Reply by the authors to the Referee #2's comments on
"Revising global ozone dry deposition estimates based on a new mechanistic parameterisation for air-sea exchange and the multi-year MACC composition reanalysis" (#acp-2017-768)

Anonymous Referee #2 (RC2)

We are grateful to the Referee for a long set of comments. In the following, we provide a response to these comments.

(1) **Comment:** *A modified version of a recently presented ozone ocean dry deposition scheme [Luhar et al., 2017] is presented. The model performance is evaluated by comparing modelled ozone deposition velocities with previously published data from oceanic cruises. Further, the global ozone ocean flux is modeled based on this new model configuration, yielding a lower oceanic ozone sink than prior estimates. The ozone ocean flux is then compared with the ozone land sink, and a new total global ozone flux estimate is derived.*

Response: Thanks for the comment.

Changes in manuscript: None.

(2) **Comment:** *Major Comments:*

While the authors repeatedly highlight their work as being a new 'scheme', as far as I understand this modeling in essence differs only in one aspect (the ocean layer is described in two, rather than a single layer) from their prior ACP publication [Luhar et al., 2017] that was submitted only ten months prior to this current paper. The article claims this ozone flux parameterization and modeling to be a novel development. However, from reading the earlier publication [Luhar et al., 2017] again, and the works by Ganzeveld et al. [2009] and Fairall et al. [2007], it appears that the the physical and ocean biochemical dependency description were mostly adaptations of principles presented in these earlier publications.

Response: All the references mentioned above by the referee are included in our paper. We clearly elucidate in our paper what the shortcomings are of the previous model formulations, including the two-layer formulation used by Luhar et al. (2017, https://doi.org/10.5194/acp-17-3749-2017) which was based on the approach of Fairall et al. (2007). We agree that our new two-layer scheme (Eq. 16) includes the same overall physical and ocean biochemical processes as in the studies by these authors, but it improves the mathematical formulation by the use of a more realistic assumption. The new scheme corrects a serious flaw (as stated in the Luhar et al. (2017) paper) of the previous two-layer scheme by including chemical reactivity throughout the oceanic mixing layer (as is observed for dissolved iodide) rather than just within the top few microns of the water surface. The new model will also apply to any other chemical compounds that are taken up by the oceanic mixing layer. We accept that this represents an incremental advance in model development, but we believe it's a significant advance. Additionally, our work also makes use of the global ozone reanalyses developed under the European MACC program to constraint the ozone dry deposition budgets better. These reanalyses have not previously been used for ozone deposition purposes and thus provide scope for a novel application.

Changes in manuscript: To consider the Referee's point, perhaps we could qualify the title a little better to read "Revising global ozone dry deposition estimates based on a new two-layer parameterisation for air-sea exchange and the multi-year MACC composition reanalysis."

Comment: *In this model the ozone ocean flux description builds exclusively on chemical removal of ozone by reaction with iodide (I-). Consideration of this reaction is not that novel, having been proposed quite some time ago. Other previous work has suggested that, while the I-reaction has high significance, other secondary reactions, such as those with dissolved organic matter (DOM) in the ocean surface microlayer, may play a role in the ozone reaction as well [Ganzeveld et al., 2009; Coleman et al., 2010]. Ganzeveld et al. [2009] showed, for example, that evaluation of the simulated O3 dry deposition velocities with a 1-layer version of the [Fairall et al., 2007] model, including only I- in the calculation of total reactivity, underestimated the measured coastal deposition velocities. The role of dissolved organic matter (DOM)-O3 chemistry was proposed to explain these discrepancies. [Coleman et al., 2010] specifically addressed the role of DOM-O3 chemistry in deposition to the Atlantic Ocean. These authors conclude: "… iodide reactions alone cannot account for observed deposition velocities. Consequently, we suggest a missing chemical sink due to reactions of ozone with organic matter at the air-sea interface." It does not appear that this Luhar et al. article takes this into consideration. The question if and how much uncertainty potentially results from this neglect is not addressed by their publication.*

Response: Part of this comment, particularly about DOM, is similar to a comment made by Referee #1. We thus refer to our Response #3 to Referee #1 where we present additional deposition calculations with DOC (equivalent of DOM) included.

