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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol nucleation spikes in the planetary boundary layer" *by* J.-P. Chen et al.

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We are grateful to the reviewer's constructive comments, and have done our best to incorporate the suggested changes. In the following we answer the reviewer's questions in detail.

1. I was very interested in reading this paper and the idea behind sounds logical. However, also this is a theoretical study and the production of the nucleation rates or the nucleation theory is not the main goal of the paper I cannot understand how the model with a sulfuric acid based binary nucleation can reach this high nucleation rates with values up 1000 and higher. I would like to see at least some more numbers concerning the assumed sulfuric acid concentrations in the model runs.

Reply: We do realize the large uncertainties in the binary nucleation theory, as stated on p.7 of the original manuscript. We also briefly discussed that, by considering an extra factor – the curvature effect on surface tension, we can get a nucleation rate that is about two orders of magnitude higher, similar to what one would get by considering the hydrate effect. But such an uncertainty does not significantly affect our results as the reviewer suspected. To further proof that, we performed a sensitivity test by artificially multiplying the nucleation by 0.1 and 10 (see Fig. A below). The results show that for the two orders of magnitude change, the number of total aerosols varied by less than a factor of 2, and the variations in D>10 particles are even less significant. A summary of this result has been added to the end of the paragraph. In our simulation the sulfuric acid concentration is not assumed but calculated with simplified photochemistry (see top paragraph on p. 7). The peak value that we calculated (see Fig. 6) is about 0.3 ppt or 0.7x107 cm-3. This value is a bit lower than the daily peak value of 107 $\sim$ 109 cm-3 measured by Kuang et al. [2008]. The lower values might be due to the high relative humidity in the studied area, under which the absorption of sulfuric acid by the aerosols tends to be stronger. Yet, the reviewer is correct to point out that other condensable gases might be involved in the particle growth in the real atmosphere.

Kuang, C., McMurry, P. H., McCormick, A. V., and Eisele, F. L. (2008) Dependence of nucleation rates on sulfuric acid vapor concentration in diverse atmospheric locations. J. Geophys. Res., 110, D10209, doi:10.1029/2007JD009253.

2. Keeping this as a theoretical study with the focus on explaining the peaks it should be clearly pointed out that a lot of simplifications like e.g. the missing effect in the particle growth by organic vapors in the aerosol dynamic have been assumed. The definition of fresh particles by particles below 30 nm is critical. The authors never mention any growth rates although they should get the data from their measurements easily. Assuming an average growth rate of 5-10 nm per hour the so called fresh particles are at least 3 hours old. By discussing the effect of turbulence I'm a little surprised that the model run show such a strong influence on the particles above 10

nm. If the authors only take sulfuric acid to grow the particles I wonder how this could fit and this is a second reason to see the assumed sulfuric acid concentrations. This is a little bit difficult to follow because we know from many publications that sulfuric acid only contributes a small fraction of the particle growth. Anyway the aim of the study is not on the growth mechanism but it should be clarified otherwise readers would be completely lost. By ignoring organic molecules in your theoretical study you have to inform the reader that this will not have a strong effect on the peak appearance but was kept simplified in this manuscript.

Reply: Although for the case shown in Fig. 1 the winds came from the open ocean, it is still possible that the airmass contained some organic gases originated from the Northeast Asian continent, but their concentrations are probably not high enough to make significant differences. We have noted this and the possible role of organics in the revision. The mass growth of aerosols comes not only from sulfuric acid but also water vapor. In fact, the largest mass fraction of our simulated aerosols is water, not sulfuric acid. Also, Brownian coagulation may also contribute significantly to the growth of particles > 30 nm. In the eddy turbulence, the growth can be further enhanced due to the strong cooling/warming effects

3. One aspect I do not like in the article is that the authors in some parts of the paper not using the well-defined definitions for particles but create their own. This is done at several places in the manuscript and I can only advice the authors to change it so the text will be easier to understand. E.g. page 26933 line 9: nuclei mode - > nucleation mode; line 10: old particles -> pre-existing particles;

Reply: Nuclei mode and nucleation mode are both commonly used terms. We have not thought much about the differences, but now we think “nucleation mode: is a more appropriate term in terms of the physical meaning. The “pre-existing particles” is also a better terms. So we gladly comply with the reviewer and made the changes accordingly.

4. All figures are very difficult to view because of too small fonts used.

Reply: All fonts have been enlarged.

5. In figure 1 and 6 the text should be corrected after using colors.

Reply: The color change was made during the manuscript typesetting and production because some of the curves do not come out correctly. But we forgot that the text also needs to be changed. Much thanks for pointing that out. The corrections have been made.

6. Figure 6a and 6b should have the same time period from 11 to 12.

Reply: Corrected.

7. I would also like to see the temperature change in the air parcel added in figure 6a.

Reply: The temperature curve has been added.

8. Page 26934 line 26: requires -> required

Reply: Corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 26931, 2010.

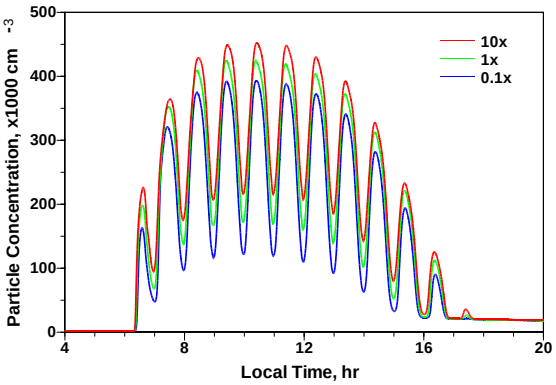

Figure A: Variation of total aerosol concentration when the binary nucleation rates are altered artificially by 10 and 0.1 times. The simulation setup is similar to that for Fig. 4, except that a longer cycling period of 1 hour and a shorter vertical amplitude of 250 m were used to make the figure more legible.

Fig. 1.