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Atmospheric Chemistry and Physics Discussions

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 # 3

The authors wish to thank Dorothy Koch for her constructive, and thorough review, and suggestions that have led to improvements in this manuscript.

Comments by referee Dorothy Koch are labeled with DK, and the author response is labeled with BC.

Major Comment:

DK: Therefore the major suggestion I have for revision is to encourage the investigators

to pursue comparison with more aircraft data, widely available for sulfate, 210Pb and 7Be. I expect that the schemes may eventually have a large effect on indirect effects for high-level clouds. So proper simulation of aerosols in that region will be important. Therefore further testing of the schemes against aircraft data would be important. Note that, in Koch et al., 2006, there is discussion of the challenge of simulating 7Be and 210Pb changes with altitude. In particular 210Pb is overestimated in the free troposphere. Liu et al. (2001) had a similar problem and corrected it by adding a cirrus-cloud settling parameterization. It would be quite interesting to see which of your schemes are most successful in comparison with sulfate, 7Be and 210Pb.

BC: We have added 2 new figures (Figs. 17 and 18) that show our simulated aerosol vertical profiles compared with aircraft data. One of these figures repeats some of the comparisons of Liu et al. (2001), and the other figure includes data presented in Kownacka (2002), and Heikkilä (2007). Modeled 210Pb concentrations were underpredicted near 5km, and the new diagnostic scheme does increase the concentrations by 20-30% for several of the mid- and high latitude profiles (CPML, WPML, Poland regions), which improves the agreement with observations. The profiles for the low latitude regions are particularly influenced by convective clouds, in addition to stratiform clouds. Additional work to revise the convective in-cloud scavenging parameterizations may further improve the agreement with observations for these regions. When comparing our model results with these observations, we must keep in mind that many of these observations are taken during short-lasting flights and reflect momentary weather conditions, and our modeled results are monthly means. For these reasons highly accurate comparison with observations is challenging, but as described in the text we have tried to match the model with the observations carefully in terms of region and month.

We also compare with a sulfate vertical profile from the TRACEP campaign of 2001, which shows sulfate concentrations are increased by up to 40-50% near 5km for the PROG-AP compared to the CTL simulation. Again, the greatest sensitivity of the modeled concentrations to the scavenging parameterization was for the middle/upper troposphere, but the sensitivity was not as great as for black carbon concentrations. The model overpredicts sulfate concentration near 5 km, but SO2 was also overpredicted. Thus, we cannot conclude that this overprediction indicates a problem with the aerosol scavenging schemes. We have added a discussion of these 2 new figures to the text at the end of Section 4.

Minor comments:

DK: The introduction is confusing, with what feels like a random discussion of various scavenging mechanisms, making it difficult for the reader to know what the study will address. Some introductory sentences clarifying and summarizing the various aspects of scavenging addressed would be helpful. For example, there is discussion of how this study focuses on diagnostic model development but then moves on discuss a prognostic approach. An introductory explanation of that both of these will be considered should follow the paragraph on p 22044 line 23. Similarly on p 22045, the 2nd paragraph should begin with a statement that this study will address not only aerosol uptake but also in-cloud impaction.

BC: A considerable portion of the introduction has been re-written to provide a clearer description of the reasons why the study was conducted, and what the study will address. Following your particular suggestions, the second paragraph of the introduction has been revised to add the introductory explanation that both the diagnostic and prognostic approaches will be considered in the study. The first few sentences of paragraphs 2 and 4 of the introduction have been revised to clearly state that the study will also examine in-cloud impaction scavenging.

DK: P22047 What are the sources for7Be and 210Pb/radon? How are they distributed onto the aerosol size distribution?

BC: The production rates for 7Be were taken from Masarik and Beer (1999). We have added this reference to Section 4.1. This is also described in greater detail in Heikkilä (2007) and Heikkilä et al. (2008). The source for 210Pb/radon is from soils and was

taken to be 1 atoms/cm2/s, following Feichter et al. (1991), Liu et al. (2001) and Koch et al. (2006). 7Be and 210Pb are not explicitly distributed into the aerosol distribution. The scavenging efficiency for 7Be is found by taking the ratio of the total sulfate mass scavenged to the total aerosol mass, whereas for 210Pb the scavenging efficiency was determined by the ratio of the total aerosol mass scavenged to the total aerosol mass. The rationale is that near the surface where 210Pb is formed, particles are composed of many chemical compounds whereas for the 7Be source in the stratosphere, and upper troposphere, sulfate is the dominant chemical compound. This was the approach presented by Heikkilä (2007). These details are also added to the text.

DK: P22051 line 2-3 “The used version of the ECHAM5-HAM” change to “The version of the ECHAM5-HAM used here”. There was an earlier sentence like this as well.