One novelty of our work is to provide a better mathematical formulation for the inclusion of waterside chemical reactivity. The work of Luhar et al. (2017) showed clearly the limitation of the one-layer scheme of Fairall et al. (2007) in describing the deposition velocity data of Helmig et al. (2012) (as a result of an overestimation of waterside turbulence-chemistry interaction in this scheme), and replaced it with a two-layer model with an arbitrary constraint on chemical reactivity. The model presented here removes this arbitrary constraint on chemical reactivity and thereby is a more realistic model of ozone interaction with ocean water.

The focus of modelling in our paper is on ozone deposition to open ocean regions. Using the one-layer Fairall et al. (2007) scheme, Ganzeveld et al. (2009) show that the inclusion of the O3-DOM reaction (with DOM represented by chlorophyll-a) significantly increased deposition velocity at coastal sites but gave mixed results compared to observations, and that for open ocean sites, there were only small changes to deposition velocity. The work of Coleman et al. (2010) also relates to coastal waters. None of the papers by Fairall et al. (2007), Ganzeveld et al. (2009) and Coleman et al. (2010) has used the more recent open-ocean deposition velocity data of Helmig et al. (2012) that Luhar et al. (2017) and the present work use (because these data had not been available at the time).

Our work suggests that the ozone-iodide reaction is able to describe the available open-ocean deposition velocity data within the uncertainty of model parameters and scatter in the data. Clearly more observations are needed to establish the relative role of additional ozone reactions.

There is some evidence (e.g. Coleman et al., 2010) that ozone deposition velocities over coastal waters are larger than those over open oceans. Our deposition approach for coastal grid cells in the model is qualitatively consistent with that behaviour and is described in our Response #7 below.

We have calculated a measure of uncertainty in the global oceanic and total ozone deposition fluxes and the details are given in our Response #5 below.

Changes in manuscript: Our Response #3 to Referee #1 on DOM will be included along with a summary of the points discussed above.

(4) **Comment:** *Further, building exclusively on O3 + I- chemistry, the proper description and consideration of I- in the ocean must be of high importance. The article does not provide any detail on what data the I- oceanic description builds on. Are these new observations? Or is the I- modeled based on other relationships? In [Ganzeveld et al., 2009], I- was estimated based on its correlation with nitrate. While this seemed to be a reasonable, and possibly the best possible approach at that time, does this paper take advantage of the much improved I- description presented by Chance et al. [2014]? Despite this progress, there certainly remains large uncertainty in the spatial and temporal representation of I-, e.g. its concentrations in high-latitude waters, which is hampered by a lack of in-situ observations. This is actually the region where, according to this study by Luhar, the largest differences in the O3 dry deposition velocities compared to the older/other deposition approaches are observed (Figure 9 in [Luhar et al., 2017]). As far as I understand, these uncertainties are likely many times larger than the rather narrow uncertainty windows in the ozone deposition budgets that are presented in this new Luhar et al. publication. Unfortunately, the authors do not elaborate on this question, which I consider a severe neglect.*

Response: It is clearly stated in our paper that the ocean iodide concentration is based on Eq. (20), which is from MacDonald et al. (2014). Chance et al. (2014) examined statistical relationships between iodide and parameters such as SST, nitrate, salinity, chlorophyll-a, and mixed layer depth and found that SST was the strongest predictor of iodide. MacDonald et al. (2014, with Chance as a co-author) used data from several cruises in the Atlantic and Pacific oceans covering the latitudes 50°S–50°N to derive their parameterisation for iodide concentration, which we have used. A sensitivity analysis involving the iodide parameterisation by Chance et al. (2014) was reported by Luhar et al. (2017) (see their Figure 5) and it was compared with the behaviour obtained using the MacDonald et al. (2014) iodide parameterisation. We have included some discussion on the use of the Chance et al. parameterisation and included a deposition velocity curve as Option 6 based on this parameterisation in our Response #5 to Referee #1.

We agree that there is considerable uncertainty in the representation of iodide concentrations, particularly in high-latitude waters, due to the lack of in-situ observations. The rather narrow uncertainty windows in our annual ozone deposition fluxes are solely due to the interannual variability inherent in the modelled meteorology and the MACC ozone concentration fields. We have now done additional calculations to determine the uncertainty range better, and this is given as a response to Comment #5 below.

Changes in manuscript: The sensitivity to the Chance et al. (2014) iodide parameterisation conducted in our Response #5 to Referee #1 will be included. Additional work on uncertainty reported below under Comment #5 will be included.

