author’s Reply:**

We have now amended Table 1 to include the dry deposition scheme that was used in the model and the number of land cover classes included. We have also included a more detailed table in the supplementary information that provides more information on the land cover classes and additional references where dry deposition treatments have been modified from the original Wesely scheme. 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. A more detailed analysis of hourly fluxes by vegetation type would certainly benefit from this information, and we have proposed this for ongoing and future model intercomparisons (CCMI, HTAP phase 2). We also acknowledge in the paper that a number of models, e.g. CAM-Chem, have undergone further development since the original HTAP study, but our analysis necessarily reflects the state of the models at that time.

### **Changes to the manuscript:**

We have modified Table 1 as described in the author’s reply. We have also included supplementary information with extra information about the deposition schemes and land cover classes

used in the different models. See also Reviewer 1, Comment 2.

**Comment 4:**

“The paper fails to refer to and discuss quite a few previous papers which provide lessons in the difficulties CTMs face with dry-deposition modeling. For Europe the papers of e.g. Tuovinen et al., 2004, 2009 discuss in detail the importance of various factors when dealing with ozone deposition in the EMEP CTM. Similarly, the Canadian AURAMS model team have documented extensive development and testing of dry deposition modules (Zhang et al., 2002b, 2006). Results from such studies should inform any discussion of global CTMs.”

**Author’s Reply:**

We have included a brief discussion of the development of dry deposition schemes in other CTMs where appropriate.

**Changes to the manuscript:**

Section 5.3, Paragraph 6:

We have extended the text here to include a discussion of previous studies (Tuovinen et al., 2004, 2009 and Zhang et al., 2002b) that have compared modelled O<sub>3</sub> dry deposition fluxes against measurement data.

**Comment 5:**

“Given these points, I miss a proper discussion of the major weaknesses of current models.”

**Author’s Reply:**

One of the main objectives of this paper is to identify the weaknesses associated with dry deposition in global scale models, as noted in the abstract. Dry deposition across different global scale models has not been reviewed recently, so it is not known how well they perform.



We clearly identify some key areas where there is substantial variation between models, in particular over oceans and tropical forests, and have discussed these in detail. Our aim in identifying these areas is to focus the attention of the modelling community on these issues so that they can prioritise further model development. A more thorough, process-based attribution of the major causes of model differences requires more detailed model output (hourly, LCC-specific fluxes, etc.) and we have requested this for ongoing model intercomparisons (CCMI, HTAP phase 2) as noted above.

**Changes to the manuscript:**

We have not made any changes the manuscript specially addressing this comment.

**Comment 6:**

“P22813, L26-27 makes the claim that ‘O3 dry deposition has not previously been reported at this level of detail’, but I would argue that the EMEP or AURUMS evaluations are far more detailed in terms of process understanding. The current paper adds a dimension with a multiple-model comparison, but makes rather broad assumptions with regard to LCC comparisons; it cannot be called illustrative (which is also useful), but not detailed.”

**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 need 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.

**Changes to the manuscript:**

Conclusions, Paragraph 6:

The text has been modified as described above.

**Comment 7:**

“The authors make no mention of the role of chemistry in perturbing real and apparent ozone deposition - for examples issues discussed in Wolfe et al (2011) for forests, or Chang et al. (2004) for oceans. Given that such process may have very important effects on measurements of O<sub>3</sub> deposition, some mention is warranted.”

**Author’s Reply:**

Near-surface and in-canopy chemical processes are of course important for controlling dry deposition, particularly for tropical forests, other forests and oceans. However, the physical scales at which these occur are typically too small to be resolved in global scale models, and hence some form of parameterization is required. 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.

**Changes to the manuscript:**

Section 5.3, Paragraph 5:

The text has been modified as described above.

**Comment 8:**

“P22798, last paragraph. It would help the reader to get some idea of differences in near-surface O<sub>3</sub> over different LCCs, since vertical gradients in O<sub>3</sub> can be substantial near the surface.”

**Author's Reply:**

It would certainly be interesting to investigate the near-surface profiles of O<sub>3</sub> and their diurnal variation, as this would provide substantial insight into near-surface chemistry and deposition in the models. Unfortunately the coarse, monthly-mean data available from the HTAP study is insufficient to permit this. However, as noted above, some ongoing and future intercomparisons are requesting hourly ozone diagnostics that would allow these profiles to be explored. Interpretation of these will require some caution, as there is likely to be a strong influence from assumptions about boundary layer mixing timescales and because near-surface gradients occur over smaller vertical scales than resolved by the models.

**Changes to the manuscript:**

We have not made any changes the manuscript specifically addressing this comment.

