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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Sensitivity of modelled sulfate radiative forcing to DMS concentration and air-sea flux formulation" *by* J.-E. Tesdal et al.

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This paper presents sensitivity runs using a GCM with an explicit aerosol scheme in order to quantify the impacts of using different seawater DMS climatologies and sea-air flux parameterizations. This study adds to the numerous studies in literature which have had similar aims at understanding the effect of DMS and impacts of changes in the DMS flux.

Unfortunately, in its current form, the manuscript does not add significantly to the literature and hence I cannot recommend its publication. Most of the work presented here has been done before and simple sensitivity studies do not add much to our current knowledge on the effect of DMS, especially when there is no comparison done with

observations.

We thank Referee #2 for his/her constructive comments. We agree that we did not sufficiently clarify the advances made in our paper. In particular, our study adds to existing literature by assessing the relative importance of spatial and temporal variation of DMS fluxes to atmospheric properties as compared with the magnitude of the global total flux. No other systematic analysis of the relationship between spatio-temporal distribution of DMS emissions and aerosol radiative effects on climate has been published to date, despite large uncertainties in specified DMS data sets in models. We recognize, however, that this motivation was not clear in the previous version of the study. In order to clarify that this is our objective, we have added some discussion of this point to the Introduction, emphasizing how the study adds to the existing literature. We also note that the Discussion paper has already been cited by Mungall et al., "Summertime sources of dimethyl sulfide in the Canadian Arctic Archipelago and Baffin Bay", doi:10.5194/acpd-15-35547-2015. (none of the authors of the present paper were involved in that study.)

The manuscript has been revised to address the referee's points and to clarify how the study adds to existing literature. Discussion of all of the referee's points follow:

1) The suitability of the model to study the impact of DMS on aerosols, cloud properties and radiation changes has not been demonstrated. Have the authors compared the model results to any observations? Several climatologies of sulphate are available, and locations with atmospheric DMS and SO2/H2SO4 observations can be also compared to show that the model is indeed close to the observations or highlight the major shortcomings of the model.

A similar concern was raised by the other referee and we would like to repeat our

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response: We have added a comparison of simulated and observed sulfate concentrations over the ocean in order to address the concern about a lack of model validation, with a focus on regions that are expected to be strongly affected by DMS emissions. These results provide evidence for realistic simulations of atmospheric DMS sources and aerosol removal processes in the marine atmosphere. In addition, the relationship between sulfate aerosol concentrations and cloud microphysical properties in simulations with CanAM and observations were previously investigated by Ma et al. (2010). Simulated aerosol concentrations, cloud microphysical properties, and unpublished results for sulfate aerosol forcings are all well within the range of uncertainty. So we have confidence that simulated impacts of DMS emissions on sulfate aerosol radiative effects on climate are meaningful.

2) What is the reason behind including sensitivity experiments based on seawater climatologies K99, K00 and L10? L10 is an updated version of the K00 and K00 climatology. It seems unnecessary to include all three considering the L10 climatology is an upgrade and covers all the data that went into the K99 and K00 climatologies. This would save a lot of discussion, which could be focused on other important features of the results. It is important to note that they are not different climatologies, but rather upgrades and hence only the latest should be used.

We agree that the L10 climatology is an update to the K99 and K00 climatologies, and thus L10 comprises the K99 and K00 data and is more reliable. However, by comparing these climatologies, we are able to demonstrate the need to resolve uncertainties in the DMS concentration fields. By demonstrating the sensitivity of the climate system to global DMS flux, we demonstrate the importance of updates like L10 (as did Mahajan et al., 2015), and the need for continued improvement to estimates of DMS concentrations. Furthermore, K99 and K00 were used in previous studies (e.g., Thomas et al, 2010; 2011; Woodhouse et al., 2010; 2013), and only recently published studies use

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L10. So, by including K99 and K00, we allow for the comparison of our results with other modeling studies. In order to clarify this to the reader, we have added the above argument to Section 2.2 in the revised manuscript.

3) Also, is there a specific reason the authors include the AN01 seawater DMS climatology? This climatology, in particular, does not compare well with observations of seawater DMS, which shows that estimating DMS using global fields of chlorophyll, nutrients and light is not an accurate method considering the complexity in the oceanic DMS cycle. Additionally, a similar exercise has been by the authors in another paper (Tesdal et al., Environ. Chem., 2015).

