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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Tropospheric distribution of sulphate aerosol mass and number concentration during INDOEX-IFP and its transport over the Indian Ocean: a GCM study" *by* S. Verma et al.

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Response to the Interactive comment on "Tropospheric distribution of sulphate aerosol mass and number concentration during INDOEX-IFP and its transport over the Indian Ocean: a GCM study" by S. Verma et al.

Anonymous Referee 1 Received and published: 27 March 2005

Response to the reviewers

We would like to thank the reviewer for his/her insightful comments that were helpful in

improving substantially the presentation and contents of the revised manuscript. We hope to have addressed appropriately all issues raised by him/her.

Point-to-point response:

Major points

1. It appears to me that the new sulfur model is generally well-designed, though I have several concerns outlined below. One general point that is not clear is the motivation behind developing a new sulfur chemistry model. What advantages and disadvantages does it have compared to the independent models developed by Boucher and Hauglustaine? The abstract says "the originality of this module is its ability to predict particle mass and number..."; the M 7 model already does this for not only Aitken and accumulation with sulfate, but also for nucleation and coarse modes for all the main chemical components of the aerosol, so it's not clear what is particular about the new model described here. M 7 and other models need to be cited here, and contrasted briefly with the new model.

Authors' Response: The Indian Ocean Experiment (INDOEX) emphasized efforts for advancing the development of global chemistry models, including refinements to chemical/physical parameterizations and improving the numerical schemes. This work is much in line with this philosophy of INDOEX, hence provides the main motivation (Section 1, highlights this point). The aerosol module (of EPA, Model-3) in the new sulphur model provides a size-segregated, two-moment distribution of aerosols that undergo processes like nucleation, condensation, coagulation and interaction with clouds. The model also has a provision to provide nucleation and coarse mode along with all other chemical component; it is however not done in the present manuscript as a first step towards understanding of sulphate aerosol forcing. As suggested by the reviewer and in view of the recent publication of Stier et al. (2004), the following statement has been added in the abstract: "This module has the ability to predict particle mass and number concentration for the Aitken and accumulation modes."

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The following statement has been removed from the abstract: "The originality of this module is its ability to predict particle mass and number concentration." The reference to the suggested article on M7 has been included in the revised manuscript. The unique aspects of the model from the independent models developed by Boucher et al. (2002) and Hauglustaine et al. (2004) have been briefly described. The other relevant model studies are also cited there in order to contrast briefly this model. The portion where the description is modified and the references are cited, reads as follows (added in the revised manuscript towards the end of section 1),

The main motivation of this study is to reproduce the observed variations in the sulphate aerosol mass and number concentration during the Indian Ocean Experiment from January to March 1999 using a GCM. The unique aspects of this study are as follows:

(i) A comprehensive numerical scheme has been implemented to deal with the gas-phase and aqueous phase reactions in a GCM, where the concentrations of radicals like OH, HO_2 and gases like O_3, H_2O_2, NH_3 and NOx are computed within the zoom version of LMD-GCM.

(ii) In most of the global sulphur models only sulphate mass is estimated as a prognostic variable. The number concentration is inferred assuming a constant size distribution. In contrast, in our implementation the sulphate number concentration is also treated as a prognostic variable. The knowledge of the sulphate aerosol number concentration is important to understand the indirect radiative forcing of the aerosols and to refine estimates of the direct radiative forcing.

The model design therefore differs from the earlier studies of Lawrence et al. (1999), Rasch et al. (2000), Boucher et al. (2002), Hauglustaine et al. (2004), Vignati et al. (2004) and Stier et al. (2005) which employ analyzed meteorology / observations or prescribed three-dimensional short lived radical fields to drive the CTM.

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2) Why is the reaction of DMS with NO3 and other species left out when Boucher et al. (2003, ACP) showed it could be 1/4-1/2 of the total DMS oxidation? I would suggest that the runs really should be redone including this reaction to make them quantitatively valid.

Authors' Response: We agree that this is a limitation of the model version described in the manuscript as only the daytime oxidation of DMS has been considered; but it has been done with a view to incorporate chemistry with a proper evaluation of sulphur budget. Thus, the model has been verified with the successful simulation of sulphur cycle to check the source-sink relationship; omission of this pathway does lead to increase in the lifetime and burden of DMS, but this should be kept in mind that all the DMS emission are oxidized to SO_2 (Langer and Rodhe, 1991). The lifetime and burden of DMS are therefore affected by the missing reaction with the NO_3 radical but the budgets of DMS and SO_2 are hardly affected as DMS has very low dry and wet deposition rates. Boucher et al. (2003) also pointed out a significant source of uncertainty in the atmospheric DMS cycle with constraints due to lack of measurements for NO3 radical. Therefore, for model to be satisfactorily extended to have the missing NO_3 pathway, a single run would not be sufficient to have quantitative assessment. Notably, the oxidation path of DMS with NO_3 requires the application of an explicit NOx parameterisation as well as explicit treatment of all the associated chemical species, which are formed /oxidized in the process. It requires a comprehensive inclusion of not only NO_3 but also N_2O_5 and NO_2 in the interactive gas-phase chemical reaction system (Lawrence et al., 1999). This manuscript is however, only the first of a series of papers on the interactive model described here. For refining the model response to DMS, inclusion of NO_3 pathway is certainly very important to be included in the next series of experiments. The above description has been added to the section 2.2.1.