BC: This wording has been changed in the first and last paragraph of Section 2.1.2.

DK: P22051 line 2, define “homogeneous freezing” and explain why this means the treatment must be different. Is homogenous freezing the only IN scheme used here?

BC: Homogeneous freezing occurs only at temperatures below 238K, and does not require an ice nucleus. Heterogeneous freezing does require an ice nucleus to be present, and occurs for temperatures between 238K and 273K in our model. Both homogeneous and heterogeneous freezing (contact and immersion freezing) are included in our model. The rationale for the different treatment of the scavenging for temperatures below 238K is as follows. For temperatures warmer than 238K, the cloud droplets, and those frozen to crystals in our model all originate from the Ghan et al. (1993) activation scheme, which includes all soluble/internally mixed aerosols larger than 35 nm in the droplet activation. Thus the aerosol nucleation scavenging by droplets and crystals, and the apportioning of the number to be scavenged between the aerosol modes can be treated similarly at these temperatures. However, for temperatures colder than 238K, the crystals do not originate from the Ghan et al. (1993) activation scheme. We decided to scavenge the soluble/internally mixed aerosol modes progressively from the largest to smallest aerosol sizes, which is consistent with the freezing parameterization of Kärcher and Lohmann (2002) used for temperatures below 238 K in our model. We have added this explanation to the second and last paragraphs of Section 2.1.2.

DK: P22052 equation 15, define E.

BC: The definition of E is now given immediately following Equation 15. This is the aerosol-ice crystal collection efficiency.

DK: P22054 I do not understand DIAG2, it uses prescribed impaction kernels in Table 2, but how do these differ for number and mass?

BC: DIAG2 uses the prescribed 'impaction' kernels in Table 2, and these do not differ for number and mass scavenging, as you have correctly noted. DIAG2 does use different 'nucleation' scavenging ratios for number and mass. We have re-worded the third sentence of Section 2.1.5 to make this distinction between changes to nucleation scavenging versus impaction scavenging clearer to readers. Also, in Table 3 we added the word nucleation to the start of the description of DIAG1, since comparing DIAG1 and DIAG2 is meant to focus on differences to the nucleation scavenging only. In the fourth paragraph of the conclusion we also revised the text to include the word nucleation in the statement related to the equating of mass and number scavenging ratios. In this study, we did not examine the impact of equating the mass and number impaction scavenging coefficients, but there may well be an additional sensitivity associated with this, particularly since we did equate mass and number coefficients for aerosol-ice crystal impaction, but not for aerosol-droplet impaction. We added a comment to acknowledge this in Section 2.1.3.

DK: There are simulations listed in Table 3 that are not described in section 2.1.5.

BC: The simulations DIAG-FULL-noimp and PROG-AP-noimp are now described in Section 2.1.5.

DK: P22056 top paragraph. The explanation for why the DIAG-FULL has more aerosol

in-cloud and in-crystal compared to CTL is not clear.

BC: The text has been re-written to make the main point of this discussion more clearly. We now state that 'These more physically based parameterizations (DIAG-FULL and PROG-AP) indicate that the mass scavenged, particularly in ice clouds, is greater by up to two-fold as compared to the mass scavenged using the prescribed scavenging fractions of the CTL simulation.'

DK: P22056 This should appear earlier, in the model description: "Our study also implemented the below-cloud scavenging parameterization of Croft et al. (2009),. . ." along with a summary of what this is. And what was used for below-cloud in Hoose et al. 2008a that makes the new scheme more efficient? And further in this sentence, the word "interstitial" should not apply to below-cloud scavenging. Although Figure 5 is similar to a figure in Hoose et al., it would be worth discussing it further here.

BC: The reference to the below-cloud scavenging parameterization of Croft et al. (2009) is included at the end of the first paragraph of Section 2. We have added a short summary to indicate that 'This physically detailed below-cloud impaction scavenging parameterization uses look-up tables to select scavenging coefficients that represent the collection of aerosols by rain and snow below clouds based on aerosol size and precipitation rates.'

Additionally, we have extended the discussion about Figure 5 to state that 'Hoose et al (2008a) implemented the prescribed below-cloud scavenging coefficients that are included in the standard ECHAM5-HAM model. Croft et al. (2009) show that the below-cloud scavenging with these prescribed coefficients is less vigorous than for the new physically detailed aerosol size-dependent parameterization of Croft et al. (2009). Sensitivity tests included in Hoose et al. (2008a) also show this same comparison, and find better agreement with observations for the detailed aerosol size-dependent parameterization of Croft et al. (2009).'

We have removed the term 'interstitial'

DK: P 22059 Lines 18-25 why does noimp have a bigger effect on PROG-AP?

