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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol nucleation over oceans and the role of galactic cosmic rays" *by* J. Kazil et al.

Anonymous Referee #1

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General:

The authors simulate aerosol formation (nucleation) over global oceans via two pathways: neutral and charged binary sulfuric acid-water nucleation. Very few global-scale simulations on atmospheric aerosol formation have been published so far, so in this respect the paper can be considered original. The paper is, in general well written and it gives a useful contribution to scientific community. I have, however, one major comment that the authors should consider carefully before the manuscript can be accepted for publication.

Major comment:

The authors start their simulation from an aerosol-free atmosphere. This assump-

tion, while understandable when considering the large uncertainties associated with explicitly simulating the whole atmospheric aerosol population, leads to a few incorrect statements and conclusions throughout the paper. The authors should definitely do something for these points.

First, in the beginning of section 4.1 (page 5549, lines 11-12) it is stated that “Ĕ., and allow us to compare the potential of different regions in the troposphere to produce aerosol”. I do not agree on this statement. In their nucleation mechanisms, the concentration of gaseous sulfuric acid plays a central role in determining the nucleation rate. The sulfuric acid concentration, in turn, is directly proportional to its production term (effectively SO2 times OH concentration) and inversely proportional to its sink term (pre-existing aerosol surface area). Since the pre-existing aerosol surface area varies by several orders of magnitude in the atmosphere, starting the simulations from an aerosol free atmosphere does not give a fair ranking between different tropospheric regions in their potential for forming new aerosol particles.

Second, given the importance of gaseous sulfuric acid in their nucleation mechanisms, it is surprising that the authors do provide pictures of simulated global sulfuric acid concentration, while doing so for SO2, OH and ionization rate (Figs 1, 2 and 4). I would recommend the author to add such a figure and to make a brief overview on how the predicted sulfuric acid compare with available atmospheric measurements.

Third, it is stated that ternary nucleation in the marine boundary layer is suppressed by the lack of ammonia (abstract, page 5561). I am not convinced that this is true. It is more likely that the large aerosol surface area provided by sea-salt particles suppresses nucleation in the MBL by acting as an effective sink for both gaseous sulfuric acid and nanometer-size nuclei.

Finally, the authors could easily estimate how sensitive their results are to pre-existing aerosols by making a sensitivity study in which an additional sink term for gaseous sulfuric acid (imitating the effect of pre-existing particles) would be present. These sink

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terms do not need to be accurate, single values representing “typical” condensation sinks in different tropospheric regions (MBL, middle and upper free troposphere) would be enough.

Minor/technical comments

The global OH concentration field is calculated with a photochemical model (Figure 2). How well this model performs when compared to observations/empirical estimates of global OH concentrations?

Satellites are able to “see” only particles that are much larger than those formed recently in the atmosphere. Therefore, relations obtained from satellite measurement (like aerosol column number concentrations) may not be applicable at all when trying to estimate the climatic effects of aerosols formed by atmospheric nucleation.

The order of pictures in Figures 1, 2, 4, 5 and 6 is strange.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 5543, 2006.