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Atmospheric Chemistry and Physics Discussions

Interactive comment on "General overview: European Integrated project on Aerosol Cloud Climate and Air Quality interactions (EUCAARI) – integrating aerosol research from nano to global scales" *by* M. Kulmala et al.

Anonymous Referee #1

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The EUCAARI project provided an umbrella for a wide array of scientific advances in our understanding of atmospheric aerosols and their role in climate. The present paper is an overview of the accomplishments under the project. The paper is very useful in providing a roadmap to the more detailed papers that have already been published and will appear in the future. The comments below pertain to elements of this large overview paper, with the goal of enhancing its usefulness to the community.

One should read the overview with the mission and objectives of EUCAARI in mind (pages 17948-17950). In particular, one notes the ambitious and specific objective of

reduction of the current uncertainty of the impact of aerosol particles on climate by 50%. Readers will undoubtedly want to assess the degree to which this objective has been achieved.

Much of the material summarized in this overview paper has references to peer-reviewed papers in the literature, which the reader may consult for details and results. In some places it is noted that a detailed paper is in preparation. In a number of cases, however, results are simply stated. While a paper will almost certainly be prepared eventually, it is somewhat problematic when results are stated here that have not yet been subject to peer review. Many of these instances will be noted below in this review. Ordinarily, a reference is not cited unless it has actually been submitted for publication. In this overview paper, I think this condition can be relaxed, so that the reader knows that a more detailed paper is in preparation. An example of this is the citation to Sierau et al., 2011 on line 5 of page 17966. Citations like this one can be listed in the References section as "to be submitted" or "in preparation."

A great deal of important results emerged from EUCAARI. It is not necessary in this review to point out each of these advances. The team of researchers is to be congratulated for an extraordinary effort at integration across groups and laboratories. The comments below are organized according to the order in which they appear in the paper. No effort has been made to list minor errors of a typographical nature.

3.2.1 Nucleation and growth: Laboratory experiments (p. 17958-17961) Several of the EUCAARI authors, e.g. Kulmala, are involved in the CLOUD experiment at CERN. That experiment, the first results from which will be reported shortly, offers considerable insight into atmospheric nucleation mechanisms beyond that provided by the collection of papers cited here. Although CLOUD is not part of EUCAARI, there is the danger that the reader will take the results cited in this paper as the last word on nucleation, while the CLOUD results, in many cases, significantly advance our understanding. A paragraph should be added that places the EUCAARI findings on nucleation mechanisms in the context of the more recent CLOUD data. The first CLOUD paper should

appear within the time frame of the preparation of the revised version of this paper for ACP.

p. 17976-80 The atmospheric measurements in India and Brazil appear to be quite important. The reviewer would encourage these results to be prepared for publication.

p. 18001 It is stated that results obtained suggest that the anthropogenic contribution to secondary aerosol formation (both primary and secondary) is dominating in most parts of Europe and that halving SO2 and anthropogenic primary emissions would result in reductions of the order of 20% on the total particle number concentration. Can a peer-reviewed publication be cited for these results? If not, then it is questionable to state these results as fact. In this case and others like it that appear in the paper, unpublished results should be presented with the disclaimer that they have yet to be subject to peer review.

p. 18003, line 23- This paragraph describes a new parameterization that has been derived. Can a reference be cited? While this material would be appropriate in a report to the sponsor, what is the reader supposed to do with this information? One cannot use the parameterization, nor have the parameterization and its results been subject to peer review.

p. 18005, line 12 “1st indirect effect reduced by about 10%.” What does the 10% refer to? Number concentration? Radiative forcing? Please explain more thoroughly.

p. 18009, line 1 – “significant improvement of the agreement between measurements and predictions of regional organic aerosol concentrations” Has a paper been submitted with these results? Without a paper, this is again just an advertisement. As noted, a qualification is needed that these results have yet to be subject to peer review.