(5) **Comment:** *Developed flux estimates are presented with error windows (see abstract line 23) that are on the order of 5%, but those windows are simply the standard deviation of the year to year variability in the modeled flux based on changing meteorology. They are not the uncertainty in the estimates of the ozone flux. Those, likely, would be much larger, making the way this is presented quite misleading.*

Response: The Referee is correct – the reported uncertainty in our annual ozone deposition fluxes is solely due to the interannual variability in the modelled meteorology (with nudging) and the MACC ozone concentration fields. We have done additional calculations to estimate uncertainty in deposition flux to the ocean better and this is described below. The global ocean and total ozone deposition fluxes with the new uncertainty estimates are **98.4 ± 30.0** Tg yr^{-1} and **722.8 ± 87.0** Tg yr^{-1}, respectively.

Changes in manuscript: We plan to include a concise version of the following calculations.

Previous global deposition modelling studies include uncertainty estimates in the modelled annual deposition loss. Stevenson et al. (2006) report a global ozone deposition of 1003 ± 200 Tg yr^{-1} for the year 2000 based on 21 models. The global deposition calculated by Wild (2007) using 17 post 2000 modelling studies is 949 ± 222 Tg yr^{-1} whereas that reported by Young et al. (2013) for the year 2000 based on a subset of six models participating in the ACCMIP intercomparison study is 1094 ± 264 Tg yr^{-1}. Hardacre et al. (2015) obtained a deposition loss of 978 ± 127 Tg yr^{-1} based on 15 global chemistry transport models driven by meteorological fields for the year 2001. The (1σ) uncertainty in all these studies is by and large all due to model to model variations.

Our calculations in the paper yield a deposition loss to the ocean of **98.4 ± 4.5** Tg yr^{-1} and a total global deposition of **722.8 ± 20.9** Tg yr^{-1}, with the 1σ error bounds in these estimates only representing the 10-year interannual variability in the modelled deposition velocity and MACC concentration fields. These error bounds do not include any other uncertainties in the deposition velocity modelling or those in the MACC concentration fields.

In our present single model study, there is no simple and reliable way of quantifying modelling uncertainty in deposition flux arising from the approximations and assumptions used. For the oceanic component, which is the main focus here, one way to estimate uncertainty is to consider the scatter in the deposition velocity observations of Helmig et al. (2012) shown in Figure 4 of our paper. This Figure shows some large fluctuations in the v_d data that are not present in the modelled values. We take the difference between the scatter in the v_d data and that in the modelled values as a measure of uncertainty that is not captured by the model. We call this difference the residual uncertainty which we aim to account for.

In order to quantify the residual uncertainty, four SST ranges, namely < 15, 15–21, 21–28 and > 28 °C were considered, which approximately correspond to the SST ranges of the cruise experiments shown in Figure 4. For each SST range, the v_d data were detrended by fitting a linear regression line and the variance (σ^2_{vdo}) of the detrended data was calculated. Similarly, the variance (σ^2_{vdm}) of the detrended modelled v_d values was calculated for each SST range. The quantity $(\sigma^2_{vdo} - \sigma^2_{vdm})^{1/2}$ is taken as the residual uncertainty, and it was 0.0046, 0.0093, 0.0049 and 0.0056 cm s^{-1} for the above SST ranges, respectively.

To calculate the uncertainty range in oceanic deposition flux as a result of the residual uncertainty in v_d, the 3-h modelled depositions velocity fields were perturbed by the above residual uncertainty values (which depend on SST) and then multiplied with the 3-h MACC ozone fields. These calculations were done for all 10 years. The uncertainty range in the oceanic deposition flux thus obtained was ± 30.0 Tg yr^{-1}, which includes the small interannual variability of ± 4.5 Tg yr^{-1} stated earlier. Our total oceanic deposition of 98.4 Tg yr^{-1} with the new uncertainty ± 30.0 Tg yr^{-1} can be contrasted with the value 340.0 ± 98.6 Tg yr^{-1} obtained by Hardacre et al. (2015). It is interesting to note that our oceanic deposition and its uncertainty are both approximately a third of the respective values obtained by Hardacre et al. (2015). There would be some uncertainty in the MACC ozone data apart from their interannual variability which we have not considered.

