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4, S3940-S3943, 2004

Interactive Comment

# Interactive comment on "Parametric sensitivity and uncertainty analysis of dimethylsulfide oxidation in the remote marine boundary layer" by D. D. Lucas and R. G. Prinn

D. D. Lucas and R. G. Prinn

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Comment 1: "The use of such a model should thus be justified more rigorously, indicating how all the processes involved in the DMS cycle of the remote marine atmosphere are well-represented and what are their limitations and the limitations of the chosen assumtions: for instance, what is the relevance of pm?"

Reply to Comment 1: We appreciate the reviewer's concerns about the limitations of our box model. The main justification for using this simple model structure is that it is computationally efficient, but still gives concentration profiles consistent with observations. Also, similar DMS models have been used previously by other researchers.

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These justifications and the limitations of the box model are now noted carefully in the last two paragraphs of Sect. 2.1.

Comment 2: "What is the reference for NO and NO3 concentrations?"

Reply to Comment 2: We have improved the discussion of the NO and  $NO_3$  concentrations. For NO we now clearly state that NO levels were below detection limit and assume a value of 1 ppt. For  $NO_3$  we previously assumed a cycle similar to the cycle used in Chen et al. (J. Atmos. Chem., 2000). Now, we have improved the treatment of  $NO_3$  by diagnosing the concentration assuming that the primary sinks are the nighttime reaction with DMS and daytime photolysis. This is described in detail in Sect 2.2.

Comment 3: "Another point concerns the use of the two analytical methods. It is difficult for the reader to understand why these two methods have been chosen: are they really suitable for the described physical system and are there other existing? This should be detailed at the beginning of section 3."

Reply to Comment 3: We have made extensive revisions to clarify our choice for using these two methods. The revised manuscript now divides the discussion into the local method (DIM) and the global method (PCM). The advantages of each are also now clearly stated.

Comment 4: "Moreover, some properties of the methods are scattered in the text (end of subsection 3.2.1: In this sense..., end of subsection 4.2: expensive to compute..., first paragraph of subsection 5.1: computationnaly...) instead of being synthetically described. I think a brief comparison of the two methods describing their advantages and inconvenients (for instance, their computational costs) is also necessary in section 3."

Reply to Comment 4: We agree strongly with this recommendation. Accordingly, we have re-organized all of the relevant methodology into just two sections of the revised manuscript, the local sensitivity analysis in Sect. 3.1 and the global analysis in Sect.

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Comment 5: "Introduction: the importance of MSA and H2SO4 in the formation of aerosols could be pointed out here. This will also justify why only concentrations of DMS, SO2, MSA and H2SO4 are discussed in the paper."

Reply to Comment 5: We focus on these four species because they tend to be the most commonly observed gas-phase DMS-related species during measurement campaigns. We now state this clearly at the end of the Introduction. We also now state that  $SO_2$ , MSA, and  $H_2SO_4$  are commonly observed because they can modify or form aerosols.

Comment 6: "2.1.3: how are fast reacting sulfur-based radicals calculated, photochemical equilibrium?"

Reply to Comment 6: Photochemical equilibrium is not assumed for the radicals. The continuity equations of all of the sulfur-containing species are solved with the stiff ODE solver. This is now clarified in the revised text (2nd paragraph, Sect 2.1).

Comment 7: "4.1: eq. 6 does not come from derivative of Eq. 1 but Eq. 5."

Reply to Comment 7: We have clarified the derivation of the first-order local sensitivity ODEs (see Eqs. 3 to 6 in the revision).

Comment 8: "5.1: in relation with point 2 in the general discussion above, it should be emphasized why three different methods have been used in order to estimate the PDFs moments (especially DIM-S)?"

Reply to Comment 8: To avoid unnecessary confusion, the revised manuscript now estimates the PDF moments using only PCM. DIM is not used for the reasons described in Sections 4.2.3 and 4.2.4 of the revised manuscript. Moreover, the Monte Carlo method (MCM) is no longer used because PCM adequately approximates the true model.

Comment 9: "5.1.1: the PCM moments are calculated assuming the variables are

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independant. This should be justified for the used concentrations."

Reply to Comment 9: A paragraph has been added to the end of Sect. 2.3 in the revised manuscript that discusses the assumption of independent variables.

Comment 10: "6.2.4: it will be also interesting to discuss the shift of sensitivity from morning to evening (for example SO2)."

Reply to Comment 10: We have improved the discussion of the time variations to the first-order local sensitivity coefficients, including  $SO_2$ . This discussion is included in Sect. 4.2.1 in the revised paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 6379, 2004.

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