We agree that the AN01 seawater DMS climatology does not compare well with observations, as is the case for all empirical models analyzed by Tesdal et al. (2016). However, AN01 produces global mean emissions similar to L10, and while it is a bit of an outlier among the empirical models in terms of its spatial pattern, it is the most skillful relative to observations by some measures. AN01 is a useful addition in our sensitivity study, given the study's focus on the relative importance of spatial and temporal variability compared with global annual mean flux. AN01 produces global mean emissions very similar to L10 but its spatial and temporal pattern is very different. This point has been made in the revised manuscript (Section 2.2)

4) Lana et al., 2011 did a comparison of the total flux of DMS using different climatologies, similar to that presented in table 3. A recent publication, Mahajan et al., JGR, 2015 (which is not cited) has done similar comparisons to those presented in this paper, but in further detail including spatial features, quantifying the impact of a change in the DMS climatology. Woodhouse et al, ACP, 2013 have studied the effect of changing the DMS flux on regional scales. This raises the question as to what is new in this study. The differences by changing the sea-air parameterisations are not surprising,

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while changes due to different climatologies have been studied before using different models.

In addition to investigating the sensitivity of radiative effects on climate to different DMS climatologies, our study also focuses on a number of questions not answered by other studies. As mentioned above, the study analyzes the relationship between spatio-temporal distributions of DMS and aerosol radiative effects and addresses the question how the spatial or temporal distribution of DMS flux influences the climate system compared to the global annual mean flux. It also presents evidence that the inclusion of air-side resistance in sea-air parameterizations is a significant factor in calculating emissions and aerosols, which was remarked upon by the other referee. The reference to Mahajan et al. (2015) is greatly appreciated, and we have mentioned the results from that study in the revised manuscript. However, while Mahajan et al. (2015) consider the effect of spatial features of DMS climatologies on atmospheric properties, they do so only in the context of the differences between the K00 and L10 climatologies, and not as a comparison of the relative importance of spatial variation to atmospheric properties in general (nor do they consider the effect of temporal variation). Further, Lana et al. (2011) provide comparisons of total flux but do not include information on any other atmospheric burdens, included in Table 3 of this study. Woodhouse et al. (2013) did not look at radiative effects on climate (as this study does) but rather focused on changes in cloud condensation nuclei (CCN).

5) Why do the authors focus only on global and annual means? Considering the large spatial and seasonal differences between the climatologies, one would expect large regional difference (e.g. Mahajan et al., JGR, 2015) and these features would be smoothed out by taking global and annual means. This could be a reason for the linear dependence of the results, when in reality regions with different background aerosol loading, or relatively lower anthropogenic SO2 emission would show highly nonlinear

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dependence considering the multistep chemistry-physics interactions to go from sea-water DMS to cloud properties.

The focus of our analysis is on impacts of DMS emissions on the global and annual mean radiation budget. Global and annual mean radiative effects of aerosols can be used to estimate changes in global energy budgets and temperature changes in the climate system. They thereby provide a simple basis for quantifying aspects of the climate response to imposed forcing agents, especially global mean temperature, and hence are widely used in the scientific community (Myhre et al., 2013).

We did not attempt to analyze regional and sub-annual variations. These are much more difficult to analyze in a statistically robust way than global and annual mean effects owing to natural variations in regional climate processes. In general, relationships between variations in radiative effects and temperature responses are highly non-linear and non-trivial in the climate system. They are addressed in only relatively few studies, often under highly idealized conditions. We therefore consider an analysis of these relationships to be well beyond the scope of this study.

6) Does the model include ternary nucleation of H2SO4 and MSA with organics, which have recently been estimated to play a major role in the conversion of DMS to aerosols? (Dawson, PNAS et al., 2012; Riccobono et al., Science, 2014).

The model does not employ any parameterization of aerosol nucleation. Only bulk aerosol parameterizations are used, as described in the paper, and in more detail by von Salzen et al. (2013). The bulk aerosol scheme is a well-documented approach which produces results that are relatively straightforward to analyze. Similar bulk schemes have been widely used in different models in the past and are used in some CMIP5 models. On the basis of the few available studies that discuss results

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from different models with bulk and microphysics schemes, we do not see evidence for considerable improvements in radiative forcing estimates based on simulations with microphysics schemes relative to bulk schemes (Schulz et al., 2006; Koch et al., 2009; Quaas et al., 2009). The manuscript has been revised to make this point (see end of Section 2.1 in revised manuscript).

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