With regards to uncertainty in deposition to non-water surfaces, since our model uses the same Wesely (1989) deposition scheme as most other models for such surfaces, we assume that the corresponding uncertainty would be similar to that in those models. Only Hardacre et al. (2015) report uncertainties in deposition fluxes to both water and non-water surfaces, with the latter calculated to be ± 80.0 Tg yr^{-1}. This value when combined with the interannual variability of ± 17.4 Tg yr^{-1} for non-water surfaces obtained in our paper leads to a total uncertainty of ± 82.0 Tg yr^{-1} for such surfaces. Hence the total uncertainty combining this non-water component (± 82.0 Tg yr^{-1}) and the revised water component (± 30.0 Tg yr^{-1}) derived above is ± 87.0 Tg yr^{-1}.

In summary, with the revised uncertainty estimation, the global oceanic and total deposition fluxes are **98.4 ± 30.0** Tg yr^{-1} and **722.8 ± 87.0** Tg yr^{-1}, respectively. The reduction in the total uncertainty compared to Hardacre et al.'s (2015) value of ± 127 Tg yr^{-1} is due to the reduction in uncertainty in the water component of the deposition flux.

(6) **Comment:** *Secondary analyses, such as comparison of modeled boundary layer ozone, global ocean flux budgets, and attribution of the oceanic flux to the total global flux that build on this modeling, are consequently highly uncertain as well. I therefore question the value of these secondary analyses. For instance, differences between the two schemes shown in Figures 9 and 10 are on the order of 0-25%. Of how much value are these results when the uncertainty in the reactivity is maybe on the order of 100-200%? To me, what I think needs to be addressed most urgently are these questions:*

- How much of the total oceanic ozone flux can be attributed to I-, versus other reactants?

- What are the oceanic I- fields? How does I- change with time and location? And how can this variability be best incorporated into the model?

Unfortunately, these questions are not identified and addressed in this paper.

Response: Please see our Response #3 to Referee #1 on DOM which demonstrates that the ozone-iodide chemistry is sufficient to describe the available open-ocean v_d measurements and their dependency on SST, and that the inclusion of other reactions (i.e. DOM) would deteriorate model performance.

As mentioned above, we used the iodide parameterisation of MacDonald et al. (2014) which is based on data from several cruises in the Atlantic and Pacific oceans covering the latitudes 50°S–50°N. This parameterisation is a function of SST, so depends on location and time. We have done additional uncertainty calculations described above. See Response #5 to Referee #1 on the Chance et al. (2014) iodide parameterisation.

Changes in manuscript: Additional calculations on DOM to be included. Details of the iodide parameterisation used will be provided. Appropriate text from our Response #5 to Referee #1 on the Chance et al. (2014) parameterisation to be included.

(7) **Comment:** *Other Comments:*

The Bariteau et al. [2010] article makes a point that ozone fluxes are higher near the coasts compared to the open ocean. Was that considered in this modeling? And if not, how much uncertainty is potentially due to this neglect?

Response: In our global modelling, the coastal grid cells that include terrestrial surface fractions are handled as follows.

A grid-box mean deposition loss rate is calculated using the individual modelled deposition velocities weighted by the fractions of the surface types present in the grid box. Our two-layer deposition scheme for the ocean is only used when the fraction of water surface in a grid box is greater than 60%. In all other cases Wesely's (1989) scheme for v_d is used, including the use of r_c = 2200 s m^{-1} for water surface. Thus for coastal water grid cells that include fractions of other surface types, the modelled deposition velocities are greater than those for the grid cells fully covered by water because terrestrial surfaces have higher deposition velocities than water and also

because of the use of $r_c = 2200$ s m^{-1} for the water title when its fraction is less than 60%. We will state this in the paper.

There is some evidence that the ozone deposition velocities over coastal waters are larger than those over open oceans (e.g. Coleman et al., 2010; Bariteau et al., 2010), which could be due to factors such as advection from land if the distance between the monitor and coastline (i.e. fetch) is limited, stronger chemical reactivity and turbulence with strong diurnal dependence. Our approach for treating coastal grid cells is qualitatively consistent with ozone deposition velocities over coastal waters being larger than over the open sea. But we do not include any additional/special processes for coastal waters.

Changes in manuscript: The above details to be summarised in the paper.

