

Interactive comment on “Coal fly ash: Linking immersion freezing behavior and physico-chemical particle properties” by Sarah Grawe et al.

Anonymous Referee #2

Received and published: 26 July 2018

This is a well-written paper concerning immersion freezing of water droplets triggered by coal fly ashes (CFA). This is a timely topic which fits very well into ACP. The authors compare samples from different sources concerning their ice nucleation activity. They use different set-ups (LACIS, WISDOM, SPIN, LINA) to do so. They correlate their findings with the physical-chemical properties of the particles. They conclude that CaSO₄ and CaO are the crucial mineral components and that thus surface hydration of these fractions can have an important impact on the ice nucleation activity of CFA.

Particularly, the physical-chemical characterization of the ice nucleation particles (INPs) makes the importance of this paper. Therefore, the precise application of the

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different methods in use is crucial. Therefore, I have listed here my concerns regarding the different techniques:

1. Alabama: I am not an expert in aerosol mass spectrometry. Therefore, I have no comments.
2. ESEM. The special resolution of the microscopic pictures is very low. I highly recommend transmission electron microscopy (TEM) pictures. This is of particular importance when investigating the spherical shaped combustion products, which are thought to origin from organics. TEM could provide the internal structure of the particles and will allow a correlation between structure and chemistry, which both have an impact on the ice nucleation (see e.g. Häusler et al. who have investigated typical constituents of soot and coal, i.e. graphenes, which have similar n_s values like some coal fly ashes).
3. EDX. Is a valuable technique in order to gather the chemical composition of materials and is easily accessible in combination with SEM. However, at concentrations below 0.1% the signal to noise ratio of this technique is unsatisfying and the results are untrustworthy. I highly recommend using micro X-ray fluorescence analyses (μ -XFA).
4. XRD. The powder diffractograms shown in the supplement are of excellent quality. Therefore, the authors can easily apply a Rietveld refinement of their data.
5. Bulk chemical composition analysis. Please specify how this was done.
6. DMPS. No comments.
7. Light microscopy. Eventually, polarization microscopy could help to differentiate the components of the particles (amorphous vs crystalline).
8. BET. Specific surface areas about 1 m² g⁻¹ are often not precisely accessible. Please, describe the detection limit of your instrument.

The authors should discuss in more detail the impact of internal structure, morphology and chemistry of the INPs on the ice nucleation activity. In particular, I miss a discussion

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of the carbonaceous particles. This can easily be performed with the data at hand and with some modifications described above. Therefore, I rate this manuscript as "accepted, subject to minor revisions".

Reference

Häusler, H., Gebhardt, P., Iglesias, D., Rameshan, C., Marchesan, S., Eder, D., Grothe, H.: Ice Nucleation Activity of Graphene and Graphene Oxides, *The Journal of Physical Chemistry C* 122 (15), pp. 8182-8190, 2018. DOI: 10.1021/acs.jpcc.7b10675

Interactive comment on *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2018-583>, 2018.

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