**Comment 9:**

“Section 5.3 lacks a lot of information and I am really not sure it adds anything. The data used are very old, which is a shame since there have been many nice ozone deposition data sets produced in the last decade (e.g. Stella et al., Biogeosci., 2011). There is no documentation of the sites methodology or data-quality issues. With ozone deposition the latter is very important, since any flux estimate is very sensitive to many factors.”

**Author's Reply:**

We have relied on well-verified long-term deposition data sets, but acknowledge that more recent measurements are available (some of which we have already include). We have now expanded the number of short term measurement sites and the range of LCCs represented for the results presented in Section 5.3. We have also modified the presentation of this data so that sites are grouped by LCC. Although it is more difficult to draw clear conclusions from short term measurements, we believe that they provide useful additional indication of model performance and weaknesses.

**Changes to the manuscript:**

Please see the changes made for Comment 1.

**Comment 10:**

“Units are given in kg/m<sup>2</sup>/s, which gives values of order 1e-10. Why use such as an awkward unit? For nitrogen deposition a common unit would be kg/ha/year, with 1e-10 kg/m<sup>2</sup>/s being ca 32 kg/ha/year - much more manageable. In Europe, fluxes tend to be given in mmole/m<sup>2</sup>/year, more consistent with the POD concept.”

**Author’s Reply**

We have used standard SI units (kg m<sup>-2</sup> s<sup>-1</sup>) throughout this study. However, we acknowledge that deposition fluxes are often presented in nmol m<sup>-2</sup> s<sup>-1</sup>, and we have therefore changed our figures to reflect this (Figs 6a, d and g; 7a and d; 8a and d; 9a-g, 10a-9). This may make comparison with other studies easier, and may aid in estimating dose-response metrics.

**Changes to the manuscript:**

For Figures 6, 7, 8, 9 and 10 (previously Figures 8 and 9), and 11 the flux units have been changed from kg m<sup>-2</sup> s<sup>-1</sup> to nmol m<sup>-2</sup> s<sup>-1</sup>.

**Comment 11:**

“The references used are typically rather old, and should be modernized throughout. As one example, on P22794, L24, the reader is asked to "look for references therein" from a 2005 paper. There have been many important studies the last 10 years, so provide more up-to-date references.”

**Author’s Reply**

Introduction (throughout):

We have now included additional references from up-to-date sources where they add substantial new information.

**Comment 12:**

“P22798, L12. Why were fluxes normalized to 30 ppb? A simpler procedure would be to just present deposition velocities.”

**Author’s Reply**

We "normalised" dry deposition fluxes using constant ozone of 30 ppbv (approximately the global average surface ozone concentration) to remove differences in dry deposition driven by model differences in surface ozone concentration. This method allowed us to compare dry deposition rates for different models and different geographical regions independent of modelled surface ozone concentrations. However, we have now modified this analysis to show dry deposition velocity rather than normalised fluxes 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). The deposition velocity is directly proportional to the normalized flux, so our conclusions are unaffected, and it is a more physical quantity. Note that the deposition velocities simulated by the models are not available from the HTAP results, but are inferred from the dry deposition fluxes and surface ozone concentrations for each model.

**Changes to the manuscript:**

Methods, Paragraph 4:

We now describe how  $O_3$  dry deposition velocities are calculated rather than normalised  $O_3$  dry deposition fluxes.

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  $O_3$  dry deposition fluxes have been changed to deposition velocity. We have modified the text where appropriate to reflect these changes in the Results sections.

Figure 1:

Average regional  $O_3$  dry deposition velocity is shown in Figs. 1d, e and f instead of normalised total  $O_3$  dry deposition.

Section 3.1, Paragraph 3 and 4:

To compare the variation in  $O_3$  dry deposition fluxes and deposition velocities across the ensemble we included a calculation for relative standard deviation (RSD). The text has been modified in these two paragraphs to include a discussion of RSD rather than differences in total  $O_3$  dry deposition.

Figure 2:

Average  $O_3$  dry deposition velocity at each latitude band is now shown instead of normalised total  $O_3$  dry deposition.

Relative standard deviation is shown in Figs. 2b and f instead of the range in total  $O_3$  dry deposition across the model ensemble.

Section 3.2, Paragraphs 2 and 3:

The text has been changed here to include discussion of the variation in deposition velocity at different latitude bands as well as total deposition. RSD is also discussed to compare the variation between deposition velocity and deposition flux across the model ensemble.

Figure 3:

Dry deposition velocity is shown instead of normalised deposition flux.