(8) **Comment:** *The performance of the deposition model leans heavily on data from the six open ocean cruises shown in Figures 4 and 5. Did the authors attempt other comparisons, for instance using any of the other data sets that were summarized in [Helmig et al., 2012]? Given that, as currently done, it appears that the validation relies exclusively on the data from a single group, it should be shown that those cruises are representative for the entirety of available data. Furthermore, these data do not appear to be publicly available, or hosted by any data center? In our research center (and I think this is becoming more common within the community) it is customary to cite the doi of the data set, invite the providers of the data for co-authorship, or at least acknowledge the data providers, whenever those data make a significant contribution to a publication, including comparisons in modeling studies.*

Response: It is correct that our model performance testing is based on published data from Helmig et al. (2012) which cover the latitudinal range 45°N–50°S. Surface based ozone flux stations employing the eddy-covariance technique enables a direct measurement of ozone dry deposition velocity. The data of Helmig et al. (2012) are the only such measurements available over the ocean.

We looked up the very sparse datasets by other researchers summarised by Helmig et al. (2012). None of these studies involved a surface-based eddy-covariance technique over the ocean. The ones that used such a technique were coastal measurements (i.e. Gallagher et al., 2001; Whitehead et al., 2010). The measurements by Lenschow et al. (1982) and Kawa and Pearson (1989) used aircraft-based eddy-covariance over the ocean.

The considerably larger sample size and the possibility of the use of improved experimental techniques in the cruise measurements of Helmig et al. (2012) compared to those reported by earlier studies provide an incentive to use these data.

On line 23–26, on Page 14, we say "As in Luhar et al. (2017), the v_d versus SST cruise data used for comparison with the model are those with the wind speed dependence retained (Ludovic Bariteau, personal communication, 2016) and not the data originally reported by Helmig et al. (2012) in which the wind-speed dependence was removed. While this approach is logically correct, there is not a large difference between the data with and without the wind-speed dependence." We will acknowledge L. Bariteau, as was done in Luhar et al. (2017).

Changes in manuscript: The other studies mentioned above can be cited. We will acknowledge L. Bariteau.

(9) **Comment:** *Page 1/Line 11: I don't see what the term 'consistent' qualifies in this context (consistent with what?)? So, I recommend deleting this.*

Response: Done.

(10) **Comment:** *1/17: As detailed above, I think the term 'new' is a bit of an overstatement. Yes, this paper does present some advancements in the ozone ocean uptake modeling, but most of the mechanisms, considered reactants, and dependencies were presented in prior publications.*

Changes in manuscript: We now use 'two-layer' in place of 'new' to qualify the work somewhat. Also, as we mentioned earlier, we will change the paper title slightly to read "Revising global ozone dry deposition estimates based on a new two-layer parameterisation for air-sea exchange and the multi-year MACC composition reanalysis."

(11) **Comment:** *1/25: Atmospheric models appear to mostly overestimate surface ozone [Parrish et al., 2014]. The results presented in this Luhar et al. manuscript show an increase of modeled ozone, thereby further increasing the discrepancy between models and observations. So, from that perspective, don't these changes go in the wrong direction?*

Response: The ACCMIP multi-model study by Young et al. (2013) shows an overall overestimation of ozone in the lower troposphere in northern mid-latitudes and underestimation in southern tropics and mid-latitudes. Parrish et al. (2014) also show that models overestimate ozone in northern mid-latitudes. Our version of UKCA in ACCESS generally underestimates observed tropospheric ozone, particularly in mid to high latitudes.

Our aim is to improve the process modelling of ozone deposition to the ocean. If a model performs worse when a particular process in improved then this points to issues with some other component(s) of the model.

To constraint the deposition fluxes better, we have used the MACC ozone reanalyses and in that case the deposition fluxes do not depend on ACCESS-UKCA's chemistry component because the modelled deposition velocity field used in the deposition flux calculation is solely a function of the input parameterisations Eqs. (20)–(23) and the physical component of the model (e.g., SST and turbulence).

Changes in manuscript: A summary of the above response will be included, noting that because we have used the MACC ozone reanalyses the deposition fluxes do not depend on ACCESS-UKCA's chemistry component.

(12) **Comment:** *3/1: Is this ('commonly') indeed still the case, given that Ganzeveld et al. [2009] published a process-based parameterization and model implementation some 8 years ago?*

Response: To our knowledge, all common global chemistry models described in the literature continue to use the Wesely (1989) approach (involving a constant value $r_c \approx 2000$ s m-1 for the surface resistance) for deposition to the ocean. Luhar et al. (2017) showed that the Wesely approach overestimates the deposition velocity data of Helmig et al. (2012). Ganzeveld et al. (2009) included the Fairall et al. (2007) one-layer scheme for ozone deposition velocity to the ocean in a global model, and found that compared to the Wesely approach it leads to 6% reduction in the total oceanic deposition of ozone and 0.5% reduction in the total global deposition. Luhar et al. (2017) showed that Fairall et al.'s one-layer scheme also overestimates the deposition velocity data due to flaws with its turbulence diffusivity assumption. Using better assumptions, the two-layer approach as presented in our paper describes these data much better.

