Atmos. Chem. Phys. Discuss., 1, S47–S50, 2001
www.atmos-chem-phys.org/acpd/1/S47/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modelling the contribution of sea salt and dimethyl sulfide derived aerosol to marine CCN" *by* "Y. J. Yoon and P. Brimblecombe"

Anonymous Referee #1

Received and Published: 3 October 2001

Review of
"Modelling the contribution of sea-salt and dimethyl sulphide derived aerosol to marine CCN"
Yoon and Brimblecombe

General: This manuscript follows in a long line of studies aimed at elucidating the source of CCN in the marine boundary layer due to the global importance of marine stratiform clouds to the global albedo along with their high susceptibility to perturbations in CCN concentrations. The current study focuses more on the contribution of sea-salt aerosol to the CCN population when compared to previous studies; however, it is somewhat disappointing to find that the model used comprises only most basic

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aerosol representation and aerosol dynamics, not to mention meteorological representation, particularly after the open debate into the usefulness of this model (i.e. low size resolution) in J. Geophys. Res. during the mid 1990s.

While I am disappointed with the limited methodology used, I believe it is important to stress the importance of sea-salt CCN in this environment, since for a variety of reasons, it is been more or less overlooked from the mid-80s to the mid-90s and, in recent years, there has been significant evidence relating to its importance in cloud nucleation along with chemical cycling of S and I species in the marine boundary layer. For this reason, I find the manuscript potentially suitable; however, the very limited model infra-structure must be compensated for by a rigorous discussion of processes omitted.

In particular, I have the following comments which should be commented on in a more detailed "Discussion" section.

Given the history of debate and conflicting theories on this particular topic, I find the Introduction too short and very uninformative.

For example: in what way has the assumption that sea-salt cannot act as CCN be challenged? How did Blanchard and Cipriano or O'Dowd and Smith challenge this?

Similarly: "Moreover, the nucleation ability of DMS derived acid in the MBL is still under debate(Katoshevski et al., 1999, Pirjola et al., 2000)"

What was the issue of the debate? These two unresolved issues are central to the whole manuscript and consequently warrant more discussion. Perhaps the issues could be outlined more clearly in the Introduction and discussed in detail in the Discussion section?

The effect of the following processes/parameters, or lack off, should be discussed in detail, if not acknowledged:

(1) The limited size resolved spectrum used (perhaps refer to discussions in JGR between P&R and Raes et al.)

(2) The total lack of coarse mode sea-salt aerosol: O'Dowd et al., 1997, illustrated that the marine aerosol condensation sink was dominated by the super-micron sized

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particles which are totally omitted from this model, therefore, the concentration of sulphuric acid could potentially be greatly over-estimated due to the underestimated condensation sink. What effect will this omission have on the results.

(3) There is no mention of nucleation rate used other than what was adapted from P&R. They have argued that this is one of the most important factors controlling the CCN production rate. What actual value was used? I presume that the nucleation rate used is some function of a nucleation rate tuner? Pirjola et al elude to the fact that the use of a nucleation rate tuner gives whatever results one wishes. They instead found that using a parameterised nucleation rate, nucleation was most unlikely in the clean MBL. This issue needs to be discussed in detail, again, in a rigorous discussion section.

(4) The modeled average values of DMS are very high. Despite the references included, can sustained concentrations of this magnitude be realistic? Pirjola et al found that the production of particles could be possible of DMS concentrations of the order of 100 ppt were sustained for many hours; however, they considered that this was unlikely. More discussion is needed.

(5) How do the modeled sulphuric acid concentrations compared to measured values?

(6) The model seems to essentially omit the aqueous phase oxidation of DMS-derived SO2, particularly in cloud, although I seem to recall that some parameterisation was included for cloud free oxidation in sea-salt in the P&R model. More recent studies have shown that more than 90% of the SO2 oxidation can occur in sea-salt, thereby reducing the production rate of Nss-CCN [e.g. O'Dowd, CD, J.A. Lowe, and MH Smith, Coupling sea-salt and sulphate interactions and its impact on predicting cloud droplet concentrations, Geophys. Res. Letts, 26, 1311-1314, 1999]. This should at least be mentioned as another influential process.

(7) The relevance of the Cape Grim CCN data should be highlighted in the context of the modeled CCN and nucleation mode concentrations. Supersaturations of 0.23% represent more the accumulation mode size while supersaturations of $>$ 1% represent

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more the nucleation mode sizes (in the context of this study, more realistically representing the Aitken mode sizes). This should be mentioned in the relevant section.

In summary, the above issues should form the material for a detailed Discussion section (between section 3.4 Comparison with other results and section 4 Conclusions). Putting the limited model study into context with respect to these other processes is important to help the reader to better understand the processes which determine the contribution of sulphate and sea-salt CCN in the marine boundary layer. Such important revisions should make the manuscript suitable for publication.

Interactive comment on Atmos. Chem. Phys. Discuss., 1, 93, 2001.