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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluation on the role of sulfuric acid in the mechanisms of new particle formation for Beijing case" *by* Z. B. Wang et al.

Anonymous Referee #1

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General:

This manuscript analyses the connection between sulphuric acid and new particle formation in a sulfur-rich polluted environment. The analysis gives a very nice addition to previous analyses performed at cleaner sites. The paper is worth to be published in ACP after some substantial revisions. Especially, the authors have made a few conclusions that should be sharpened. Furthermore, recent literature has brought up new findings that should be mentioned and briefly discussed in this manuscript. My detailed comments in this regard are given below.

Major comments:

Section 2.2.1. The recent analysis by Korhonen et al. (2011, ACP, pages 3051-3066)

points out several problems that may arise when the cluster growth rate between 1.5 and 3 nm (GR) is estimated from the time delay between the increase in H2SO4 concentration and that of 3-6 nm particle concentration. The authors should discuss this issue further in section 2.2.1 by considering these findings. Most importantly, Korhonen et al. (2011) showed that a zero time delay, as observed in some of the cases here, may not necessarily indicate very fast growth of nucleated clusters.

Page 24171, lines 13-16. I do not understand why the authors give specifically the exponent 3 here? Later on they provide exponents with broad range of values. Would it rather be better to say that exponents for J1.5 vs. H2SO4 clearly in excess of 2 are indicative some degree of thermodynamic influence on the nucleation process. I suggest that the authors replace "3" with "n>2.5" consistent with their analysis presented later in the paper.

Page 24172, lines 14-27. The authors conclude that it is the pre-existing aerosol concentration rather than gaseous sulphuric acid concentration determining the occurrence of NPF at the site. Is this a firm conclusion? I have a difficulty in following the reasoning of this conclusion.

Page 24174, lines 18-20. This is another conclusion I have a difficulty in understanding based on available data.

The recent analysis by Sihto et al. (2009, ACP, pages 2933-2947), Vuollekoski et al. (2010, Atmospheric Research 98, pages 229-236) and Korhonen et al. (2011) using aerosol dynamical model simulations challenge the interpretations that can be made based on the relation between the "nucleation exponent derived from atmospheric observations" and the "nucleation mechanism". Most importantly, these simulations show that 1) the connection between the "real" and "observed" nucleation exponent is more complicated than previously thought, and 2) on average, observed nucleation exponents tend to be on higher than the real ones, i.e. activation mechanism would typically produce "observed" exponents substantially larger than unity and kinetic mechanism

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would produce "observed" exponents typically larger than 2. The authors should carefully consider these results when discussing their findings in sections 3.2, 3.3, 3.4 and 4, as well as in abstract.

Minor comments:

Section 2.1. Have the authors made any estimates on the accuracy of the gaseous H2SO4 measurements? If this information is available, please provide it here and give a possible citation to work in which such an estimate have been made.

Page 24170, line 9. The assumption made here should read GR6=GR1.5-3. Since there are potential problems in determining GR1.5-3 (see my first major comment) and since GR1.5-3 may not be a good representative for GR6 due to size-dependencies in cluster growth rates, why did not the authors try to estimate GR6 from particle number size distribution measurements? I would think that the growth rate of the nucleation mode would be closer to GR6 than the highly inaccurate value of GR1.5-3.

Page 24167, lines 16-17. Could the authors provide some examples of observations of NPF in urban polluted environments?

Page 24171, lines 2-3. Please add some reference for the kinetic cluster formation theory.

Page 24172, lines 3-6. The authors are certainly correct but based solely on Figure 1, it is hard to see that N3-6 and H2SO4 have similar trends on NPF days.

Section 3.4. The authors should mention that the nucleation coefficients A and K were derived for all the events regardless of which nucleation mechanisms obtained for that particular event.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 24165, 2011.