p. 18011, line 18- These results on radiative forcing are important and address one of the major goals of EUCAARI. They are described in more detail over the next few pages, but it might be good to provide the key references at this point. The same can

be said for the results beginning on line 28.

p. 18014-6 The result on the sensitivity of simulated indirect aerosol forcing to the presence of an assumed background level of cloud drops seems important. The predictions of the reduction of the simulated aerosol indirect effect due to ice nucleation by soot particles (line 23-) by +0.2 to +0.55 W/m2 seem unusually large with a very large spread. Perhaps the two original references justify the reasons for this in more detail, but simply citing such a large forcing difference and such a large spread for this effect without explanation leaves the reader wondering. Likewise, such a large range of estimates of present-day indirect aerosol forcing from -0.27 to -1.16 W/m2 due to the effect of nucleation is surprising. What is the explanation for this large spread in estimates? It is not clearly stated in fact what is meant by the effect of nucleation in these simulations. Does this mean that nucleation is entirely shut off? What is the explanation for the large increase in present-day forcing?

p. 18023, line 3- Have the updates to ECHAM5-HAM been documented? Even if not documented, this description is important since researchers may want to use ECHAM5-HAM and they need to know its latest features. (These advances do not fall in the category of advertisement.)

4.1.1 Aerosols and climate: reducing uncertainty This section addresses a primary goal of EUCAARI. The project has produced an array of results that ostensibly improve upon those in IPCC AR4. The extent to which the uncertainty has been reduced by 50% is addressed on page 18029, lines 12-17. The authors caution that the conclusion that this reduction has been achieved is based on a limited number of results. The nature of some of the results cited here under EUCAARI, and in fact for many of those in the literature relating to forcing estimates, is to consider one effect in isolation and to report a forcing estimate for that effect. Examples here include those for soot particle effects on freezing and cloud lifetime and of nucleation. (As we study more and more phenomena, the uncertainty bounds on aerosol forcing may even grow with time before they can shrink.) It would seem that the large uncertainty bounds on soot

ice nucleation and on new particle formation by nucleation that are cited here have not been taken into account when claiming that the uncertainty in radiative forcing estimates has been shrunk by 50%. In view of the climate forcing calculations done under EUCAARI, it is questionable whether the 50% goal can really be claimed. What appear to exist here are selected forcing calculations, but not of sufficient breadth to claim that aerosol forcing uncertainty on the whole has been significantly decreased. Perhaps it is beyond the scope of EUCAARI, but an important issue is whether these new results on radiative forcing and climate response will become part of IPCC AR5.

p. 18064, lines 22-26 What can be said about the validity of the semi-empirical nucleation rate parameterization in light of the emerging results from CLOUD? The literature contains a seemingly endless succession of atmospheric nucleation parameterizations, each one claiming to be definitive. Some guidance to the reader would be helpful here.

p. 18067, line 26 In the global modeling study it is stated that isoprene was identified as a major precursor to the formation of glyoxal. That glyoxal is a product of the gas-phase oxidation of isoprene has been known for some time. Probably a better way to state the results of the study in question is that oxidation of dissolved glyoxal is a major source of oxalate, but this also has been known for some time (see work of Ervens, Turpin, Volkamer and others), so this also cannot be claimed as originating in this paper.

p. 18079, final paragraph This paragraph describes results concerning climate effects of black carbon. The lack of any references to work done outside of EUCAARI has the danger of conveying the impression to the reader that the references from EUCAARI are the only ones of importance. The climatic effects of BC are of intense interest, with many recent papers on the subject. It is not necessary to list all these other papers here, but acknowledgment to the array of results in the literature on the climatic effects of BC and the extent to which they agree needs to be made.

Figures 4-11, 16-20 References to peer-reviewed papers, or submitted papers, need to be added to the figure captions. In the cases where the data have not yet been

submitted for publication, that fact needs to be stated in the figure caption.

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