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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

Interactive comment on "Influences of in-cloud aerosol scavenging parameterizations on aerosol concentrations and wet deposition in ECHAM5-HAM" *by* B. Croft et al.

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Author Response to Referee Comment #2

The authors wish to thank this referee for the insightful comments and suggestions that have helped to improve this manuscript.

Referee comments are with labeled RC, and author comments are labeled with BC.

Major points:

1) RC: For the entire paper and for specific sections explanations of why the investigation is being conducted needs to be included.

BC: We agree with the referee that the manuscript needs to have a clearer description of why the investigation is being conducted. The abstract, introduction and conclusion of the manuscript have all been re-written with an emphasis on explaining why the investigation is being conducted. The investigation was conducted for several reasons:

a) The development of a new diagnostic in-cloud scavenging scheme allows us to examine the relative importance of the nucleation versus impaction scavenging processes for warm, mixed and ice phase clouds from a global and annual mean perspective.

b) Since a variety of in-cloud scavenging parameterizations have now been implemented in the ECHAM5-HAM GCM, we are able to assess whether uncertainties in the representation of in-cloud nucleation and impaction scavenging can lead to significant differences in predicted aerosol concentrations, burdens, and deposition.

c) The comparison between in-cloud scavenging parameterizations also allows us to examine the strengths and weaknesses of the various parameterizations, and based on this to provide recommendations in regard to the modeling of in-cloud scavenging in global models.

We have included these motivations in the revised introduction, particularly paragraphs 2 and 3. We continue to address these motivations throughout the text.

2) RC: Explanation of what the results mean would significantly improve the paper.

BC: We also agree with the referee that a clearer explanation of what the results mean and a clearer statement of our conclusions and recommendations would improve the manuscript. As a result, the abstract and conclusions have been re-written to provide a clearer explanation of the meaning of the results. Specifically we have added the following conclusions and recommendations.

a) We found that in the global and annual mean, the aerosol mass scavenged in stratiform clouds is primarily attributed to nucleation processes (>90%), whereas the aerosol

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number scavenged is primarily attributed to impaction processes (90%, with 99% of this total occurring in clouds with temperatures below 273 K). This leads us to the recommendation that that impaction scavenging in mixed and ice phase clouds must be given careful attention in the latest generation of microphysical aerosol models that are becoming available. Additionally, we acknowledge that there are uncertainties associated with the impaction process, particularly for mixed and ice phase clouds, and thus we recommend further research efforts to improve understanding of impaction processes.

b) We found that aerosol concentrations in the middle and upper troposphere were most sensitive to differences in the parameterization of in-cloud scavenging. For warm phase clouds in the lower troposphere, in-cloud scavenging fractions are often close to unity for the aerosol modes that contain the majority of the aerosol mass, and thus there is a lower sensitivity to the in-cloud scavenging parameterization. Different in-cloud scavenging parameterizations were found to change black carbon concentrations in the middle troposphere by up to an order of magnitude. Thus, we recommend modelers to give particular attention to scavenging in mixed and ice phase clouds.

c) In comparing the strengths and weaknesses of the various parameterizations, we found that while scavenging by prescribed fractions gives reasonable results, and is computationally less expensive, the more physically detailed parameterizations generally performed better in comparison with observations, particularly evident in the middle troposphere. Additionally, prescribed fractions do not represent the considerable variability of scavenged fractions that occur within the size and temperature ranges implemented by the prescribed fraction method of our model. We now recommend the diagnostic and prognostic approaches as preferable to prescribed fractions.

Specific Points:

1) RC: The abstract and conclusion are key places where the reasons why the study is being conducted and what the results mean should be placed. The introduction of course needs to include the reasons for the study.

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BC: The abstract, introduction and conclusion are now re-written to specifically focus on the reasons why the study was conducted, and what the results mean.

2) RC: One paper that was not cited or discussed was the one by Jacobson (2003) JGR who presented a 1-dimensional parameterization of chemical and aerosol cloud processes. How does his work compare with this current study.

BC: Thank you for pointing out this omission. We have now included this reference in the paragraph 4 of the introduction. Our work agrees with the findings of Jacobson (2003) in that we found for stratiform clouds that the aerosol mass scavenged is primarily (>90%) due to nucleation processes, and the aerosol number scavenged is primarily attributed to impaction processes (>90%). We have introduced a new table (Table 11) that shows the annual and global mean aerosol number removal by the processes of nucleation versus impaction scavenging for warm, mixed and ice phase clouds, and comparing the simulations CTL, DIAG1, DIAG2, and DIAG-FULL. We found that scavenging in mixed and ice phase clouds accounted for 99% of the aerosol number scavenging in stratiform clouds, and that changes to the assumptions about impaction scavenging can change the global and annual mean number scavenging rates by up to 7 -fold, with a particular sensitivity for ice clouds. These results are discussed in the new Section 3.5.2.

3) RC: In the results section, there is a lot of detail about how one simulation compared to another. I think it would be better to reduce the detail and point out what is important to learn from the figure or table.

BC: We have re-written the text of the results section to start the discussion of each figure or table with what is the most important point to learn.

4) RC: One thing that I am not sure that I learned is whether it is worthwhile calculating separate aerosol mass and number scavenging coefficients or not. . . . Could the authors provide a more definite recommendation?

