Response to reviewer

Anonymous Referee #1
Received and published: 22 July 2011

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.

Response

We consider that EUCAARI has significantly improved the level of knowledge in this field and it is very difficult to create unbiased estimates of the actual level of uncertainty. We have changed the text accordingly (section 4 in general and 4.1.1)..

Much of the material summarized in this overview paper has references to peerreviewed 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."

Response:

We have updated the references available at the moment and made sure unpublished results are indicated as such.

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.

Response

Thank you for these positive comment

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.

Response

We added a new subsection (3.2.1.4. Future needs) to the end of section 3.2.1, which now addresses the points raised by the reviewer. The subsection contains the following text:

"Although our understanding on atmospheric nucleation has enhanced substantially during EUCAARI, several issues requiring further research can be identified. Firstly, it is unclear whether atmospheric nucleation is dominated by a single nucleation pathway, or whether multiple different mechanisms are competing with each other. Secondly, we have not yet resolved the relative importance of kinetic and thermodynamic factors controlling the atmospheric nucleation rate. Thirdly, the identity and role of organic vapours in the nucleation process are still unknown. Finally, although our results suggest ion-induced nucleation to be of minor significance in continental boundary layers, this may not be the case in the free troposphere or above the oceans.

In order to address the remaining issues and scientific questions, we need to find out how the nucleation rate is connected with the chemical composition, physical properties, evaporation rates and dynamics of the smallest atmospheric clusters. Getting such information required further advances in both experimental tools and theoretical approaches. These include construction and application of highly sensitive and selective instruments capable of operating at the sub-2 nm size range, making laboratory experiments at highly-controlled conditions (e.g. Kirkby et al., 2011), developing a new generation of kinetic molecular-scale models, and using various theoretical approaches relying on both quantum chemistry and classical thermodynamics."

Ref: Kirkby J. et al.: Role of sulphuric acid, ammonia, and galactic cosmic rays in atmospheric aerosol nucleation. Nature, 476, 429-435, 2011.

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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.

Response

We thank you for your encouragement. For e.g. India, we have published lately

A.-P. Hyvärinen, T. Raatikainen, D. Brus, M. Komppula, T. S. Panwar, R. K. Hooda, V. P. Sharma, and H. Lihavainen. Effect of the summer monsoon on aerosols at two measurement stations in Northern India - Part 1: PM and BC concentrations, Atmos. Chem. Phys., 11, 8271-8282, 2011

A.-P. Hyvärinen, T. Raatikainen, M. Komppula, T. Mielonen, A.-M. Sundström, D. Brus, T. S. Panwar, R. K. Hooda, V. P. Sharma, G. de Leeuw, and H. Lihavainen. Effect of the summer monsoon on aerosols at two measurement stations in Northern India - Part 2: Physical and optical properties, Atmos. Chem. Phys., 11, 8283-8294, 2011

K. Neitola, E. Asmi, M. Komppula, A.-P. Hyvärinen, T. Raatikainen, T. S. Panwar, V. P. Sharma, and H. Lihavainen; New particle formation infrequently observed in Himalayan foothills - why? Atmos. Chem. Phys., 11, 8447-8458, 2011

T. Raatikainen, A.-P. Hyvärinen, J. Hatakka, T. S. Panwar, R. K. Hooda, V. P. Sharma, and H. Lihavainen. Comparison of aerosol properties from the Indian Himalayas and the Indo-Gangetic plains. Atmos. Chem. Phys. Discuss., 11, 11417-11453, 2011

(and hopefully we will submit more soon)

We have added the references on suitable place in the text.

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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.

Response

The results that the reviewer is referring to are not yet published in a detailed peer-reviewed study. They are, however, summarized in Table 2 of the present manuscript. We have added a reference to Table 2 in the revised version of the manuscript to the section where the sensitivities of particle number concentrations to natural and anthropogenic contributions are discussed.

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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.

Response

This parameterization is described in more detail in Hoose et al (2010a, AMS conference proceedings), see the reference in line 10 on p 18003. We agree with the referee that these results will have to be subjected to

peer review to make them fully available to the scientific community, and are currently preparing a manuscript.

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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.

Response

We agree with the reviewer, and have now replaced "indirect effect" by "indirect forcing", in agreement with the terminology elsewhere in the paper.

We have also corrected two errors that we found in this sentence: 1) The term "1st" (indirect effect) is wrong, because also the lifetime effect is included in these estimates. In fact, in the text that follows it becomes clear that the lifetime effect is the main reason for the reduction of the indirect forcing. 2) The indirect forcing is in fact reduced by about 25%, not 10% (comparing results from Hoose et al., 2009 to those of Hoose et al., 2010c).

