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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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The manuscript by Tesdal et al. uses a global atmospheric GCM, which includes a representation of aerosol chemistry and processes, to evaluate the effect of various DMS emission estimates on sulfate burden and the subsequent radiative impact. The study builds incrementally on pre-existing work. The manuscript is well written and structured, and the content is appropriate for ACP. However, before publication I recommend that the authors consider the points below, particularly about evaluating the model's ability to reproduce observed quantities.

We thank Referee #1 for his/her insightful comments. In particular, we appreciate the

point that the article did not sufficiently address the issue of model assessment, to which we have responded by adding a new subsection comparing simulated sulfate concentrations with observations. We have revised the manuscript to address all of the referee's points and to clarify our terminology in line with the suggestions made. Specific responses to all of the referee's points follow:

Major comments

The suitability of the model for the study is not demonstrated. If we are to have faith in the results of the model's response to different perturbations (as part of the sensitivity study), we need to first know that the model is capable of representing the quantities in question. Has the model ever been evaluated against observations of DMS, SO2, sulphate, CCN, CDN, radiation...? Particularly in regions which are dominated by marine aerosol (Southern Ocean). Does the model simulate the observed seasonal cycles of these quantities adequately?

The model uses a basic (bulk) representation of aerosol which does not fully simulate aerosol microphysics (e.g. competition between condensation and new particle formation, coagulation / interaction between different aerosol species etc). Bellouin et al. (2013, ACP 13: 2027) find stark differences between a bulk and a microphysical aerosol scheme in how they simulate aerosol direct and indirect effects, including the response to a DMS perturbation. Some discussion of the limitations of the aerosol scheme is therefore necessary, and would be particularly useful when connected to an evaluation of model skill vs observations. Calculation of the 'CCN sensitivity' for comparison with the equivalent values calculated in Woodhouse et al., (2010) and others would also be valuable.

We would like to respond to issues raised in the first and second paragraph together

since they are related.

We have added a comparison (new Section 3.1) of simulated and observed sulphate 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 sulphate 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 sulphate aerosol forcings (unpublished but assessed as part of the model development process) are all well within the range of uncertainty so that we have confidence that simulated impacts of DMS emissions on sulphate aerosol radiative forcings are meaningful. This is discussed in the revised manuscript (Section 3.1).

We agree that bulk and microphysical schemes typically tend to be very different in terms of the processes that are considered and results for radiative forcings. According to our own experience with the CCCma bulk and aerosol microphysics schemes, large differences exist for some of the results. However, we consider the bulk approach to be appropriate and perhaps even desirable in the context of the current study. 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. Based on the few available studies that are based on results 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). The Discussion section of the revised text also explicitly raises the point that we consider

a single AGCM in this study, and neglect uncertainties in the radiative effect of DMS based on different model formulations.

Unfortunately, we are unable to calculate a "CCN sensitivity" for comparisons with results from Woodhouse et al. (2010) owing to the fact that the model does not simulate CCN concentrations. The simple parameterization of aerosol indirect effects in the model supplies cloud droplet number concentration as a function of sulphate concentration.

I was surprised that greater prominence was not given to the air resistance result and the impact that can potentially have on DMS flux / aerosol. That seems like an important conclusion, and the biggest contribution.

We agree that this is a significant result from our modelling exercise. We have emphasized it further in our conclusion.

The word 'forcing' is used incorrectly in the manuscript (including in the title). Radiative forcing is the difference in radiation budget between two time periods (e.g. preindustrial and present day). I think the term 'radiative effect' is what is meant, as the manuscript in question doesn't consider different time periods (except in one paragraph on page 23948: line 25).

In the paper, radiative forcings were defined as net changes in the TOA energy balance due to changes in sulfate aerosol concentrations which are produced by DMS emissions and subsequent oxidation of DMS in the atmosphere. A distinction between present-day and preindustrial time periods is different than our analysis of the impacts of different DMS emission data sets on climate. We believe that our approach is con-

sistent with the definition of radiative forcings given in the IPCC AR5 report:

"RF is the net change in the energy balance of the Earth system due to some imposed perturbation. It is usually expressed in watts per square meter averaged over a particular period of time and quantifies the energy imbalance that occurs when the imposed change takes place. Though usually difficult to observe, calculated RF provides a simple quantitative basis for comparing some aspects of the potential climate response to different imposed agents, especially global mean temperature, and hence is widely used in the scientific community. Forcing is often presented as the value due to changes between two particular times, such as pre-industrial to present-day, while its time evolution provides a more complete picture."(Myhre et al., 2013)

We generally agree that 'forcing' has been used incorrectly in the original text and removed the term "radiative forcing" as per the referee's request. In order to account for the ambiguous usage of the term "radiative forcing" in previous literature and to clarify what we mean by "radiative effect", we added the following text (Introduction):

In the following, net changes in the energy balance at the top of the atmosphere due to changes in concentrations of DMS-derived sulfate will be referred to as the radiative effect, which is sometimes also referred to as radiative forcing.

