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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Formation and growth of nucleated particles into cloud condensation nuclei: model-measurement comparison" *by* D. M. Westervelt et al.

Anonymous Referee #2

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This paper represents an interesting model vs. measurement comparison related to cloud condensation nuclei formation due to atmospheric nucleation. The paper is suitable for publication in ACP after the authors have addressed the few issues raised below.

The authors have chosen to include a long, review-type introduction in their paper. I am fine with such an approach, but with the reservation that it should be made more carefully than done here. Firstly, I do not think that the chosen literature reflects the current understanding on atmospheric nucleation mechanisms, nuclei fate (growth vs. scavenging) and resulting CCN production to the extent that would be desirable for

this kind of an introduction. Secondly, the text should not contain loose statements, or statements that may not be right. For example, is there a firm basis to claim that nucleated cluster are stable in the atmosphere and that their initial size is typically 1 nm (page 8336, lines 13-14)? Third, I have a hard time of seeing how large nucleation rates and nucleation probabilities as low as 10 to the power -8 would be related to each other as stated on page 8337, lines 9-13. Think an extreme case where nucleated particles grow solely by their self-coagulation, not by condensation at all. It takes a thousand 1 nm particles to make one 10 nm particle, so the survival probability against self-coagulation would be 0.001 at 10 nm and 10 to the power -6 at 100 nm. Lower survival probabilities are possible only if larger pre-existing particles are the main sink for growing nuclei, but if this is the case then the survival probability would be independent of the nucleation rate (as nuclei themselves do not contribute to the sink). Forth, is there some specific reason to select these two sites (Hyytiala in Finland and South Africa) when discussing observed particle growth rates? Certainly, both clearly smaller and larger growth rates have been observed in other locations. Finally, what is meant by CCN sensitivity to nucleation and can it be measured using percentages? Normally, sensitivity means how much a change in one quantity affect a change in another quantity.

The survival probability requires some clarifications. First, the definition of this quantity in section 2.5.4 includes coagulation losses only. The authors should tell the readers that this is not necessarily the real survival property of the nucleated particles because they may be removed by other ways before reaching CCN sizes. For example, in many cases removal by wet scavenging is much more efficient than removal by coagulation. Second, how the survival probability is determined in practice from measurements and model simulations? By determining the relevant time scales from observations and simulations and then using equation 6, or by trying to determine this quantity somehow more directly from observations and simulations? What are the related uncertainties in calculated survival probabilities and do these uncertainties affect the model-simulation comparisons in Figures 5-9? Thirdly, nucleated particles grow, on average, too slowly

too reach 100 nm within one day at most of the sites? The authors mention this briefly, but do not discuss the consequences of this fact on the results and their interpretation.

Figure 3 and related text: depending on the assumed nucleation mechanism and site, the model predicts whether nucleation takes place or not with 50 to 64% probability. Noting that purely random guess should produce 50% probability of being right, these number do not sound very large. The authors should open up this issue a bit further and not just state about the overall success of the model (page 8360, line 5).

Most of the measurement sites used in the analysis are kind of urban locations, and no remote site is included. This should be mentioned somewhere in the text. Does this fact have any consequences on how the main conclusions can be generalized?

Minor issues

Beginning of section 2.2.1: EC is not part of the organic aerosol.

Section 2.2.2: The authors should mention briefly that organic may influence not only the growth rate but also the nucleation rate.

page 8351, line7: Seinfeld and Pandis (2006), not 2005.

page 8357, line 7: what is meant by feedback between nucleation rate and condensation sink?

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 8333, 2013.