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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Chemical composition of ambient aerosol, ice residues and cloud droplet residues in mixed-phase clouds: single particle analysis during the Cloud and Aerosol Characterization Experiment (CLACE 6)" *by* M. Kamphus et al.

Anonymous Referee #5

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The manuscript by Kamphus et al provides a valuable dataset detailing the composition of ice nuclei and CCN within a mixed phase clouds sampled at a mountaintop site. However, the analysis of the data provides only general conclusions about nucleation processes that are not well established. The authors should address the major points listed below as well as comment on minor points prior to publication.

Major points:

1. The principal limitation of the study is that of comparing ice (IR) and droplet residue (DR) data to background (BG) data for a variety of meteorological conditions. This approach is understandable because the sampling method inherently gives low particle rates and poor statistics for individual cloud events. However, the useful information gained by using this approach is very limited. One example in the text (15397, line 1) describes a lack of spectra classes 5 & 6 in DR compared to mission-averaged BG abundance rather than BG abundance for that event. Considering the variability of aerosol properties within different air masses, what conclusion can be drawn from this comparison? Other examples are 15402 line 8 and 15404 line 11, where DR data could be compared to biomass aerosol in the BG. In these DR cases and wherever feasible, the authors should compare DR and IR to the BG data for that specific event, which unlike residue data, should be plentiful.

Additionally, two possible general resolutions are 1) divide the analysis into 2-3 broad scenarios based on wind direction, cloud type, or an appropriate tracer such as CO, NOx, or black carbon; or 2) focus on the IR intensive March 1-2 cloud event as a case study. In some way, demonstrate that chemical and meteorological conditions are similar for any DR & IR comparisons with BG.

2. The definition of aerosol types based on classes from a clustering algorithm does not appear to efficiently separate spectra based on chemical composition or IN capability. Throughout the manuscript, some spectra cluster classes are labeled 'mineral dust', eg., SPLAT classes 1 & 2. However, all of the SPLAT classes in Fig 3 except class 3 have metallic components (other than K) characteristic of crustal material – Fe, Ca, Si. The authors then present fractional abundance of these classes in Fig 4, where it appears that the vast majority of BG aerosols contain crustal material. Is this expected? Or instead is this a product of a small fraction of spectra in each class that contained large metallic signatures that when averaged, produce minor peaks in the cluster centers? Furthermore, SPLAT classes with low/no metallic signatures (3 & 6) are the only classes that are underrepresented in IR, which also demonstrates the

lack of selectivity of the clustering routine. Please elaborate on the homogeneity of populations within the clusters. Explain the ramifications on the analysis (for example, the small fraction of mineral dust aerosols present in multiple clusters), and explain the compositional labels for classes.

Similarly, the separation of cluster classes is necessarily subjective. The authors should consider refining some classes based on spectra features. As an example, it appears that SPLAT classes 2 & 4 are essentially the same, ie, processed mineral dust, that differ only slightly in relative organic signal.

Minor points:

Fig 3. The intensity of the negative ion peak at 55-56 m/z tracks the HSO4– intensity. It is probably a secondary electron peak created by a process (e.g., impaction) within the time-of-flight region. Could a similar process occurring during mass analysis also explain the high intensity signals at negative <10 m/z?

15383 line 25. List typical residence times and temperatures for the sampling inlets.

15385 lines 3-10. Specify that cut-off diameters are lower cut-off diameters.

15385 line 14. List temperature of this section.

15385 line 23. By sampling only the smallest ice crystals that have presumably nucleated ice most recently, is it likely that the Ice-CVI sampling will miss the most efficient ice nuclei (those that formed initially and have grown beyond the Ice-CVI size range)? Please comment here.

15386 line 17. 'IN concentrations' measured by the OPC?

15387 line 4. Does the 300-400 nm maximum represent the overall 'detection' efficiency for the entire instrument (inlet transmission + light scattering detection + ionization hit rate) or just the transmission efficiency of the aerodynamic lens?

15393 line 11. Fig 2 shows a strong size dependence for CCN, particularly in the

ATOFMS data, where only very large CCN (hundreds of nm) appear to be activating preferentially. Please comment on whether the large size of DR is an effect of the sampling method or is representative of a cloud microphysical processes, e.g., very small supersaturation of water.

15397, l 25: DR are relatively enhanced in sulfate-containing spectra, or equivalently, spectra with metallic components are depleted in DR compared to BG (for this event?). Since largely, CCN activity follows aerosol size rather than composition, it would be interesting to mention whether the CCN-inactive mineral dust aerosols presumably in the background were larger than the sulfate-rich CCN-active aerosols. It also appears that mineral dust aerosols were coated with secondary material. This does not appear to enhance their CCN properties. Please provide a few comments.

15399 line 26. The authors state that the dominant BG class (ATOFMS class 5) is consistent with biomass burning aerosol. Is this 79% level typical of other tropospheric ATOFMS studies? Does this indicate a high biomass burning influence throughout the campaign? Do all of the class 5 particles contain potassium?

15404 line 2. State whether any of the SPLAT IR class 4 spectra show indications of insoluble material.

15406 line 9. The authors should consider shortening the lengthy comparison to airborne IR data (although highlighting enhancement of metallic and dust components across various IR data is important), and instead compare to previous mountaintop measurements during the INSPECT campaign referenced in the text. State how the overall conclusions of IR composition and nucleation mechanisms for the CLACE study compare with those of INSPECT.

Technical corrections:

Fig 4a. Reds and greens are too similar. Change colors for clarity.

15404 line 17. replace are with is.

15380 line 23, 15396 line 28, 15408 line 22: replace extend with extent.

15395 line 26. Replace Different with Compared.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 15375, 2009.