Hence, we have replaced the original sentence

"With the new ice nucleation scheme in place, the simulated aerosol 1st indirect effect is reduced by about 10% compared to simulations that do not treat ice nuclei."

by the following:

With the new ice nucleation scheme in place, the simulated aerosol indirect forcing is reduced by about 25% compared to simulations that do not treat ice nuclei.

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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.

Response

The reviewer is correct - we have not demonstrated a clear imrovement in the sense of comparing to previous studies. We have, however, demonstrated a very good agreement between the modeled and measured organic aerosol concentrations in Fig. 17 of the ACPD manuscript. We will therefore replace the sentence pointed out by the referee with "simulations with the regional model developed within EUCAARI result in very good agreement with measured organic aerosol concentrations on the European continent"

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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.

Response

We have added references

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.

Response

The CAM-Oslo model has a relatively large aerosol indirect effect in liquid clouds of -1.88 W/m2 (difference in SWCF, Hoose et al, 2009). This is due to low background aerosol concentrations over ocean, as discussed in the previous paragraph. Compared to this value, the reduction due to effects in mixed-phase clouds is not particularly large (10 to 26%), and other studies find similar effects (Lohmann, 2002; Storelvmo et al, 2011). It should also be noted that the reduction is not only due to the soot ice nuclei, but also to the introduction of an extended cloud microphysics scheme with a prognostic treatment of ice crystal concentrations. We have added this information to the text: "We have also investigated the influence on the simulated indirect effect of **a prognostic treatment of ice crystal concentrations (Storelvmo et al, 2008) and** ice nucleationby natural and anthropogenic particles (soot). Hoose et al. (2010c) found a reduction in the simulated aerosol indirect effect by about 10 to 26%. The reduction is **mainly** caused by a reduced lifetime effect dueto anthropogenic soot (glaciation effect). This is because soot particles stimulate the freezing of supercooled water, thereby stimulating the release of precipitation from the clouds. The net effect is a reduction in cloud lifetime."

References:
Storelvmo, T., C. Hoose and P. Eriksson (2011): Global modeling of mixed-phase clouds: The albedo and lifetime effects of aerosols. *Journal of Geophysical Research,* 116, D05207, doi:10.1029/2010JD014724
U. Lohmann (2002), A glaciation indirect aerosol effect caused by soot aerosols, *Geophysical Research Letters 29, 4, doi:10.1029/2001GL014357.*

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?

Response

The large spread in the present-day indirect forcing can be partly explained by the spread in modeled global cloud droplet number concentrations: 202 # cm-3 with nucleation and 83 # cm-3 without nucleation. In simulations without nucleation, both binary homogeneous nucleation and boundary layer nucleation are shut down, and hence aerosol numbers arise only from primary emissions. The available sulphuric acid can still condense on existing particles. The spatial distribution of the nucleation-increased CCN establishes a strong effect on indirect forcing. Kazil et al. (2010) showed that the present-day contribution of nucleation on the total aerosol effect is -2.55 W m-2. Considering the linear sulphuric acid dependence of the nucleation mechanism used in ECHAM5-HAM and an increase in SO2 emissions from 0.06 Tg(S)/year (year 1750) to 54 Tg(S)/year (year 2000), the effect of nucleation on present-day forcing seems reasonable.

Kazil, J., Stier, P., Zhang, K., Quaas, J., Kinne, S., O'Donnell, D., Rast, S., Esch, M., Ferrachat, S., Lohmann, U.,

and Feichter, J.: Aerosol nucleation and its role for clouds and Earth's radiative forcing in the aerosol-climate model ECHAM5-HAM, Atmos. Chem. Phys., 10, 10733-10752, doi:10.5194/acp-10-10733-2010, 2010.

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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.)

Response

Changes with respect to Stier et al. 2005

1. new nucleation scheme (charged + non-charged)
Kazil, J., Stier, P., Zhang, K., Quaas, J., Kinne, S., O'Donnell, D., Rast, S., Esch, M., Ferrachat, S., Lohmann, U., and Feichter, J. (2010): Aerosol nucleation and its role for clouds and Earth's radiative forcing in the aerosol-climate model ECHAM5-HAM, Atmos. Chem. Phys., 10, 10733-10752, doi:10.5194/acp-10-10733-2010.

2. new Kappa-Kohler water-uptake scheme
O'Donnell, D., Tsigaridis, K., and Feichter, J.: Estimating the direct and indirect effects of secondary organic aerosols using ECHAM5-HAM, Atmos. Chem. Phys., 11, 8635-8659, doi:10.5194/acp-11-8635-2011, 2011.