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BC: We have revised the conclusion to recommend that it is worthwhile to calculate separate mass and number nucleation scavenging coefficients. We also discuss this more clearly in the comparison of simulations DIAG1 and DIAG2 in Section 3.2 and 3.3 related to nucleation scavenging. For impaction scavenging, we used separate scavenging coefficients for the case of scavenging by cloud droplets, but not for scavenging by ice crystals. We now have added a note to the second to last paragraph of Section 2.1.3 to indicate that this is an additional uncertainty related to impaction scavenging by ice crystals that we did not address.

5) RC: I found the results of the different scavenging parameterizations on sulfate and black carbon burdens (page 22069, line 23-24) to be significant with 22

BC: This comment appears to be truncated. Did you mean page 22059, lines 23-24? This comparison illustrates the importance of impaction scavenging parameterization for these species, particularly in the case of the prognostic scavenging. We have added a sentence here to further discuss this finding.

6) Figure 4 shows aerosol mass concentration (zonal and annual mean) while Figure 7 shows aerosol mass mixing ratio (zonal and annual mean). Why is it important to show both of these very similar quantities? An explanation could included near the top of Section 3.3.

BC: We show the aerosol mass concentration that is scavenged into the droplets and crystals (zonal and annual mean) in Figure 4, and the aerosol mass mixing ratio (zonal and annual mean) in Figure 7. We decided to show both of these quantities since it is not intuitive whether greater scavenged mass would be associated with greater or lesser mass mixing ratios. We add a discussion related to this in the first paragraph of Section 3.3.

7) RC: In discussing results for aerosol number distributions, the authors point out that models that use a simple scavenging parameterization tend to overestimate the production of new particles. From this finding are there important parameters from the

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previous studies that could be impacted (radiation, chemistry, aerosol burden)?

BC: Yes, simulations F100 and F100-INT show that using large prescribed scavenging fractions (set to unity) leads to excessive numbers of new particles since the surface area available for condensation on to larger particles is reduced, and thus more new particles form. We agree that further studies to investigate the impacts of this overestimation of new particles on the aerosol direct and indirect effects on the radiation budget, chemistry and aerosol concentrations would be worthwhile. Although, the nucleation mode particles do not greatly influence the radiation budget directly, there may be feedbacks that are important. Further work to examine how much the new particle production needs to change before there is a significant impact on the radiation and chemistry would be useful. From an air quality perspective, the predicted number of fine mode aerosols is also of importance to human health. We add a few sentences in Section 3.4 to comment on these points.

8) RC: Why is BC poorly predicted (p 22066)?

BC: Differences in the parameterization of in-cloud scavenging do contribute significantly the differences in the prediction of black carbon concentrations, particularly in the middle and upper troposphere as shown by Figures 15 and 16. Black carbon is an aerosol that can be internally or externally mixed depending on how the aging and coating processes are represented in any given model, and this also contributes to differences in scavenging rates and predicted concentrations. Additionally, black carbon mass removal in mixed phase clouds, and number removal in mixed and ice phase clouds depends strongly on the representation of impaction scavenging, which can vary significantly between models. Thus, from a scavenging perspective black carbon is difficult to represent since it can have a significant mass in the externally mixed Aitken size mode, which is sensitive to the representation of impaction scavenging. Also, depending on the parameterization of the black carbon ageing varying amounts of the mass might be in the internally mixed accumulation size range, which would be scavenged more readily by nucleation processes. These are some of the factors

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related to scavenging that make prediction of black carbon more difficult. We add a comment related to this in the Section 4 discussion of Figs. 15 and 16.

9) RC: I recommend strengthening the conclusions with recommendations instead of saying 'some consideration should be given' or 'future work could be directed'

BC: The conclusions have been re-written to present the recommendations more clearly.

Technical Details

1) RC: P. 22043, line 1 It would be more clear to say 'cloud nucleation scavenging scheme' so as not to confuse with particle or aerosol nucleation.

BC: This change is made.

2) RC: P. 22043 line 23-25 The last sentence of the abstract is missing a verb

BC: Verb is now added.

3) RC: P. 22049, line 6, -> must be apportioned

BC: Now corrected

4) RC: P. 22052, line 14, what size is assumed for the monodisperse ice crystals

BC: The monodisperse ice crystal size is calculated following Equations 1-3 of Lohmann et al. (2008). The size calculation depends on the ice crystal number concentration and the ice water content. We now make reference to this calculation in the text.

5) RC: P. 22054, line21, reference to the table should be at the beginning of the paragraph

BC: Reference to table is now at start of paragraph.

6) RC: P.22060, line 19, -> interstitial

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BC: Spelling corrected.

7) RC: P.22061, line 7, ->associated

BC: Spelling corrected

8) RC: P.22064, line 4-5, I do not see that DIAG-FULL and PROG-AP are different than how the CTL simulation behaves in Figures 10 and 11. Could this sentence be clarified?

BC: The discussion has been changed such that we no longer claim that there are any significant changes for this comparison.

9) RC: P.22068, line 1, It would be more clear to say ‘cloud nucleation scavenging scheme’ so as not to confuse particle or aerosol nucleation.

BC: Suggestion has been implemented.

10) RC: P. 22069, line 17-18, I suggest combining sentences to more easily explain why separate mass and number scavenging coefficients should be used.

BC: These sentences have been re-worded to make the explanation more clearly.

11) RC: Multi-panel plots with colorbar for each panel are hard to read. Try enlarging panels and using only one colorbar for each column or figure.

BC: Figures 4, 6-9 have been corrected following this suggestion.

12) RC: Figure 10 and 11, please state what the acronyms mean (NADP, EMEP etc.)

BC: Explanation of acronyms is now included.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 22041, 2009.