In many places, however, we did not feel it was necessary to substitute the one phrase for the other. We rather modified the text wherever the term appeared, so as to include enough detail about the underlying processes that the meaning should be clearer than in the original text, with the term "radiative forcing" removed. In one place we retained the term radiative forcing, because it refers to specific outputs of particular models, which are documented in the literature cited.

It would be very informative to see the spatial responses (change in DMS, sulphate,

CDN, radiation... as a map) to the DMS perturbations. Presenting this information would make it much easier to compare and interpret the results against previous work.

Since our work is primarily concerned with global changes, we do not feel it is within the scope of the current contribution to include these maps in the main text. But we agree that these can be very informative for the interested reader, so we have added them as supplementary material.

Minor comments

These questions have helpfully pointed toward needed clarification, and we have added further detail to the corresponding sections.

Page 23939, line 19: How is the ensemble created? Varying initial conditions?

Following sentence has been added:

Each ensemble member uses the exact same model configuration, but in each a different seed was used in the random number generator used in the radiation code.

Page 23939, line 22: 'realizations' = 'ensemble members'?

Yes. We changed 'realizations' to 'ensemble members'.

Page 23939, line 22: It would be useful to state what the spread was between the realizations, for direct comparison to the response from altered DMS emissions.

Please note that the spreads of the ensemble members for each model run are also shown in Figure 2 and 3. We added the following in the result section:

The largest ensemble spread in DMS emissions among the simulations is less than $0.06\,\mu\mathrm{mol\,m^{-2}\,d^{-1}}$, which is negligible compared to the overall range of DMS emissions of the different model runs ($3.15\,\mu\mathrm{mol\,m^{-2}\,d^{-1}}$).

Page 23944, line 28: Has the statistical significance of the changes been calculated?

Statistical significance is determined from Figure 9 by comparing the mean differences among the model runs with the corresponding error bars. We have added a reference to Figure 9 in the revised text and added the following to the caption of Figure 9 (Please note that Figure 9 in the revised manuscript is Figure 7 in the previously submitted version.):

To derive the error estimates, all treatments (control, temporally invariant, spatially uniform, and no air-side resistance) were pooled after their separate means were removed; error bars are $\pm$ two standard deviations of the pooled data ($n = 12$). Statistical significance is determined by comparing the mean differences among the model runs with the corresponding error bars.

Page 23945, line 7: Discussion or presentation of the spatial patterns would be useful in understanding the increase in SO2 but decrease in sulphate. Are any further model diagnostics available to probe this further? It's an interesting outcome and one which it would be useful to understand.

As discussed above, we consider a detailed analysis of spatial patterns to be beyond the scope of the current contribution, but the relevant maps are presented as supple-

mentary material.

Page 23948, line 20: Vallina et al., 2007 don't go as far as calculating the atmospheric/ climate response.

We agree. Vallina et al. (2007) mainly looked at the response of DMS given a change in climate. With "weak effect" bias we are referring to a link between DMS and climate, both as the influence of DMS on climate and vice versa. We acknowledge that this was not clear in the original manuscript and the text has been changed accordingly:

Previous studies have found a relatively weak link between DMS fluxes and climate (e.g., Kloster et al., 2007; Vallina et al., 2007; Woodhouse et al., 2010).

Page 23948, line 20: Wouldn't a low DMS flux result in a lower background aerosol concentration, thus making the system more sensitive to DMS perturbations...?

The referee's question points to a lack of clarity in the wording, which we have revised to prevent misunderstanding. The effect described by the reviewer could conceivably occur, but it would require a large change in the relative flux, and there is no particular reason for that not to occur in cases with a higher 'baseline' flux as well. Because the radiative effect on climate is more or less linear with the total global DMS source, perturbing a DMS concentration field that has a higher global total flux will generally have a greater effect than perturbing a field with a lower total flux.

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