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Interactive comment on "On-line single particle analysis of ice particle residuals from mountain-top mixed-phase clouds using laboratory derived particle type assignment" *by* S. Schmidt et al.

Anonymous Referee #2

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The manuscript by Schmidt et al. presents ambient and laboratory single particle mass spectrometer data to identify chemical components in aerosols and ice residuals on Jungfraujoch and to draw conclusions on the ice nucleation ability of different aerosol particle components. Given the importance of ice nucleation processes for precipitation and the climate, and the many uncertainties related to these processes, such studies are needed. The combination of laboratory experiments and ambient measurements makes this study especially useful for the single particle mass spectrometer community. I therefore recommend publication of this well-written manuscript in ACP after the following comments have been addressed:

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General comments

The limitations and uncertainties of single particle mass spectrometry and the respective data should be addressed in more detail. There are several aspects:

- More care should be given as how one describes the quantities. It should be made clear, e. g. in the introduction, that “a larger relative amount” cannot be interpreted as “more aerosol mass”, but can only refer to “a larger number of particles” assigned to a certain category.

- The presentation of marker mass fragments and reference spectra from different particle types is highly desirable for the single particle mass spectrometer community. For the spectra to be useful for other groups/instrument types however, information on uncertainties needs to be given. One of the issues of laser ablation single particle mass spectrometers is the (weak) repeatability of spectra/measurements; depending on particle size, chemical composition/morphology, and on the placement of the particle in the laser beam, the amount of ablated/ionized material can vary significantly. According to p. 4, l. 36, you were not size-selecting particles for your laboratory tests, which might have led to larger spectrum-to-spectrum differences. Showing e. g. standard deviations of the averaged spectra would give an idea on the uncertainty and variability of the marker spectra.

- Related to the above – how were the marker peaks identified? Given the overlap of marker peaks between different particle types, it seems to me that presenting the data as marker spectra would almost make more sense. How important are the individual markers for identification as opposed to the whole spectrum? In Table 3 you give the ratio of the number of spectra containing marker peaks (all marker peaks? just some?) to all spectra of a particular particle type. A better way to present uncertainties would be the standard deviation of the avergaed spectra (see above). The ratios in Table 3 are fairly low, I thus expect a relatively high scatter of spectra per particle type.

- More information should be given on the choice of clustering algorithm and the un-

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certainties of its outcome. There are a lot of references given to Roth (2014, 2016), but the main points should be conveyed to the reader in the manuscript (also in Table 1).

A general result I infer from the ambient measurements is that secondary (here in the sense of formed from a chemical reaction/in combustion) components (both organic and inorganic) or particles containing a lot of secondary (organic) components (e. g. biomass burning) are less effective ice nuclei than primary organic particles such as e. g. biological particles. In this simple categorization, however, it is hard to place the engine exhaust particles. It would be helpful if the engine exhaust and PAH particle types were discussed in the manuscript (which is not the case now).

I am confused about the similarity of sea salt and cooking emissions fragmentation pattern. How were the sea salt particles produced, what kind of cooking emissions were investigated? The variety in cooking activities is incredibly large (food cooked? what method? fuel used? etc.), and without further information a “cooking spectrum” is not very meaningful.

Soil dust and minerals are categorized as “natural” aerosol, in accordance with classifications in literature. However, soil dust aerosol concentrations can directly be influenced by anthropogenic activities (e. g. farming, mining, forestry).These are all important factors, especially when looking into/modelling anthropogenic influences on climate via aerosol-cloud interactions. I am fully aware that soil dust source apportionment lies outside the scope of this paper, but I suggest leaving out the word “natural” and potentially add a sentence on this issue.

Specific comments

P. 3 - 4, l. 37 – 2: Please give the reason for choosing the fuzzy c-means algorithm over the other two possibilities.

P. 5, l. . 3 – 9: See general comment on uncertainties and limitations of marker/spectra identification.

P. 5, l. 25 – 28: Was the inlet heated to 20°C to prevent condensation? Did you perform any assessment of the influence of the heating on the chemical composition (e. g. evaporation of semi-volatile material)? What was the residence time in the inlet? The evaporation of semi-volatile material could be especially important for SIA and SOA. Were parts of the ICE-CVI heated as well? Please add more information on sampling/inlet conditions.

P. 5, l.34: What about the transmission of particles with diameters larger than 500 nm? A relatively large fraction of the particles measured at Jungfraujoch was larger than 500 nm, according to Figure 3.

P. 7, l. 7 – 8: Many of the marker mass fragments of the biological particles (especially bacteria and pollen) have negative marker mass fragments. How were they identified in ambient air where you only had positive spectra?

P. 9, l. 31 – 38: See general comment above. A more thorough discussion on the properties and uncertainties of engine exhaust and PAH spectra in comparison with biomass burning spectra might shed some light on their differences in ice nucleation behavior.

P.10, l. 34 – p. 11, l. 3: Please elaborate further on the comparison and discrepancy of OPC and ALABAMA size distributions. The size distributions shown in Figure 3 are completely different and basically do not allow to draw any conclusions. Are there no artefacts of the Sky-OPC?

P.11, l. 22 – 34: Whereas the meteorological conditions were similar for the two periods, they do no coincide in termns of time of day, which however can have large influences on (anthropogenic) emission patterns (e. g. engine exhaust, cooking…). Please take this into account in your data interpretations.

P. 12, l. 27: This “conclusion” is a rather minor finding of your study (or put in different words, a potential reason for differences in composition between activated and non-

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activated aerosol particles that can be ruled out). This sentence should not be at the end of the results section.

P. 13, l. 1 – 28: Again, in your summary, please a add a few sentences on uncertainties and the limitation of the method concerning single particle marker spectra identification and particle type detection/identification in ambient air.

Technical comments

P. 1, l. 19 – 21: “As outcome. . .” – weird sentence structure, rephrase

P. 1, l. 32: “and” instead of “an”

P. 4, l. 37: Phrase structure; should read “Coating experiments were also . . .”

P.7, l. 4 – 5: Weird sentence structure

P. 9, l. 5: Dot at the end of the sentence is missing.

P. 12, l. 15: Sentence structure: Should read "It has to be noted further..."

P. 12, l. 27: conclusion

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