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Interactive comment on "Ice nucleation by combustion ash particles at conditions relevant to mixed-phase clouds" *by* N. S. Umo et al.

Anonymous Referee #3

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Review Umo et al. (2014) Ice nucleation by combustion ash particles at conditions relevant to mixed-phase clouds

Summary of the presented work

This study investigated different combustion ashes regarding their potential atmospheric relevance as ice nuclei. To motivate the study, the authors emphasize the injection into the atmosphere. However, ice residual measurements do not distinguish ashes from minerals yet, so that the atmospheric presence remains unclear. Additionally, the ice nucleating behavior of combustion ashes was also not investigated until now.

The study distinguished two different combustion ash types with respect to their

sources. The first class is bottom ash coming from complete combustion processes of coal in households and power plants. The second class is fly ash, which is emitted during combustion processes like wild fires, biomass burning or domestic combustion and may include other materials.

As an example for the bottom ash particles the authors used first wood and coal solid fuel combustion ashes and second combustion ash from wood combustion in a typical household. For the fly ash class the authors used a filter sample from a power plant. The samples were characterized regarding their surface and morphology, their size distribution and their mineralogical composition and finally their ice nucleating ability. To this end, they used the μL- and nL-NIPI experimental setup.

The coal fly ash (CFA) sample shows compared to the bottom ashes a smaller BET specific surface area. Regarding the size distribution show the bottom ash samples a much narrower distribution with a smaller mean diameter than the CFA. Also in the mineralogy the CFA clearly separates from the bottom ashes. The results for the ice nucleating properties show that the CFA has higher ns-densities than the other samples. The reason suggested in this study for that could be the difference in the combustion process or the morphology. The parameterization of the ns-density for CFA has a temperature dependency similar to biological particles. In comparison to other aerosols the combustion ashes have an ns-density comparable to clays and minerals.

The authors call for future studies to improve the separation of ash particles from the mineral class in ice residual measurements. Furthermore the differences between the ash samples have to be investigated in more detail.

General comments

This study is technically impeccable and original. Most parts of the article are well written. The main problem which I have with this work is its relevance for the atmosphere. Although the authors go to great lengths to justify the atmospheric importance of these particles and cite a lack of suitable analysis methods as the reason for why they haven't been in focus until now, it could also simply be that they occur only very locally and in small number concentrations. The size of the particles investigated here (after artificial disaggregation!) is simply to large to keep them aloft in the atmosphere for significant times. Nevertheless, I support the publication of this paper subject to a number of corrections and clarifications and hope that future research in the field will elucidate the questions raised in this study.

Detailed comments

Section 1:

- Is any information available on the size distribution of ambient ash samples?

- What number or mass concentrations of ash are found in ambient air? If this information is not available for ash specifically, what are the total aerosol concentrations close to ash sources?

- It seems likely that the composition is of what is termed “ash” is size-dependent, and that at the smallest sizes there is a transition to soot. Please comment.

- It could also be that the ice nucleation ability is size-dependent. Was any indication for this observed in the experiments?

- page 28848 line 20ff: The study by Block and Doms (1976) which is cited to underline the atmospheric relevance of fly ashes is quite old. During the last 40 years the inefficiencies within the collection systems in power plants have certainly improved.

- page 28849 line 13/14: when citing the McCluskey et al (2014) paper, please add the conditions (T and RH) under which ice nucleation was measured.

Section 2: Sources and generation of combustion ashes:

- It did not become quite clear to the reviewer what “solid fuel” is, as the term is ambiguous (at least to non-native speakers). A better description of the material and where it was bought would improve the understanding of this section.

- Furthermore the question arises whether the domestic ash was produced in a typical stove in a kitchen or in a stove in a living room. When it was produced in a kitchen stove the authors have to justify whether this kind of cooking is really relevant these days (or in which parts of the world it is relevant).

Section 3: Preparation of ash suspension and freezing experiments:

- The samples were preprocessed before the freezing experiments were done. The question is whether the samples are representative of atmospheric particles after the processing. This applies in particular to the stirring which breaks down the aggregates.

- page 28853 line 12ff: For a better comparison between fractal agglomerates the indication of the fractal dimension or the size of the primary particles as for soot agglomerates would be better. Additionally the used software Image J should give this information.

Section 4.2: Size distribution of combustion ash particles:

- Why show all 4 samples 2 modes in the size distribution?

- From these size distributions you can get the geometric surface area. Therewith a direct quantitative comparison of the BET surface area and the geometric surface area is possible and also of interest for further studies.

- Volume mean diameters of 8-10 μm are very large compared to typical atmospheric aerosol particles sizes. This should be mentioned.

Section 5.2:

- page 28858 line 21 to page 28859 line 3: This section is difficult to understand. A more detailed description is necessary. Please comment on how large the difference is to the previous method and whether the published results change when the new calculation is applied.

- The use of different fit functions for the different samples is not well justified. In particular, the domestic bottom ash appears to have a very similar temperature dependence as the other botton ashes, but the fit function includes a third free parameter. Of course this improves the quality of the fit, but it does not add any information or does not aid the physical interpretation. How much worse would the fit be with just two parameters?

Section 5.3: Comparison of ice nucleation activities of combustion ashes to INPs with varied mineralogies:

- It should be mentioned that the study from Niemand et al. (2012) used geometric surface areas to determine the ns-density whereas the other studies used BET specific surface areas. Therefore, a direct comparison is not possible. Furthermore you have to mention which parameterization (BET or geometric ns) from Hiranuma et al., 2014 you used.

- A comparison with volcanic ash immersion freezing experiments e.g. by Welti et al, 2011 and Steinke et al., 2011 would also contribute to the study.

Section 7:

- There is some work on pyroconvective clouds (e.g. Sassen and Khvorostyanov, 2008, doi:10.1088/1748-9326/3/2/025006) which could be mentioned here.

Technical comments:

- page 28863 line 19 “in the this category” - either the “the “or the “this” is too much

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 28845, 2014.