BC: When the in-droplet and in-crystal aerosol concentrations are treated prognostically, impaction has a greater effect on the predicted burdens since impaction continues to add aerosols to the existing in-droplet and in-crystal concentrations over successive time-steps, unlike for the DIAG-FULL simulation. We have added this to the text in Section 3.3 as part of the discussion of Table 4. We can also see the important influence of impaction on the PROG-AP vertical profiles shown in Figures 14 and 15.

DK: Figs 10 and 11, I suggest plotting these up to 18 instead of 20-something in order to see the points better, especially for Fig. 11 where the data/model values are so small.

BC: The figure has been revised as suggested as you have suggested.

DK: P22065 line 13 the underprediction of PROG-AP over Asia is present in all cases so should not be attributed to the scavenging scheme.

BC: The text has been revised to exclude this reference to Asia. We do not want to mislead the reader to think that is underprediction over Asia should be attributed to the scavenging scheme.

DK: Fig 14 provide more information on the locations of the profiles.

BC: Further detail about the location of the profiles is now included in the text and also in the caption for these figures.

DK: P22066 I think this: “For the simulation PROG-AP as compared to PROGAP-noimp, black carbon concentrations are lower by up to a factor of five, and two in the middle and upper troposphere, respectively.” Should be reworded: “For the simulation PROGAPnoimp as compared to PROG-AP, black carbon concentrations are lower by up to a factor of five, and two in the middle and upper troposphere, respectively.”

BC: We have reworded this sentence to read ‘For the simulation PROG-AP, black car-

bon concentrations are lower by up to a factor of five and two in the middle and upper troposphere, respectively, as compared to simulation PROG-AP-noimp.’

DK: P22066 Since PROGAP-noimp agrees better with observations does this suggest a problem with the impaction scheme?

BC: This does not necessarily suggest a problem with the impaction scheme since there may be other factors that could influence the comparison between the profiles such as uncertainties in local emissions, vertical transport and differences in spatial and temporal averaging. We have to be careful not to over-interpret this comparison with aircraft profiles, particularly since there are some locations shown by Figures 14 and 15 that have poorer agreement for PROG-AP-noimp. We have added a comment about these uncertainties in making our comparisons at the end of Section 4.1.

DK: P22067 Line 18. The age of the radionuclide measurements does not matter. However for 7Be is it important to have the correct point in the solar cycle or to average over an entire cycle. This should be pointed out as a source of model bias (if the source does not match the observation strength in solar source.)

BC: We agree that the production of Be-7 is modulated by the solar (and geomagnetic) activity. However, on short time scales (such as the one year considered in this study), the correlation between the global mean production rate and 7Be deposition fluxes or surface layer concentrations can be difficult to detect in observations (e.g. Graham et al, 2003, Heikkilä et al, 2008). Thus, we consider the variability in the solar cycle to be a very minor bias to our comparisons. Greater uncertainties in the comparison with observations are related to discrepancies in estimates of the source strength itself (e.g. Masarik and Beer (1999), Webber and Higbie (2003)), and particularly discrepancies between the modeled and observed meteorological conditions at the measurement sites, and the grid size of the model which does not resolve local conditions at the measurement sites. In the text we have added a note about these uncertainties near the end of the second paragraph of Section 4.1.

Conclusions

1. DK: Would it be worthwhile to make the FULL scheme prognostic?

BC: Yes, it would be worthwhile to make the FULL scheme prognostic. Particularly, the detailed size-dependent impaction parameterization, and the determination of separate nucleation scavenging fractions for the aerosol mass and number distributions are aspects of the FULL simulation that should be coupled with the prognostic scheme. These recommendations are now stated more clearly in the conclusion.

2. DK: Since there was minimal difference among the sensitivity studies when comparing with most observations, it would really be interesting to see how the direct and indirect effects change for the schemes. Maybe add a paragraph anticipating where the big changes might occur. For example, maybe cirrus cloud

BC: Yes, we agree that further investigations to check how the direct and indirect effects are changed for the schemes would be worthwhile. Differences to the parameterization of in-cloud scavenging has the greatest impact on aerosol concentrations for the middle and upper troposphere, unlike for the warm phase scavenging of the lower troposphere where the in-cloud scavenging ratios, particularly for the soluble/internally mixed coarse and accumulation modes, are relatively similar between the schemes. As you note, this means that the greater changes to the direct and in-direct effects in response to the parameterization of in-cloud scavenging could be related to the middle and upper troposphere, and particularly in response to changes in predicted black carbon concentrations. This discussion is now added to the last paragraph Section 3.4, paragraph 5 of Section 4 related to Figs. 14 and 15, and the last paragraph of the conclusion.

Additional References (not included in the ACPD version of our paper):

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Kownacka, L.: Vertical distributions of beryllium-7 and lead-210 in the tropospheric and lower stratospheric air, Nukleonika, 47, 79 . 82, 2002.

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