Figures 4 and 5:

Deposition velocity is shown in Figs 4c, d and Figs. 5c, d instead of normalised deposition fluxes.

Figure 6b, 6e, 6h, 7b, 7e, 8b and 8e:

A plot of modelled v. measured dry deposition velocity replaces the normalised modelled v. measured dry deposition fluxes for each measurement site.

**Comment 13:**

“P22794, L16-22. This paragraph needs references!”

**Author’s Reply**

We have included some new references for this paragraph.

**Changes to the manuscript:**

Introduction, Paragraph 1:

“Ozone is a significant trace gas constituent in the troposphere. The two main sources of tropospheric ozone are transport from the stratosphere and in situ chemical production via the oxidation of hydrocarbons and CO in the presence of nitrogen oxides (NO<sub>x</sub>) (Crutzen et al., 1974; Liu et al., 1980; Atkinson et al., 2001). Tropospheric ozone, in addition to being a greenhouse gas (IPCC 2013), is the primary driver of chemical oxidation in the troposphere as a source of OH radicals (e.g. Prather et al., 2001) and is also a potent pollutant in its own right (Royal Society Policy Report, 2008).”

**Comment 14:**

“Odd not to mention here EMEP, the one (also global) CTM which implements DO3SE. In fact, wasn’t DO3SE developed in cooperation with the EMEP team? There are a long list of papers covering this linkage.”

**Reply to Comment 14:**

We have now included a statement that the EMEP model utilises the DO3SE model and referenced it accordingly. We hope to see these more integrated treatments of dry deposition

processes become more standard in global models in future.

### **Changes to the manuscript:**

Introduction, Paragraph 8:

“Comparatively recent process models such as DO3SE (Emberson2000a, Emberson et al., 2000b, Emberson et al., 2001 Buker et al., 2007), which was developed to estimate stomatal ozone flux, do parameterize the effect of soil water deficit and vapour pressure deficit on stomatal conductance. DO3SE has recently been included in the EMEP model (Simpson et al., 2012), however, these significant developments have not been generally implemented in global scale models.”

### **Comment 15:**

“P22799, L13-15. This is a little confusing. I assume you mean normalised fluxes when taking ratios?”

### **Author’s Reply:**

We now present deposition velocities rather than normalised fluxes. These are derived from the partitioned fluxes and monthly surface ozone from each model, and we have added a short statement to describe this.

### **Changes to the manuscript:**

We have not made any changes the manuscript specially addressing this comment.

### **Comment 16:**

“P22806, L6. I agree that deposition to water is important for the sink strength, but deposition to the land ecosystems has important implications for growth aand hence carbon sequestration.”



## **Author's Reply**

From an impacts perspective, deposition to vegetation is clearly very important. We have included a statement here about the wider ranging effects of dry deposition to land-cover.

### **Changes to the manuscript:**

Conclusions, Paragraph 3:

We included the following text.

“The relative magnitude of the deposition to oceans means that it is important for the global ozone budget to constrain this term. However, deposition to terrestrial ecosystems has important implications for many other components of the Earth system including carbon sequestration, hydrology and atmospheric composition.”

### **Comment 17:**

“P22807, L16. Agreement between models should not be the aim of any exercise. The aim is to agree with the real world”

### **Reply to Comment 17:**

The reviewer is of course correct to point this out, and we have modified our statement to avoid implying that agreement between models is the main aim. Nevertheless, disagreement between models is indicative of disagreement with the real world and provides useful insight. Discrepancies over oceans and tropical forests are especially large, highlighting specific issues that need to be rectified. For example, a tropical forest land cover class is not included in the deposition schemes of some models. Including tropical forests would remove one source of uncertainty in these models and give a better representation of dry deposition at the global and regional scale.

### **Changes to the manuscript:**

Section 4.3, Paragraph 4:

We have changed the final sentence as follows:

“However, improved constraints on the seasonal amplitude in fluxes to coniferous forests, mixed coniferous forests, agricultural crop land and high latitude LCCs, in addition to more coherent representation of land cover and LAI across the models would contribute to a better representation of dry deposition in models.”

### 3 Other changes

Section 4.1, Figures 6, 7 and 8:

Please note that in Figures 6, 7 and 8 the model-measurement comparison plots for the long term data are now grouped by land cover type. Figure 6 presents data from Coniferous Forest sites, Figure 7 presents data from deciduous forest sites and the newly added Figure 8 shows data from a grass land and a citrus orchard.

Section 4.1, Paragraph 1:

We have included a referenced estimate of uncertainty in the eddy covariance technique commonly used to measure  $O_3$  dry deposition fluxes.