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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Sources of uncertainties in modelling Black Carbon at the global scale" *by* E. Vignati et al.

Anonymous Referee #1

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This paper examines uncertainties in model processes affected the black carbon aging and its removal rate. Using a chemical transport model. The paper tests two treatments of aging, a fixed removal rate and one based on some interactions in the model. The two treatments give a significantly different result from each other in the global average, but close to source regions, concentrations are found to be similar in both cases. The authors find a better agreement with observations with the more complex approach. The authors also test scaling wet removal rates to estimate resulting effects and find a difference.

Overall, I find that this study uses treatments of particle aging and wet removal that are much less rigorous than the state of the art in modeling, so cannot give a reliable answer to the questions raised. Aside from the physical processes represented/numerical

methods used, further evidence of the problems with the treatment is the fact that results are significantly different from that of a more detailed model and from what we would expect physically. Also, the study uses a chemical transport model that cannot account for feedbacks of processes, further raising questions about the accuracy of the conclusions since feedbacks would alter the rates of precipitation and internal mixing. As I believe a publication should encompass not only new information but information that represents an advance over what has been published previously, I recommend against publication of the present results, which will certainly change upon modification of the model, possibly drastically. More detailed comments are given below.

1) First, the empirical equation for scavenging efficiency (Equation A1, Appendix) is arbitrary, as it depends only on the precipitation rate and does not consider the hygroscopicity of particles or the probability of collision of particles of one size versus those of another with precipitation particles of different size. As at least some global models treat the size and composition dependence of aerosol particle removal by precipitation particles of different size (e.g., Jacobson, 2004, JGR 109, D21201, doi:10.1029/2004JD004945 using the methodology in JGR 108, doi:10.1029/2002JD002691, 2003), it is difficult to see how the use of an empirical scavenging efficiency represents an advance over previous work.

2) The difference in methodology (both the use of a CTM versus a coupled climate-CTM and the use of empirical removal treatment) most likely contributes to the significant difference in result between the present study and the Jacobson (2004) result, wherin the author found that wet deposition removed > 90% of black carbon. Here, the result using an empirical treatment was 40-70%. It seems implausible that dry deposition could account for the remainder (60-30%) of BC removal from the atmosphere as most BC particles are too small to sediment noticeably. There is little discussion of how dry deposition is actually calculated in the model and no discussion to how it is physically plausible for up to 60% of BC to be removed by dry deposition.

3) Further, for the model to predict wet removal accurately, it is necessary for the pre-

cipitation rate to be accurate. The authors need to show the global distribution of precipitation and compare this with observed precipitation.

4) With respect to aerosol aging, the present study uses the model of Vignati et al. (2005). This represents an improvement over the schemes used in many global models but is still an over simplification, as it treats modes instead of discrete size bins, which some other models treat (e.g., Gong et al., 2003, JGR 108, doi:1029/2001JD002002; Jacobson 2002 cited in the authors' manuscript). Gong et al. (2003) found that a minimum number of sections is needed to represent aerosol microphysical processes reasonably, more than the few modes treated here.

5) The present model assumes a single coagulation kernel when coagulation between modes is considered, whereas in reality, modes consist of particles of different composition and size, where the coagulation kernel varies as a function of size and composition. Vignati et al. (2005) shows a comparison of the coagulation scheme against an analytical solution for total number, but in that scheme, the coagulation kernel is constant and the actual size distribution is not shown. The authors should at a minimum show a realistic size distribution and compare the modal treatment of coagulation with a sectional and/or analytical solution when the coagulation kernel in the sectional treatment varies as a function of particle size (thus, the modal coagulation kernel must represent some integrated value over the section value).

6) Treatment of condensation with a modal method is also a problem, as condensation varies as a function of particle size and composition. A sectional method can account for Raoult's law and the Kelvin effect, but a modal method cannot. The authors need to discuss and quantify to the best extent they can the inherent errors in treating condensation with a modal scheme versus a sectional scheme in cases other than ideal cases (e.g., Vignati et al., 2005).

7) The paper states that "The model transport has been extensively validated using Rn and SF6." However, there is no discussion of the actual advection scheme used

nor whether it has been evaluated even in one dimension against peak-preserving schemes. If no reference for such a comparison exists, the authors should perform a comparison and present the results. Transport schemes in global CTMs are notoriously diffusive, often losing 30% of their peaks in 50 grid cells of advection. Rn and SF6 tests are not sufficient for determining diffusivity of such schemes. The authors should also show the modeled globally-averaged vertical distribution of black carbon to illustrate whether the vertical profile is diffusive or not.

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