

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 # 1

The authors thank this referee for the helpful comments, questions, and suggestions, which have led to improvements in the quality of this manuscript.

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

General comments:

RC: As there are many uncertainties in predicting global aerosol distributions, this paper presents a method to improve the accuracy of model results from improvements

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in in-cloud removal schemes. This finding is very useful in helping other models in improving the wet removal process.

BC: In order to present our conclusions and recommendations more clearly in the revised manuscript, we have re-written both the abstract and conclusion sections. We now point out that impaction scavenging was found to account for more than 90% of the aerosol number scavenged in stratiform clouds, and 99% of that total was attributed to scavenging in mixed and ice phase clouds for our diagnostic scavenging scheme. As well, 50% of the dust mass scavenged in stratiform clouds was attributed to impaction scavenging. Thus, global modelers should give careful attention to the representation of the impaction scavenging process for the mixed and ice phase clouds of the middle and upper troposphere, and for dust at all cloud temperatures.

Specific comments:

1) RC: A gamma distribution for cloud droplets is chosen in this study. It should be noted that using different droplet spectra can cause scavenging coefficients to differ by a factor of 3-5. Using different collection kernels can cause scavenging coefficients to differ by an order of magnitude. A brief discussion regarding potential uncertainties related to these different choices of inputs is needed.

BC: At the end of the second last paragraph of Section 2.1.3 we have added a description of these uncertainties.

2) RC: Figure 1 shows size-dependent scavenging coefficient, but this is not really for a specific size, but rather for a whole aerosol spectrum that has this geometric mean. There is nothing wrong with the figure, but the related discussion in the text implies that the scavenging coefficient is for a specific aerosol size. Some clarification is needed.

BC: In the first paragraph of Section 2.1.3 after the introduction of Fig. 1, we have added the sentence to clarify that the radii are the geometric mean radii for the assumed lognormal aerosol distribution.

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3) RC: The comparison between DIAG-FULL and CTL runs with observations show some degree of improvement by DIAG-FULL. Is this improvement statistically significant?

BC: For the scatterplots shown by Figs. 10, 11, and 16, the correlation coefficients do not change enough between the simulations DIAG-FULL, PROG-AP, and CTL that we could argue that there is any statistically significant improvement. We have revised the discussion related to sulfate wet deposition in the first paragraph of Section 4, and also the second paragraph of Section 4.1 related to ^{210}Pb and ^7Be to state that there is not any statistically significant difference between the simulations for our comparisons with observed wet deposition or surface layer concentrations. This result is not unexpected since the majority of the aerosol mass, and thus the aerosol mass deposition, is associated with warm phase scavenging in the near surface layers. For warm phase clouds and the soluble/internally mixed accumulation and coarse mode aerosols (containing the majority of the aerosol mass), the in-cloud scavenging fractions are near to unity, and do not change significantly between the various simulations. Thus, we would expect that all of our simulations would compare similarly with the observations of sulfate, ^{210}Pb and ^7Be wet deposition and surface layer concentrations. We found that the various scavenging approaches differ more for the mixed and ice phase clouds that usually do not coincide with the locations of the greatest aerosol mass concentrations. Black carbon concentrations in the middle troposphere increased by up to one order of magnitude for DIAG-FULL as compared to the CTL simulation, which improved agreement particularly with high latitude observations since those concentrations were underestimated by near to 2 orders of magnitude for the CTL simulation. This is discussed in paragraph 5 of Section 4.

4) RC: Is the size distribution simulated from the model comparable with observations? How does the impact of size distribution on in-cloud removal compare with the impact between DIAG-FULL and CTL?

BC: The size distribution from the ECHAM5-HAM compares well with observations as

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shown by Stier et al. (2005), ACP (See Fig.6) and Hoose et al. (2008), ACP (See Fig. 11). We now make a reference to these comparisons at the end of paragraph 4 of Section 4, and also add a sentence to note that the correct simulation of the size distribution is also essential to representing the in-cloud removal processes properly since removal is a size-dependent process. Thus, the scavenging influences the size distribution, but the size distribution does also influence the scavenging. A completely erroneous size distribution would cause greater errors in the in-cloud removal than the current uncertainties that we have found associated with the in-cloud scavenging parameterizations, but this investigation is beyond the scope of this paper.

5) RC: Figure 2. . . There is a sharp decrease in the collection efficiency in the 1-2 μm range. Is this physical and what kind of impact would this have on the aerosol size distribution after in-cloud scavenging.

BC: This is called the 'zero scavenging zone (ZSZ). The physical explanation is presented in a paper by Miller and Wang (1991). This minimum occurs when the combination of forces at work on the aerosol particle and collector cause a near zero probability of collision. We now mention the ZSZ in Section 2.1.3 in the text related to Figure 2. In reply to your second question, aerosol particles in the 1-2 μm size range in clouds will be readily scavenged by nucleation scavenging processes, and so this sharp decrease in the in-cloud impactation scavenging should not be a significant limitation to the removal of particles in that size range.

6) RC: . . . the DIAG scheme gave better agreement for black carbon profiles. For other aerosols the budgets are changed by the DIAG scheme but no information was given if the scheme made better agreement.

BC: Since aerosol mass concentrations in the middle and upper troposphere show the greatest sensitivity to the various in-cloud scavenging parameterizations, we have now added further comparisons with observed vertical profiles of 210Pb, 7Be, and sulfate. Figures 17 and 18 show these comparisons, and there is an associated discussion

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added to the end of Section 4.1. Similar to our findings for black carbon, the greatest changes are found in the middle troposphere. ^{210}Pb concentrations increase by up to 30%, which improves the agreement with certain mid-troposphere observations at mid- and high latitudes.

7) RC: As there are many variations of the DIAG scheme tested in the model, it would be nice to see a summary of the applicability of these tests to offer some guidelines for global models in implementing in-cloud schemes.

BC: We have now re-written both the abstract and conclusion to more clearly present our recommended guidelines. In the new Section 3.5.2, we point out that aerosol number scavenged in stratiform clouds is primarily attributed to impaction scavenging in mixed and ice phase clouds and we point to the importance of careful attention to this process in the global models. Comparison between DIAG-FULL and DIAG2 shows the sensitivity to the impaction parameterization and this is discussed in the Section 3.5.2. The scavenging of aerosol number and mass with separate nucleation scavenging ratios is also recommended, and is discussed in the first paragraph of Section 3.2 and paragraph 4 of Section 3.3 with respect to comparisons between DIAG1 and DIAG2. In the discussion related to Figs. 15 and 16, we also conclude that the greatest uncertainty related to the in-cloud scavenging schemes was for mixed and ice phase clouds. These recommendations and conclusions are now incorporated more clearly in the abstract and conclusion.

Technical corrections:

1) RC: Page 22043 line 24: change 'are' to 'is'

BC: This correction has been made.

2) RC: Figure 4: the labels on the plots should be enlarged to be seen properly

BC: We have re-plotted Figs. 4, and 6-9, and removed the excessive number of color-bars to make the figures easier to read.

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3) RC: Page 22068 line 6 insert 'were' before increased

BC: This line is removed in the new text, and so this correction is not applicable.

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