Changes in manuscript: A summary of the above response can go into Introduction.

(13) **Comment:** *3/9: Ganzeveld et al. [2009] should also be cited here?*

Response: We do not think the Ganzeveld et al. (2009) reference is appropriate here because they used a one-layer scheme (of Fairall et al., 2007) and not a two-layer scheme.

Changes in manuscript: None.

(14) **Comment:** *3/28: How is the oceanic layer between the surface and 10 m depth represented?*

Response: The assumption regarding how the top oceanic layer is represented goes in the derivation of an expression for the waterside component of deposition velocity (or conversely surface resistance). This expression is implemented in the dry deposition module of our atmosphere-only global model. The global model itself does not include any explicit oceanic layer since there is no ocean model coupled. The prescribed SSTs are used in some of the input parameterisations for the waterside deposition velocity (Eq. 16) obtained using the two-layer scheme. Our scheme assumes that: chemical reactivity (or reactant) is present throughout the oceanic layer; in the top few micros of the oceanic layer ozone loss is dominated by chemical reaction (with no turbulent transfer); and in the oceanic layer below, both chemical reaction and turbulent transfer act together. We now provide a diagram (as suggested by Referee #3) to make this clearer (see our Response #2 to Referee #3).

Changes in manuscript: A new diagram as suggested by Referee #3 to be provided to make this clearer (see our Response #2 to Referee #3).

(15) **Comment:** *3/24: As mentioned earlier already, a section is needed here explaining how oceanic I- concentrations were derived and included in the modeling.*

Response: The following paragraph will be added:

Changes in manuscript: To add "This parameterisation is based on iodide data from cruises in the Atlantic and Pacific oceans covering the latitudes 50°S to 50°N, and is a function of SST (which is prescribed), so depends on location and time. It yields highest concentrations in warm tropical waters and lowest in cool waters at higher latitudes. Chance et al. (2014) examined statistical relationships between iodide and parameters such as SST, nitrate, salinity, chlorophyll-a and mixed layer depth, and found that SST was the strongest predictor of iodide in surface waters. In the study by Ganzeveld et al. (2009), oceanic surface nitrate was used as a proxy for iodide concentration. Luhar et al. (2017) used Eq. (20) in their modelling, but also presented a sensitivity of deposition velocity to the Chance at al. (2014) iodide parameterisation involving a dependence on SST." Appropriate text from our Response #5 to Referee #1 on the Chance et al. (2014) parameterisation also to be included.

(16) **Comment:** *4/14: ...considered, but a ....*

Response: Done.

(17) **Comment:** *5/1: Why 'consistent' ?*

Response: The word has been deleted.

(18) **Comment:** *5/21: …. included, and a ….*

Response: Done.

(19) **Comment:** *16/1: ….that the new ….*

Response: Done.

(20) **Comment:** *16/2: …..(2017), but unlike the latter, the new ….*

Response: Done.

(21) **Comment:** *Figure 5: As mentioned earlier, this figure nicely shows that improvements made through this work are merely nuances, while very large uncertainties and deficiencies in other areas are overlooked.*

Response: The preference for the new scheme is summed up in Lines 1–3 on Page 10: "The model performance presented in Figure 4 leads us to conclude that new scheme performs as well as the two-layer reactivity scheme in Luhar et al. (2017) but unlike the latter the new scheme does not unrealistically/artificially limit the chemical reactivity to within a fixed depth of the order of a few micrometres and has consistent asymptotic limits."

As we said in our Response #1 to a comment by Referee #1 and in the paper, there is only a small difference between the new and old two-layer schemes in terms of their performance compared to data. However, it can be said that unlike the old two-layer scheme, the new scheme leads to right results for right reasons. The present work also provides a good understanding of the impact of chemistry-turbulence interactions and how they differ in the new and old deposition schemes.

Changes in manuscript: Some changes in the relevant text to better clarify the differences between the schemes.

