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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric nucleation: highlights of the EUCAARI project and future directions" *by* V.-M. Kerminen et al.

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The manuscript gives an excellent overview of the cutting-edge experimental, observational, modeling, and theoretical methods employed in the investigation of atmospheric aerosol nucleation within the EUCAARI project. It is very well written and a pleasure to read. In addition, it is a very helpful resource of information on current understanding of atmospheric aerosol nucleation that will serve well as a reference in the future. My only major criticism is that some statements in the manuscript imply almost global validity (explained below), while the observations are dominated by data from boundary layer sites in Europe: The resulting findings may not necessarily hold in other regions of the world. A brief addition to the text that addresses this eventuality may not hurt. Beyond this and a few other points listed below, the manuscript is in great shape.

1) Generalization of findings

"All the results obtained during EUCAARI indicate that sulphuric acid plays a central role in atmospheric nucleation, in addition to which other vapours, especially organic ones, are needed to explain the nucleation and the subsequent growth processes." (16499/12-15)

Would this also hold in regions of the atmosphere where organic vapors, or e.g. ammonia, are not plentiful? An example would be the Pacific Ocean, and in particular its upper troposphere, where concentrations of organics and ammonia are arguably much lower than over Europe.

"Overall, our observations are indicative of frequent, yet moderate, ion-induced nucleation usually outweighed by much stronger neutral nucleation events in the lower troposphere." (16499/21)

I do not object the finding, but a bit of caution may not hurt: Since the strong neutral nucleation events in the lower troposphere of the EUCAARI domain are likely caused by compounds such as organics, ammonia, amines, etc., it cannot be excluded that in the lower troposphere of regions where these compounds are less plentiful, say over the Southern Ocean, ion-induced nucleation may play a more important role.

2) Nucleation parametrizations

The same argument on generalizing the validity of the results can be made for the nucleation parametrizations (16522/18-25) that have been derived from the EUCAARI field data: Do these work (or should they be expected to work) in environments that are very different (temperature, gas phase composition) from those at the field sites, such as over the open oceans, or in the upper troposphere?

3) Binary sulfuric acid-water nucleation

"The new H2SO4–H2O nucleation experiments are in line with EUCAARI field observations" (16509/25)

The laboratory experiments treat nucleation of H2SO4 and H2O, and yet they are in line with the field observations? That sounds not right. It would mean that either the lab experiments have a problem with impurities (organics ?), which lead to a similar nucleation process as observed in the field, or that the nucleation in the field is due to homogeneous nucleation of H2SO4 and H2O.

"Roughly the same number of H2SO4 molecules in a critical cluster and similar H2SO4 concentration levels needed for a given nucleation rate in both the laboratory and the ambient atmosphere suggests strongly that particles are formed via a similar H2SO4-driven nucleation mechanism in these two environments." (16510/7)

Same thing: The laboratory experiments investigate binary H2SO4/H2O nucleation, but the nucleation process responsible for the nucleation observed in the field is likely a different one, for instance cluster activation. How can the two produce similar nucleation rates at comparable H2SO4 concentrations?

4) Nucleation probability

In equation 3, the exponent in exp(-I*t) seems not to be unitless, as one would expect, is that correct? Also, please give a few more details on the nucleation time t.

5) Coagulation sink

"... is their coagulation sink ..." (16525/1)

Please give a few more details on the coagulation sink (= sink for coagulation on pre-existing aerosol?).

6) Manuscripts in preparation

In several places, manuscripts that are in preparation are cited. I understand the difficulty of timing the publication of the results in a project of the size of EUCAARI, but still, it would be a more robust approach to cite only works that have progressed farther in the publication process. Maybe this is the case by now.

7) Ideology (16505/7)

Maybe there is a better word than ideology.

8) Bipolar ions (16506/3)

This may be just my personal ignorance, but I don't know what bipolar ions are.

9) References

Missing umlaut in the reference Kulmala et al. 2009 (Pöschl) (16532/23)

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 16497, 2010.