3. prognostic CDNC calculation and changes for mix-phase cloud microphysics
Lohmann, U., Stier, P., Hoose, C., Ferrachat, S., Kloster, S., Roeckner, E., and Zhang, J.: Cloud microphysics and aerosol indirect effects in the global climate model ECHAM5-HAM, Atmos. Chem. Phys., 7, 3425-3446, doi:10.5194/acp-7-3425-2007, 2007.
Lohmann, U. and Hoose, C.: Sensitivity studies of different aerosol indirect effects in mixed-phase clouds, Atmos. Chem. Phys. Discuss., 9, 15045-15081, doi:10.5194/acpd-9-15045-2009, 2009.

4. new SOA scheme
O'Donnell, D., Tsigaridis, K., and Feichter, J.: Estimating the direct and indirect effects of secondary organic aerosols using ECHAM5-HAM, Atmos. Chem. Phys., 11, 8635-8659, doi:10.5194/acp-11-8635-2011, 2011.

5. AG activation scheme
Stier et al. (2011) in preparation

6. CCN diagnotics
Stier et al. (2011) in preparation

7. updated dust emission
The global aerosol-climate model ECHAM5-HAM, version 2 (ECHAM5-HAM2), K. Zhang, D. O'Donnell, J. Kazil, J. Feichter, P. Stier, S. Kinne, U.Lohmann, S. Ferrachat, J. Quass, S. Rast, M. Esch, and B. Croft

8. updated seasalt emission
The global aerosol-climate model ECHAM5-HAM, version 2 (ECHAM5-HAM2),

K. Zhang, D. O'Donnell, J. Kazil, J. Feichter, P. Stier, S. Kinne,
U.Lohmann, S. Ferrachat, J. Quass, S. Rast, M. Esch, and B. Croft

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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.

Response:

We agree with the referee and we have modified the text as follows:

Preliminary analysis from EUCAARI model analyses of the direct radiative forcing by the aerosols ranges from -0.049 and $-0.311 Wm^{-2}$. In Quaas et al. (2009) several additional global models were used to derive the direct ($-0.4 \pm 0.2 Wm^{-2}$) and indirect effect ($-0.7 \pm 0.5 Wm^{-2}$) radiative forcings for atmospheric aerosols. We caution that the EUCAARI results are only from the three models participating in the project, more complete and comparable results will be available later for IPCC AR5 analyses.

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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.

Response:

We modified the text on p. 18064 into the following form:

"A major outcome of the EUCAARI nucleation studies is the new semi-empirical nucleation rate parameterizations for neutral and ion-induced nucleation based on field observations (Riipinen et al., 2007a; Paasonen et al., 2010; Nieminen et al., 2010). These parameterizations, while compatible with the atmospheric measurements made during the EUCAARI project, probably need further refinements to

become more accurate at varying atmospheric conditions, especially at conditions encountered outside the continental boundary layer. Refining the existing parameterizations requires additional field and laboratory measurement data, such as those coming from the on-going CLOUD experiment (Kirkby et al., 2011).

Direct application of nucleation rate parameterizations is not possible, or at least not desirable, in large-scale modeling frameworks (see Kerminen et al., 2010). As a result, an additional aerosol formation rate parameterization is needed. In EUCAARI, the existing and widely-applied aerosol formation rate parameterization by Kerminen and Kulmala (2002) was further improved (Lehtinen et al., 2007; Anttila et al., 2010).“

Ref:

Kerminen, V.-M. and Kulmala, M.: Analytical formulae connecting the “real” and the “apparent” nucleation rate and the nuclei number concentration for atmospheric nucleation events, J. Aerosol Sci., 33, 609–622, 2002.

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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.

Response:

We agree with the reviewer's remark. However, Myriokefalitakis et al (2011) preformed the first global modeling study that explicitly parameterized and evaluated the importance of multiphase chemistry for organic aerosol formation, in particular through oxalic acid formation. The modeling results have been supported by extensive comparison with observations. That study clearly demonstrated that multiphase reactions in the global atmosphere can produce significant amounts of water soluble organic aerosol and precisely 5-10 Tg-C/yr of oxalate that is about 5-10% of the model's water soluble organic carbon.
The sentence is modified accordingly.

Myriokefalitakis, S., Tsigaridis, K., Mihalopoulos, N., Sciare, J., Nenes, A., Kawamura, K., Segers, A., and Kanakidou, M.: In-cloud oxalate formation in the global troposphere: a 3-D modeling study, Atmos. Chem. Phys., 11, 5761-5782, doi:10.5194/acp-11-5761-2011, 2011

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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.

Response:

We agree that there should be no claim that only EUCAARI has done work on this. We have added additional references on some of the key works in the field.

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.

Response: These have been corrected