

Interactive comment on “Sensitivity study of dimethylsulphide (DMS) atmospheric concentrations and sulphate aerosol indirect radiative forcing to the DMS source representation and oxidation” by O. Boucher et al.

J. Hjorth (Referee)

JENS.HJORTH@JRC.IT

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General Comments

The paper investigates how the modelled atmospheric concentrations of DMS and of sulphate aerosol depend on the use of different descriptions of the oceanic DMS climatology, the parameterisation of the sea-air transfer of DMS and the field of atmospheric oxidants (including aqueous phase chemistry). It also addresses the question of how the estimation of the indirect radiative forcing of anthropogenic sulphate aerosol is influenced by the uncertainties on the DMS-derived natural background aerosol.

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I believe that the discussion by the authors of their results is very appropriate: The potential importance of various parameters is highlighted, but the conclusions do not go further than what the uncertainties on the available information allow. For example, the large potential impact of BrO is explained, but it is also said, that little is known about the actual concentrations of BrO in the atmosphere. Thus it is made clear, that this really is a 'sensitivity study', that has the scope to give an idea about the impact of important uncertainties in our understanding of emissions and chemistry of DMS on the estimation of sulphate aerosol concentrations and of indirect radiative forcing. The paper finally makes recommendations regarding future research efforts that would help to reduce the uncertainties that, according to the results of this study, appear to be important. I find that the paper is well written and it appears to be scientifically sound. I believe that it should be published on ACP after a few minor corrections and amendments that I will discuss in the following.

The paper is generally clear, but the title is an exception from that! It is, in my opinion, awkward and I would suggest to rephrase it.

The influence of DMS on the indirect radiative forcing by anthropogenic aerosol is treated by the paper and mentioned in the title, but not addressed neither in the abstract and nor in the 'Introduction' part of the paper. I think that this is a mistake and suggest that the authors include a sentence about their conclusion regarding indirect radiative forcing in the abstract and that they also give an introduction to this issue.

Specific comments

P. 1183, last paragraph: The authors may also mention the paper by Campolongo et al., which also comes to the conclusion that aqueous phase oxidation of DMS appears to be important (Campolongo, F., Saltelli A., Jensen, N.R., Wilson, J., and Hjorth, J., 1999, The role of multiphase chemistry in the oxidation of dimethylsulphide (DMS). A latitude dependent analysis, Journal of Atmospheric Chemistry, 32, 327-356).

P. 1184/1185, paragraph 2.1.: I would suggest to add a few lines, summarising how

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the oxidation of SO_2 is treated in the model (particularly the heterogeneous oxidation pathways), since this is obviously also relevant for the outcome of the present study.

P. 1185, 2nd paragraph: H_2O_2 is an important species because it oxidises SO_2 to sulphuric acid in droplets. Here it is said, that its concentration is calculated from prescribed HO_2 concentrations and H_2O_2 photodissociation rates. However, according to the description of the same model by Bouchet et al. (2002), the treatment of H_2O_2 is more complex and involves also wet and dry scavenging as well as depletion due to reaction with SO_2 . Could the authors please clarify this point?

P. 1188, l. 2: As already pointed out in another on-line comment to the paper, there is an error in this line (EXP6 does not include BrO chemistry).

l. 11: The value of the gas phase rate constant for the reaction $\text{O}_3 + \text{DMS}$, determined by Martinez and Herron in 1978, is an upper limit only. Thus also the estimate of the contribution of this reaction to the oxidation of DMS is an upper limit, and this should be made clear in the text.

P. 1189, l. 8: The statement, that a range of 10 to 40 Tg S per year is 'usually accepted for DMS emissions', is too vague! The DMS-fluxes calculated in this paper are about twice as high as the recent estimates cited by the authors; this seems to be a quite significant difference, and I think it may be relevant to discuss the reasons for this more in details. The authors should also mention the estimates of DMS emissions that point to higher values, in order to qualify their statement about the 'usually accepted' range.

P. 1191: It would be useful to have a brief indication of where the Albatross Cruise was performed (e.g. by inserting a map of the cruise track in Fig. 6).

P. 1193, line 4: Rather than 'evidence for the existence of BrO' I would suggest to write 'evidence for the importance of BrO'.

Paragraph 3.5: When discussing the sources of DMSO and the uncertainties related to these it should also be taken into consideration, that there is a significant uncertainty

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on the yield of DMSO from the OH-initiated oxidation of DMS.

P. 1195, conclusion no. 4: As I have mentioned above, the contribution of the gas phase reaction of ozone with DMS is only an upper limit.

Technical corrections P. 1184, l. 5: point -> points P. 1187, l. 13: acetone is not a hydrocarbon P. 1187, l. 18: There is no information about where the data on emissions from biomass burning and lightning comes from. P. 1209, Fig. 5: The data seem to be monthly averages. This should be explained in the figure caption P. 1213, Fig. 8: The figure caption is not clear. Is the figure showing the ratio between the annually averaged zonal and global DMS mixing ratios?

Not being a native English speaking person I hesitate to suggest language-corrections, particularly regarding the use of prepositions (I may well be wrong myself!). However, I would suggest to the authors that they take care, that their paper is checked for such minor errors. E.g. on page 1193, it is said that 'DMS is oxidised in DMSO in high latitudes'. I would say 'oxidised to DMSO at high latitudes', but I am not quite sure about this!

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