Manuscript Review: Luhar et al., Revising global ozone dry deposition estimates based on a new mechanistic parameterisation for air-sea exchange and the multiyear, ACPD–2017-768.

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.

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.

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 O_3 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)-O_3 chemistry was proposed to explain these discrepancies. [*Coleman et al.*, 2010] specifically addressed the role of DOM-O_3 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.

Further, building exclusively on $O_3 + I^-$ 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 O_3 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.

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.

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.

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?

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.

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

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.

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?

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?

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

3/28: How is the oceanic layer between the surface and 10 m depth represented?

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

4/14: …considered, but a ….

5/1: Why 'consistent' ?

5/21: …. included, and a ….

16/1: ….that the new ….

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

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.

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?

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

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

20/10: Replace 'concentration' with 'mixing ratio'.

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

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 r_c of Wesely's scheme, as already demonstrated by *Ganzeveld et al.* [2009]. Their process-based approach arrived at a global O_3 oceanic deposition budget that was not that different from models using Wesely's constant r_c. 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.

26/29: …., whereas that ….

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

28/8: ….deposition, an increase ….burden, and an ….

Literature cited:

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