Response to referee #1

The authors wish to thank this referee for the thorough and insightful comments and questions.

General comments:

The referee was asking if we could conduct a simulation with aerosol-cloud-precipitation feedbacks removed. The climate of the model deteriorates with the fixed cloud droplet and ice crystal numbers. Thus, we preferred not to conduct such studies since we wanted to make comparisons with observations. To address this concern, we have added a subplot to Fig.6, which shows how the total precipitation changes between the simulation with the aerosol size-dependent below-cloud scavenging and the standard model. In this manner the reader can better interpret the geographic plots and identify localities where changes in the deposition and burdens are more influenced by the revised scavenging, as opposed to changes in the precipitation between simulations. We have also added a discussion to Section 3.1 (second paragraph) and Section 3.4 (second paragraph) about the localities where feedbacks on precipitation did occur, and thus the wet deposition change is influenced by this feedback in addition to the change in the scavenging parameterizations in those regions.

We also adjusted the discussion of the interpretation of the feedbacks between aerosols and clouds for these nudged simulations in Section 3.4 (first paragraph) to emphasize that we must use caution in interpreting these results since nudging can reduce the magnitude of the aerosol-cloud feedbacks. This is further discussed below in reference to the second to the last specific comment.

Specific comments:

Below we respond to the specific comments of this referee.

Q. What exactly is the meaning of below-cloud scavenging?

In regard to terminology, we used the term below-cloud scavenging since we did only calculate the process of aerosol impaction scavenging by rain and snow below cloud base. We now explain this explicitly in the last paragraph of the introduction.

Q. Which emissions are taken from the AEROCOM project?

p.7877, l19-20 The natural emissions of sea salt, dust, and DMS from the oceans are calculated on-line based on the meteorology of the model. Emissions for all other aerosol species are taken from the AEROCOM emission inventory. We have corrected this in the first paragraph of Section 2.

Q. What about the tendency to enhance new particle formation in the lower troposphere, since without condensation surfaces the H2SO4 cannot condense on those
In regard to the scavenging of ultra-fine particles with radius smaller than 10 nm, please note that following the comments of referee 2, we have now removed the assumption of zero collection of these particles from all simulations except for two sensitivity simulations, now labeled BCS2-ULOW and BCS2-UHIGH. Otherwise, we assume collection by Brownian motion following the results of Wang et al. (1978) and as outlined in Tables 1 and 2. We have re-calculated all of the model simulations and re-plotted all of the figures with the revised assumptions. The various assumptions in regard to scavenging of particles less than 10 nm change the global and annual mean mass and number burdens for any aerosol species or mode by less than 5%. In regard to the referee’s question, we do find evidence of enhanced new particle formation in the lower troposphere and have now added an additional figure (Fig. 12) that shows increases in the annual and zonal mean nucleation mode number concentrations by up to 30% in the lower troposphere. The discussion at the end of Section 3.3 (last paragraph) is supplemented in regard to this new figure and the enhanced new particle formation.

Q. I cannot confirm the statement that the two coefficients are similar.

We have corrected the wording such that we do not state that Snow-A and Snow-B are similar. We outline the differences, comment that these arise from assumptions about the snow morphology, and indicate that we have implemented the coefficients from Slinn (1984) in this study.

Q. The convective fraction was chosen with a relatively high vertical velocity, and thereafter, it is relatively small. This also explains the small contribution to the total atmospheric deposition.

We have added a discussion at the end of the last paragraph of Section 3.1 to discuss the limitations of using a relatively high fixed vertical velocity, and the not well developed representation of the convective sub-grid scale effects on the parameterization of convective below-cloud scavenging.

Q. Some of the more regional effects are related to the aerosol-cloud-precipitation feedbacks, which are difficult to analyze.

