

Interactive comment on “Why formation rate of 3 nm particles depends linearly on sulphuric acid concentration?” by M. Kulmala et al.

Anonymous Referee #1

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Evidence is presented which supports the argument that new particle formation rates in the atmosphere of a boreal forest vary in proportion to the sulfuric acid concentration raised to some power from one to two. From the nucleation theorem it is concluded that the critical cluster contains one to two sulphuric acid molecules. The paper argues that the first power dependence might be explained by activation processes that could include heterogeneous chemical reactions that might involve polymerization or heterogeneous nucleation, and that could typically involve organic compounds. This hypothesis (i.e., the proposed “activation theory”) is the focus of the paper. Previous work, some of which is cited in the paper, shows that a squared dependence on sulfuric acid concentration would occur if new particle formation were limited by the rate at

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which sulfuric acid vapor molecules collide.

This an interesting hypothesis, and it might be right. The challenge, of course, will be do the hard work required to show that it is right. This hard work will involve the development of new measurement methods, and will likely take some years.

Speaking of hard work, two types of data were required for developing this hypothesis: number concentrations of 3 to 6 nm particles and sulfuric acid vapor concentrations. I was unable to find a reference to the individuals who carried out these measurements or to the methodologies that were employed. Given that the hypothesis could not be proposed without these data, I feel that this is an oversight needs to be corrected.

I would be more comfortable if the paper were to include a detailed analysis of all data collected by this research team that explores the relationship between new particle formation rates and sulfuric acid concentrations. The results shown in Figures 2 and 3 are interesting, but apply to only a single day. The text states “Not all event days during the campaign show such a clear linear dependence. We can, however, confidently state that in all cases the power-law exponent is between one and two.” Why not present all of the evidence? The paper would be much stronger if this were done. Are some days more nearly consistent with a squared dependence, while others are more nearly consistent with a first power dependence? What is the logic for shifting all data by the same time shift (90 minutes, for the data shown in Figure 2) when concentrations of condensing vapors (and therefore, perhaps, particle growth rates) may vary by a factor or ten or more during the course of a day? I feel that these are substantive issues that ought to be explored in more depth. I think such an analysis would provide a firmer scientific basis for the proposed activation theory.

I recommend that the paper be focused on scientifically sound observations and established scientific principles. In my view these include the nucleation theorem and the observed relationship between $N(3-6)$ and sulfuric acid concentrations. I would then include in the discussion the proposed activation theory, which is one possible expla-

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nation. I have reservations about having the activation theory as the primary focus of the paper. The hypothesis could serve as the bases for a wonderful research proposal, but I don't think it should be the primary focus of a journal article at this early stage.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 11277, 2005.

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