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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Contributions of anthropogenic and natural sources of sulfur to SO_2, H_2SO_4(g) and nanoparticle formation" *by* D. D. Lucas and H. Akimoto

Anonymous Referee #1

Received and published: 18 June 2007

General:

This manuscript deals with a very important issue, namely atmospheric aerosol formation associated with natural and anthropogenic sulfur emissions. My opinion on this paper follows to a large extend that by the other anonymous reviewer, so I will detail only the comments that somehow differ from the other review report. After considering carefully these comments, the paper should be acceptable for publication in ACP.

Major comments:

While sections 1-4 of the manuscript are in general very well written, there are a num-

ber of issues in section 5 that require some refinement. Some of these issues need to be taken into account in other sections as well.

The first issue is the role of other trace gases in new particle formation. It is well known that at least over continental areas, aerosol formation depends not only on sulfuric acid concentration but also of other trace gases such as ammonia and low-volatile organic compounds. This should be brought up explicitly in introduction (lines 5-10 on page 7683), in Table 1 and associated text on page 7686, in the discussion on page 7687 (lines 15-20), and perhaps also in conclusions. The fact that these co-pollutants are emitted from a surface certainly assist new particle formation taking place in the boundary layer, while from the text one too easily gets the impression that SO2 need to be transported away from source areas to initiate new particle formation.

Second, I do not think that the nucleation rate is a proper measure when discussing the relative importance of different SO2 sources to atmospheric aerosol formation and the relative importance of different nucleation mechanisms. There are at least two reasons for this: 1) ternary nucleation rate is directly proportional to the arbitrary chosen "scaling factor". Here it was chose to be 0.0001, but in principle it could be something totally different. 2) depending on their growth rates and the available particle surface area, the probability that nucleated particles survive to give a significant contributing to the particle number concentration budget varies a lot between different atmospheric compartments. As a result of these two points, comparisons should be based on particle number concentrations resulting from nucleation and associated processes, not on actual nucleation rates.

In the analysis starting from page 7700, the authors have assumed that the NP formation rate is proportional to the sulfuric acid concentration to the power 3. Why this assumptions? All available atmospheric observations refer to a power law dependence that is in the range 1-2.

I am quite surprised that no comparison to another, closely-related global modeling

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study on atmospheric aerosol formation (Spracklen et al., 2006, Atmos. Chem. Phys.) has been presented in the paper.

Minor comments:

Page 7688, line 4: DMS may lead to high H2SO4 concentrations but the levels of SO2 are practically always very low as compared with continental regions.

Page 7688, lines 15-25: there are number of studies dealing with aerosol formation associated with convective uplift and cloud outflows in the free troposphere. At least some of these studies should be mentioned here.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 7679, 2007.