Although this is admittedly a weakness of the analysis, in this manuscript, the following sentence has been added to section 4 clarifying this as one of the model uncertainty arising from such a limitation. "It is finally remarked that the results reported here do

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not take into account the DMS oxidation with NO_3. Further studies are needed to include this pathway and to see the resulting changes."

3) Assuming Henry's Law equilibrium for SO2, H2O2 and O3 can be rather inaccurate when the aqueous phase components are quickly reacting away with each other; can the authors justify the use of this approximation here?

Authors' Response: We do not take explicitly into account mass transfer rates between the gas and the aqueous phase in view of an examination on mass transfer effect by Schwartz (1986) and Venkataraman et al. (2001) who have shown that the mass transfer effect does not limit the rate of sulphate formation.

4) Why is a prescribed liquid water content used (section 2.3)? Doesn't the LMDZ microphysics provide more accurate and appropriate values?

Authors' Response: So far, in-cloud scavenging in LMDZ has been described in a static way. We require more dynamic description and generous approach to clarify the assumptions. Unfortunately in the LMDZ liquid water content is not completely realistic due to the lack of a sub-grid scale distribution of the cloud properties in the vertical.

5) Not distinguishing between liquid and ice scavenging (i.e., treating ice as if it were simply liquid) is likely to be an important deficiency; a sensitivity run would help to elucidate this (should be relatively easy given the simple representation of scavenging).

Authors' Response: We do not distinguish between liquid and ice scavenging. Ice-phase processes might also be of importance in precipitation systems. In an idealized simulation using a global chemical transport model, Crutzen and Lawrence (2000) investigated the sensitivity of trace gas removal by the ice particles and found that scavenging by the ice phase is generally much less efficient than the aqueous

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phase while inclusion of ice uptake essentially extended the vertical region of gas scavenging. However the sulphur scavenging by ice is not very well understood in the global models. Therefore as a first approach we have treated all parts of cloud as liquid. We have added the reference of Crutzen and Lawrence (2000) and the above description of in-cloud scavenging in section 2.3.

6) The way in which the release of gases from evaporating precipitation is treated will err strongly towards too much release of highly soluble gases (like HNO3), which should remain in the droplets until nearly the entire population evaporates; this should be commented on, and again a sensitivity run to elucidate whether this effect is as important as one might expect would be valuable. Likewise, the choice of 0.5 retention for aerosols in evaporating droplets is extremely arbitrary; does it have any physical basis, and how sensitive are the runs to the uncertainty in this parameter?

Authors' Response: We agree here that the choice of 0.5 retention for aerosols in evaporating droplets is arbitrary. In general, the contribution from below cloud scavenging to the concentration of dissolved gas is of less importance (Asman et al., 1995); following that the reevaporation of sulphate is a small term in the budget, it does not affect much the distribution. Wet deposition of highly soluble trace gases is an uncertain factor in global scale modelling. As more dynamic description and generous approach to clarify the assumptions in the model are underway, we have chosen to leave the sensitivity test for this choice until more is known about the process. The above explanation has been incorporated in the revised manuscript in section 2.3.

7) The split operator treatment of doing scavenging before deep convective transport is certainly better than the opposite order of operations, but it still may not be sufficient, especially if the scavenging scheme is (properly) limited to only scavenging the fraction of the grid cell where precipitation is falling, and the deep convective mass fluxes are (as typical) applied to the entire grid cell. It is, however, quite an effort to do better; the

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authors should simply make this clear and expect it to possibly lead to discrepancies in their results.

Authors' Response: As said, this is quite an effort to do better. In the present manuscript we have added it as suggested by the reviewer in section 2.4. It should be noted that we also take into account an additional scavenging term (section 2.4) related to the convection (where there is detrainment of aerosols from the updraft). This additional term accounts for the processing of air from the environment in convective updrafts (Crutzen and Lawrence 2000; Reddy et al., 2004).

8) The second really major problem I see with the manuscript is that the evaluation is taken much too lightly. Saying that modelled levels agree with observations within a factor of 1-2 does not say much, and in this case is not even correct (in March the discrepancy goes up to 2.5-3x). The agreement for aerosol quantities shown here is much poorer than that shown for gases like O3 and CO by de Laat, Lawrence, and others. What can we learn from this? The term "reasonable" agreement is used a lot, which again is not very informative (admittedly, the term is used very often by modellers, and I've also been guilty of this in the past, so this paper should not be singled out, but it is important to try to take a step beyond this). The evaluation needs to be made more quantitative, and the authors should not see a discrepancy as a bad feature of their model, but rather as an opportunity to learn some about what is missing in our current theoretical understanding (to the extent that it is represented in the model). Where specific sources of uncertainty are suspected, this needs to be discussed, rather than simply citing previous studies (like Wilson et al. and Ackermann et al.).