(22) **Comment:** *Figure 6: Ganzeveld et al. [2009], in their Figure 3a and 3b provide similar analyses for January and July. Unfortunately, they do not show annual mean analyses. However, comparing their data with this Figure 6 reveals some very large differences. While Ganzeveld et al. [2009] report the high latitude oceans exhibiting the highest ozone deposition velocities, this Figure 6 shows that the ocean deposition velocity is highest over the tropical oceans. Isn't that a rather large disagreement that should trigger an in depth analysis and discussion?*

Response: The work by Ganzeveld et al. (2009) has been adequately commented upon by Luhar et al. (2017), including their Figure 3a and 3b (see in the latter paper the last para on Page 3761, and also in the first para of Section 7.3 on Page 3762). We do not think it is necessary to repeat that exercise here.

Changes in manuscript: None.

(23) **Comment:** *18/9-10: ACCESS-UKCA then seems to differ from other models that seem to overestimate surface ozone [Parrish et al., 2014]?*

Response: As shown by Woodhouse et al. (2015) and Luhar et al. (2017) our version of UKCA in ACCESS generally underestimates observed tropospheric ozone, particularly in mid to high latitudes. The potential reasons for the model underestimation of tropospheric ozone include

inaccuracies in the emission fields of precursor species, and shortcomings in chemical or physical processes simulated in the model. However, as mentioned earlier, we determine the ozone deposition flux using the MACC ozone reanalyses and the modelled deposition velocities which do not depend on model's chemistry component. Thus model's performance for ozone is not relevant here.

Changes in manuscript: The above response to be included.

(24) **Comment:** *20/6: As mentioned earlier, this seems to disagree with the results from [Ganzeveld et al., 2009]?*

Response: That is true, and as mentioned earlier the differences with the Ganzeveld et al. (2009) have been adequately commented upon by Luhar et al. (2017).

Changes in manuscript: None.

(25) **Comment:** *20/10: Replace 'concentration' with 'mixing ratio'.*

Response: Done.

(26) **Comment:** *21/5: This really should not be called 'uncertainty' then. Maybe use the term 'error bar'.*

Response: We now say error bounds.

(27) **Comment:** *26/15-21: In this discussion about the differences between this and the previous studies, changes are attributed to a better representation of the commonly applied constant rc of Wesely's scheme, as already demonstrated by Ganzeveld et al. [2009]. Their process-based approach arrived at a global O3 oceanic deposition budget that was not that different from models using Wesely's constant rc. This, in my opinion, calls for a discussion of how these large differences between these two process-based approaches, one being extended to two layers, only considering I-, and the other one using a single layer but including more reactants including DOM, can be reconciled.*

Response: The Referee's is correct in saying that the one-layer, process-based scheme (of Fairall et al. (2007)) implemented by Ganzeveld et al. (2009) gives a global O3 oceanic deposition budget that is not too different from models using Wesely's constant surface resistance (r_c) approach. We have clarified the reason for this earlier, and it is discussed at great length by Luhar et al. (2017). The main reason is that the one-layer scheme overestimates the turbulence-chemistry interaction in the waterside viscous sublayer by assuming a turbulent diffusivity that increases linearly with depth. This assumption is not valid for the viscous sublayer. The two-layer approach fixes this problem. What this also implies is that getting the waterside turbulence-chemistry interaction correct in the model formulation is more important than including additional reactants (e.g. DOM).

The topic of DOM is discussed in detail in our reply to Referee #3.

Changes in manuscript: As mentioned earlier, a discussion on DOM based on our Response #3 to Referee #1 will be included.

(28) **Comment:** *26/29: ...., whereas that ....*

Response: Done.

(29) **Comment:** *27/26: Given my reservations detailed above in my opinion this is a rather subjective and invalid evaluation.*

Response: In our work, we developed a new two-layer parameterisation for deposition velocity that builds upon, and corrects a flaw of, the previous process-based schemes, tested it within the limitations of the available data and input information required, and used it with the 10-year MACC global ozone reanalyses for calculating deposition budgets, with a comparison of these budgets with those from other studies.

While we do not agree with Referee's comment, we have clarified various points (e.g. uncertainty, DOM, iodide, and coastal grids) raised by the referee in our replies above, which we think address this particular comment.

Changes in manuscript: None.

(30) **Comment:** *28/8: ….deposition, an increase ….burden, and an ….*

Response: Done.