Response to Referee 1

S. Metzger et al.

16 August 2018

We thank the referee for her/his constructive comments and manuscript review. Please find our point-by-point reply below. The MS will be revised accordingly. We hope to have satisfactorily addressed all comments.

Reply to comments.

1. In Figure 8 the authors find that aerosol water mass concentrations are highest in the western desert of the U.S. Liao and Seinfeld 2005 and Carlton and Turpin 2013 find it is highest in the eastern U.S. where sulfate mass concentrations and RH are higher. Can the authors provide a context for this discrepancy?

Our water mass results are lowest in the western desert of the U.S. in agreement with Liao and Seinfeld 2005 and Carlton and Turpin 2013. Please note the inversion of the color scale. We apologize for the missing note in the caption of figure 8. We will include it in the revised MS.

The authors stress the importance of "aging" in their manuscript and this is not well described. I think they mean changing particle hygroscopicity with time but this is not clearly stated. Also, "aging" is a not a precise term. Do they mean increased oxidation due to longer OH exposure in the atmosphere, and the subsequent changing chemical composition important water uptake?

We agree, "aging" is a not a precise term. We will replace it throughout the work with "chemical aging" and add that this indeed changes the (bulk) particle hygroscopicity with time. In our model, the uptake of inorganic acids on bulk compounds, and the associated neutralization reactions and water uptake, occurs over time, i.e., during aerosol transport. This will be clarified in the revised MS.

The authors do not make a compelling case in the introduction for their work and I found this confusing. Below I provide comments that think help address my concerns and I hope the authors find them useful.

We highly appreciate these constructive comments.

The introduction is not directly linked to the premise that aerosol water is crucial. I find it difficult to understand why the introduction starts with the importance of desertification and subsequent dust emissions to properly describe AOD when their title and abstract focus on aerosol water. Do the authors mean to say that even in arid regions, AOD is not properly described in models unless water uptake is considered? That is a compelling argument and would help to connect the introductory desert discussion with aerosol water.

Yes, indeed, even in arid regions the AOD is not properly described in models unless water uptake is considered. The reason is the interaction of air pollution with e.g., mineral dust (the issue we recently have addressed with Abdelkader et al, 2015). The missing link is the uptake of acids on mineral dust as it can alter the ability of bulk dust to take up water vapor even at a very low ambient humidity – in case of condensing hydrochloric acid, calcium chloride can be formed over time which can cause water uptake at a relative humidity as low as 28%.

Sentence 1: "providing realistic projections of climate change is one of the most difficult tasks of climate modelers..." I would state "Providing realistic projections of climate change is difficult due to many unknowns and large uncertainties . . . " As written the sentence is awkward, the authors seem to say, conducting climate simulations is the hardest thing for climate modelers to do.

Yes, we agree. We will change the sentence to "Providing realistic projections of climate change is difficult due to many unknowns and large uncertainties that still exists ...".

The authors might not be aware of this paper using actual measurements of particle-phase ions and meteorology coupled with ISORROPIA-II to calculate aerosol water to better connect surface particle mass measurements to satellite AOD by Nguyen in Geophys. Res. Letts.:. I think it would help with their argument to link the importance of aerosol water with AOD and then subsequently radiative forcing calculations important for climate modeling efforts.

Yes, we agree. We will include Nguyen et al. in the revised MS as their work helps to link the importance of aerosol water with AOD.

Page 4, Line 13: is dry deposition based only properties of the surface? Do different chemical species all deposit at the same rate?

This is a typo. We will change it to "Dry deposition fluxes are calculated

as the product of the surface layer concentration and the dry deposition velocity, which reflects the efficiency of the transport to- and destruction at the surface (Ganzeveld et al., 2006)".

Page 4, Line 9/25: Why are some subroutines listed together, while others are separated out? For example CLOUD, CVTRANS, JVAL, TROPOP, H2O, ORBIT, and RAD are listed below in a similar fashion.

The submodel listing on line 9 was left accidentally in the MS. We will clean it up in the revised MS.

Page 4, Line 16/17: what do "...water isoprene concentration" and "methanol water deposition. . . "? Perhaps the authors mean isoprene concentration in ocean water? Do they mean dry deposition of methanol to water? I read this sentence multiple times and I am still not sure.

The sea-air exchange submodel (AIRSEA) calculates the transfer velocity for certain soluble tracers (e.g., DMS, isoprene, methanol). This will be clarified in the revised MS.

Page 4, Line 20: Does "Our chemical mechanism for the troposphere is similar to the one used in poz" mean the mechanism is the same as used in Pozzer et al., 2006, cited earlier? Sometimes the authors write "poz" and "Pozzer". I am not sure if they mean the same thing.

This is a typo. "poz" should read Pozzer et al., 2012 (Atmos. Chem. Phys., 12, 961-987, www.atmos-chem-phys.net/12/961/2012/). This will be corrected accordingly and the reference added in the revised MS.

Page 5, last sentence and continuing to the next page: "It was shown by Metzger et al. (2016b) that the ν_i -approach allows to analytically solve the gas-liquid-solid partitioning and the mixed solution water uptake by eliminating the need for numerical solutions . . ." Is the Metzger approach not a numerical solution?

Again, a typo. "solutions" should read "iterations".

Page 16, line 6: take out "only"

Yep, "only" will be taken out in the revised MS.

The text regarding "Kindly" provided emissions seems like language that should be in the acknowledgements.

Also true; parts of the sentence will be moved to the acknowledgements.