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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Boundary layer nucleation as a source of new CCN in savannah environment" *by* L. Laakso et al.

Anonymous Referee #1

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This study compared global model simulation of aerosol nucleation events with the 18-months observations conducted in a site over the South African savannah region. The observation data have been discussed in another paper, so the focus of this study is to deduce aerosol formation and growth mechanisms from the difference between model and observation results. The subject is certainly interesting and suitable for publication in ACP. However, the main conclusion that biogenic organic vapor is an important source of aerosol for this region is not strongly supported by their analysis. The authors should try to rule out other possibilities as well as evaluate the uncertainties in the model. This manuscript may be considered for publication if the following comments can be responded satisfactorily.

Major comment:

1. A main point of this paper is to relate the underestimation of aerosol production and growth in the model to the omission of organic vapor. I have quite a few concerns about this assumption (seems to be regarded as a conclusion in this paper). (1) No direct supporting evidence is provided. For example, is there any measurement of secondary organics in the aerosol? (2) The model that used seems to consider OC aerosol emission (p. 5). Does it consider mechanisms for organic aerosol formation? If so, then a sensitivity test could further elucidate the importance of organics. Figure 4a shows the fraction of growth from H2SO4. What are the rest of the fraction composed of (any information on OC)? (3) The evolution of size spectrum is not totally controlled by nucleation and condensation. Other factors, such as coagulation and emission are also important, and the inaccuracy in them may contribute to the discrepancies in Figs. 3-5. For example, the biomass burning emission used in the simulation is based on climatological satellite data, but the real biomass burning evens are usually sporadic and have strong inter-annual variations. Such an uncertainty might invalidate the suggestion of a strong contribution from organic vapor (e. g., the model may simply underestimated biomass emission). The uncertainty in biomass emission was mentioned in section 3.3 to explain the difference in CCN-sized particles, and this should be applied to Fig. 3 also. (4) The general underestimation of particle production and growth in Fig. 3 could also be due to the inaccuracy of the model physics. For example, Chen et al. (2011, Atmos. Chem. Phys., 11, 7171–7184) argued that surface tension decreases with particle size, and ignoring this effect would result in underestimation in both nucleation rate and condensation rate. Also, the lack of RH dependence in Eq. 1 could lead to a lack of minimum in the particle production during the dry season.

2. p.3, 8-5 lines to the bottom: I do not have access to the Zhou (2001) thesis and thus don't know why there should exist a 50% RH threshold. The hygroscopic growth (or the threshold of it) depends on chemical composition as well as whether efflorescence occurs. For example, sulfuric acid solution does not dehydrate under typical conditions, so there is no RH limit for the hygroscopic growth such droplets.

3. p. 5, Eq. 1: Eq. 1 seems to be very crude. According to the nucleation theory, H2SO4-H2O binary nucleation is a strong function not only [H2SO4] but also relative humidity and temperature. This could be part of the reason why the modeled fresh particle formation rates do not show a minimum in the dry season as in the observation. Another question is why it is necessary to use a different scheme for the boundary layer than for the free-troposphere since both assume H2SO4-H2O binary nucleation?

Minor comments:

1. p.4, line 17-18: “minimum, mean and maximum sizes that growing particles reach during the event” How do you define “growing particle”? Some of the particles might be pre-existing. Are you also assuming that there is no atmospheric advection or spatial inhomogeneity?

2. Fig. 3 This figure shows the seasonality of particle formation or growth rates as a means to verify model results. The seasonality is a rather crude parameter in relating to the particle formation mechanisms. Why not show the dependence of “particle formation rate” on more direct physical or chemical parameters such as SO2 concentration, solar radiation etc.?

3. p. 7, last paragraph: It is unclear what the “dip in accumulation mode” means. Are the authors referring to the dip in the tri-modal (or multi-modal) distribution that suggested by Whitby? The authors seem to relate the missing of the “dip” to wet removal (specifically rain scavenging). If the authors are referring to the Greenfield gap concept, then the scavenging process should produce a maximum in the accumulation mode, not a dip. More detailed explanation is needed for general readers to understand what message the authors are trying to convey.

4. It would be helpful if the authors can demonstrate the performance of the aerosol model by showing the simulated evolution of size spectrum and compare it with Fig. 1.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 8503, 2012.