Authors' Response: We have corrected this sentence in the manuscript that modelled levels agree with observations within a factor of 2-3. We are grateful to reviewer for pointing out this mistake. Further, to see the agreement of model simulated trace gases concentration to observations, we plot the CO surface concentration (Fig. 5).

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The simulated CO concentration remarkably agrees with observations between day-62 and day-82 while the model underestimates the observations by a factor of 2 between day-42 and day-62. Next, we plot simulated latitudinal ozone mixing ratios from 0 - 3 km altitude range. It is evident from the Fig. 5 (lower panel) that model is able to simulate the general north-south gradient in the ozone concentration in lower troposphere (de Gouw et al., 2001). The model simulated ozone concentrations of 8 to 31 ppbv agree well with the observed ozone profile (5 to 25 ppbv) obtained after averaging in the region lying between 8°S to 8°N. The model has thus reproduced faithfully the trace gas observations. This description has been added in the manuscript at end of section 3.2.2.

We have omitted the usage of word "reasonable agreement", wherever applicable.

There are not much compilations available on the size-distribution measurements suitable for model for INDOEX-IFP, therefore for the sake of evaluation of number concentration with observation we look at the time average over the month for each ship measurements for 1998 as well. The caption of Figure 6 is therefore rewritten in order to point out that simulation has been done only for the period of January to March 1999. As the simulations are not done in the nudged mode, for the sake of number concentration evaluation, we show the comparison with FFP-1998 observations as well. We have removed the sentence, which cite the references of Wilson et al. and Ackermann et al. Also we have extended the discussion on latitudinal variation of number concentration (end of section 3.3) in the light of observations from Kamra et al. (2003).

9) My final major point is that the analysis needs to dig in deeper; most of the points are reconfirmations of earlier findings, such as the north-south gradient, and the contrast between Aitken and accumulation modes. The only major new point I found was the difference between the results using the two inventories, though this was also only treated very roughly - it was not clear what in the underlying development of the inven-

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tories may have been responsible, and also whether one is really better (the RV run did come out better often, but generally only marginally better). Since INDOEX has been chewed on a lot, and since this paper also takes on the task of describing a new model, I don't think the authors need to overdo it in terms of trying to find new points (it is understandable that one can't do everything in each paper), but I do think that the paper does need a bit more depth in the analysis in order to reach the standards of final publication in ACP.

Authors' Response: In this work we emphasize on the role of Indian emissions over the Indian Ocean because previous research shows the presence of dense anthropogenic aerosol layer over INDOEX region (Ramanathan et al., 2001). In order to assess the role of Indian emissions in increased pollution levels, we present the Indian source contribution to sulphate burden in section 3.5. To clarify the Indian inventory formulation we have extended the section 2.6 and added a figure with the following description in section 2.6. "The simulated SO_2 emissions in the RV and GEIA inventories averaged for the INDOEX-IFP period are shown in Fig. 2. The identification of large point sources in the RV inventory is an advantage over the global inventories, where the emissions are estimated at country level. The RV emission inventory is constructed with very high source resolution. The sulphur emissions are estimated using plant specific fuel combustion."

To extend this work we now also show the differences obtained in the radiative forcing and AOD by the two inventories (Fig. 15). Our result indicates that Indian emission does contribute significantly to the AOD and radiative forcing over the Arabian Sea while there is meagre to very small contribution over the Bay of Bengal. The work has been extended and included in section 3.4.3.

Minor points:

1) There is some redundancy in the abstract (regarding the model reproducing the

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observations)

The abstract has been modified and rewritten again.

2) Andronache's work with a cloud-resolving model (in the JGR INDOEX special issues) should be cited along with the other earlier studies (introduction).

Done

3) The set up of the model runs is not clear; in 3.1 it says it was run over the IFP, spinning up from 20 December 1998, but then later it appears that a run was also done for the FFB (Jan-Mar 1998), while on the other hand in section 3.2.3 it is stated that "the model meteorology is different from the actual one" (this could be interpreted as meaning for a different period, or simply meaning that there are errors in the ECMWF analysis, in which case it should be stated more precisely).

The simulations are done with the initial state prepared from the ECMWF analysis for 20 Dec. 1998 and model is integrated for 100 days to represent the results from January to March 1999 (IFP-1999). However, since the simulations are done without nudging, for the sake of evaluation of number concentration with observations we look at the time average over the month for each ship measurements for FFP-1998 as well. A clarifying comment has been added to caption of Figure 6 in revised manuscript.

4) section 3.3.1: "captured by the model" - since this is not compared to observations, it is difficult to see what feature the model is "capturing"

Appropriate change in the text has been done in this section.

5) section 3.3.2: "highest AOD is observed" - should read "is simulated"

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Done

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