Below-cloud scavenging and nucleation scavenging cannot occur at the same model level at any given time-step in our model. As clarified above, we only studied impaction scavenging by rain and snow below cloud base, and this requires a completely cloud-free layer. Thus, the order of process calls should not contribute to this enhanced below-cloud scavenging being associated with reduced nucleation scavenging. The physical explanation is now presented in Section 3.1 (third paragraph). This is expected since the greater aerosol removal below cloud base allows less aerosol to be available for transport upward to the altitudes where in-cloud scavenging occurs. Sedimentation and dry mass deposition rates are also reduced in response to the lower aerosol concentrations.
Section 3.2 ‘Column’ has been added to the title.

Q. Is there a physical explanation why sea salt is affected much stronger than the other aerosol types?

p. 7887, l.24-30 Table 8 caption had a typographic error, which has been corrected from ‘sea salt’ to ‘dust’ since the table does refer to dust only. The physical explanation for sea salt being more strongly influenced by the below-cloud scavenging parameterizations has been added to the second paragraph of Section 3.2 in the discussion of the tables.

Sea salt is most strongly influenced by the size-dependent below-cloud scavenging parameterizations since this aerosol species has a considerable fraction of total mass in the coarse mode, and the scavenging coefficients for this mode are greatly enhanced, by one to two orders of magnitude, as compared to the prescribed coefficients of Stier et al. (2005) (shown in Fig. 2). Dust also has a considerable mass in the coarse mode, but the lifetime reduction is less, 7% as opposed to 15%, for sea salt between the BCS2 and CTL simulations. This occurs since dust tends to be emitted in regions of lower precipitation, and is lofted above the altitudes of below-cloud scavenging while being aged to a soluble/mixed state, which can be scavenged by cloud nucleation processes. On the other hand, sea salt emissions are generally in regions of stratiform precipitation, and are more susceptible to removal by below-cloud scavenging shortly after emission.

Q. The prognostic rain scheme has a much stronger effect on impaction scavenging, due to subsequent evaporation of rain below-clouds. However, this is to a large extent compensated by nucleation scavenging. This should be pointed out better.

p. 7888, l.4-20 We have added a sentence to the middle of the last paragraph of Section 3.2 to discuss the reduction of in-cloud scavenging following the increased below-cloud scavenging for simulation BCS2-PR.

Q. According to the cloud scheme presented by Lohmann et al. (2007), the hydrophobic particles act as ice nuclei.

p. 7890, l.11-15 We have corrected the sentence in relation to hydrophobic particles to read: ‘...these effects are greatest for the hydrophobic aerosols, which do not act as cloud condensation nuclei in our model, but can be ice nuclei.’ Thank you for pointing out this error in the text.

Q. The description of Fig. 12 and Fig. 13 leads to the conclusions that the cloud properties are less affected by the aerosol. Can this be affected by nudging as well?

p. 7890-7890 We have added a discussion to the first paragraph of Section 3.4 noting that our study cannot be a true feedback study since the nudging does also contribute to the differences in the clouds and precipitation that occur between simulations. Thus, caution should be exercised in interpreting the feedbacks, we can only show what can happen in the framework of nudged simulations, and we wish to examine this to clarify how greatly our deposition results might be influenced by these feedbacks in addition to the revisions to our revised below-cloud scavenging parameterizations. We present Fig. 13 to show that the cloud and precipitation in our model are reasonable. We do not want the reader to conclude that cloud properties are less affected by aerosols since, as the referee indicates, the nudging may reduce the magnitude of these feedbacks and we have tried to make this point more clearly in Section 3.4. At the end of Section 3.4 we have also included the acknowledgement that these small scale local events can increase and modify the climate system due to model non-linearities, and can influence dust mobilization (citing Northern Africa as an example) as the referee has noted.

Q. How does the comparison look like for the other simulations?

p. 7892, l. 7-14 We have compared all simulations with the NADP observations, and the results are very similar between all simulations. Thus, we did not add a table since these results differ very little between simulations. We have added a sentence to Section 3.5 to indicate that we completed these comparisons and found similar agreement...
to the results shown. The simulation with thermophoresis did about equally as well as
the BCS2 simulation for sulfate, and sea salt deposition with the prognostic scheme
was not significantly different in comparison with the observations.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 7873, 2009.