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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evidence of an elevated source of nucleation based on model simulations and data from the NIFTy experiment" *by* P. Crippa et al.

Anonymous Referee #2

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The authors present an interpretation of near-surface tower measurements of class A nucleation events made during the NIFTy field experiment in southern Indiana at a forest site. The interpretation is largely based on the skill of UHMA model simulations in replicating the observations, but are also supported by arguments based on the observations. If the case for a source of nucleation aloft were proven, the paper would certainly be worth publication. The arguments seem to provide reasonable support for the hypothesis, but appear far from incontrovertible proof. If the authors are able to address the points below, the manuscript may be suitable for publication.

Observational evidence for an elevated source of new particles is stated as i) the great-

est increase in concentration of 6-10 nm particles being earlier than the peak of nucleation intensity at the 46m tower height and corresponds to the sharp increase in turbulence intensity associated with breakdown of the nocturnal inversion, ii) the lack of correspondence of near surface condensation sink just prior to the largest rate of change of ultrafine particles and the concentration of ultrafine particles, iii) low backscatter layer (inferring a low CS) associated with the inversion and residual layer and iv) a closed size distribution inferring that the smaller particles were not transported from where they were formed, only the resulting larger particles. All the above may reasonably be stated as consistent with the hypothesis, though none are proof and may have alternative explanations or play no role in a causal relationship. Also, I encourage the authors to directly address the comment of Kari Lehtinen and Ari Laaksonen to give confidence in their interpretation.

The model simulations show some systematic biases compared with the observations and tuning model parameters to improve the fit is used to infer reasons for the discrepancies. Again, it appears that the arguments are plausible within the limitations of the varied parameters, however none of these provide definitive proof, as acknowledged in the final paragraph of the paper.

I guess that both the observational and model-derived arguments provide the "evidence" that is promised in the title, but it is difficult to evaluate whether the burden of proof is sufficiently advanced by the analyses to warrant publication given some of the limitations of the study. The authors should try to address the following two major uncertainties in the modelling aspects of their analyses in a revised manuscript to better allow such an evaluation.

i) As identified by the other reviewer, a significant problem with the modelling approach relates to the treatment of organic compounds within UHMA. The stated aim of the treatment of organics in the model is to "broadly represent the potential for the production of oxidation products with low volatility that might partition into the particle phase". The condensational growth by low volatility oxidised organic compounds produced at

varying rates by direct ozonolysis or OH attack of a mixture of biogenic precursors of widely varying reactivity, or further OH attack of these first-generation products will not necessarily lead to a growth rate reproducible by the simplistic broad representation employed here. Can the authors place any reasonable bounds on the contribution to growth rate from organics from such processes? Is there any way that the model skill in reproducing growth rate can be used to infer the location of nucleation in the absence of a more realistic and constrained organic oxidation and condensation mechanism?

ii) Has a sensitivity to the nucleation rate parameterisation been conducted? The data from which the collision-controlled mechanism was inferred and the kinetic coefficient and exponent derived shows a great deal of scatter. There is likely a great sensitivity to potential nucleation rates from other fits to the data within the confidence limits. Tied into this is the role that the organic compounds may play in the nucleation mechanism itself. An "activation" type mechanism with n=1 would allow for direct participation of low volatility organics which would control the A constant. The nucleation intensity (and hence use of model skill in reproducing this to infer the location of nucleation) will be very sensitive to the assumed form of expression.

Minor: The figure labels are frequently unreadable without very high magnification (e.g. the location label for the MMSF site in Figure 3)

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 11979, 2012.