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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Cloud processing, cloud evaporation and Angström exponent" *by* G.-J. Roelofs and V. Kamphuis

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Response to the reviewers of *Cloud processing, cloud evaporation and Angström exponent* by Geert-Jan Roelofs and Vincent Kamphuis

We want to thank both reviewers for their careful reading of the manuscript and their helpful questions and comments. Based on their suggestions we have altered the manuscript, as described below.

Reviewer 1.

Specific comments.

1. We have mentioned the wavelengths used for calculation of the Angström exponent (i.e., 553 and 855 nm) more prominently than in the first version (see section 2, last paragraph).

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2. The single refractive index of 1.333 is associated with pure water. For solutions, the value is a little larger, but not enough to significantly change our results. We have mentioned this in section 2, last paragraph.

3. The first version of the manuscript was not complete in its description of the base case aerosol. The aerosol size distribution in the base case simulations is representative of marine conditions, hence the relatively large coarse mode fraction, but with a moderate influence of pollution, expressed into enhanced fine mode particle concentrations as compared to a typical marine aerosol population (see Table 1). We have added simulations for aerosol representative of different pollution levels for comparison, discussed in section 3.3.

4. We have included a figure with the liquid water volume distributions of the base case simulations (Figure 3).

5. This comment motivated us to include a comparison of simulations for different pollution levels. The results indicate that the RH-dependency and the impact of cloud processing and of kinetic limitations differ significantly between clean and polluted atmospheres, and we discussed this in the light of the contradictory observations for marine and continental aerosol, referring to the work of Loeb and Schuster (2008) and Koren et al. (2007) (section 3.3).

Reviewer 2.

Reviewer 2 asks for a clearer description of the intention of our paper. It is an exploratory study that investigates and describes the influences of the aerosol-humidity effects on aerosol size spectra and their significance for AOT and AE. In the first version of the manuscript we suggested that the Angström exponent could be used to detect cloud processing of aerosol. However, based on the additional simulations for the revision of the paper we concluded that the combined aerosol-water interactions exert a relatively complex influence, highly dependent on characteristics of the aerosol population, on the optical parameters and that further research is necessary to untan-

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gle these influences from aerosol optical thickness measurements. Nevertheless, our results may be relevant for studies on, for example, improvement of emission inventories or estimation of aerosol climate forcings. These studies greatly benefit from more accurate retrieval of aerosol parameters from satellite observations and from surface remote sensing.

Specific comments.

1. We have added more information on the Angstrom exponent and its typical values in the introduction (section 1).

2. We have altered the reference.

3. The model uses a detailed bin representation for the aerosol size distribution. Log-normal modes are used only to initialize the model, as explained now in more detail in section 2.

4. When compared to the theoretical typical aerosol populations for different pollution levels from Whitby (1978; see also Table 1), the initial aerosol size distribution in the base case simulations is representative of marine aerosol, hence the relatively large coarse mode fraction, but with a moderate influence of pollution, expressed into enhanced fine mode particle concentrations as compared to a typical marine aerosol population (see Table 1). It is not based on observations.

5. The model is capable of considering external mixtures of aerosol, e.g., a fine aerosol mode comprised of ammonium bisulfate and a coarse mode comprised of sea salt. However, several size distributions must be considered for this and simulations require a longer simulation time, mainly because of collision/coalescence calculations. Soluble coarse mode aerosol, regardless whether it consists of ammonium bisulfate or sea salt, exerts a strong Raoult (solute) effect and activates easily to cloud drops during cloud formation. In that sense there are no significant consequences for cloud development when a different soluble species is used to initialize coarse mode aerosol. We have

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discussed this now in section 2.

6. We use 533 and 855 nm. We have added a comparison with Angstrom exponents calculated with wavelengths used by different measurement platforms currently used in atmospheric research, and found that most significant differences occur at relatively small RH (see section 3.1).

7. We have added several figures for the sensitivity studies (Figure 5).

8. We have replaced this simulation by a series of simulations that are initialized on typical aerosol size distributions for different environmental pollution levels (Table 1), ranging from marine to heavily polluted, and discussed the results in more detail than in the first manuscript (section 3.3).

9. The AERONET data presented by Koren et al. (2007) reflect relatively polluted conditions influenced by biomass burning. The observed Angström exponent agrees well with values computed in our aged urban pollution simulation, at 85% RH (section 3.3).

10. In the first version of the manuscript we suggested the Angström exponent may be used to detect cloud processing of aerosol. However, this was premature. The additional simulations performed for this revision demonstrated that the combined aerosol-water interactions exert a rather complex influence on the optical parameters, and that further research is necessary to untangle these influences from aerosol optical thickness measurements. We have altered the discussion accordingly (section 4).

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 12721, 2008.