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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 #1

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General comments

This paper describes measurements from CLACE 6 from two single particle mass spectrometers at the Swiss High Alpine Research Station Jungfraujoch. Chemical composition data are compared for background aerosol, droplet residues and ice residues. Results are consistent with previous work, suggesting an enhancement of mineral dusts in ice residues and an enhancement of sulfates in the droplet residues.

The data are a useful addition to the existing literature, and the manuscript should be published, but I believe that some additional analysis would make the dataset most useful to the reader.

Specific comments

p. 4, line 9. Given the appropriate ice nucleus, ice nucleation can occur in theory at any temperature below 0 °C, and ice nucleation has been observed very near this limit for some biological particles (see summary of [Szyrmer and Zawadzki, 1997]).

p. 17, lines 1-3. Was the ATOFMS connected to the ice-CVI out of cloud for a long enough time to 'verify zero particle counts' with any confidence? From Figure 1, it appears that the ATOFMS was on the Ice-CVI inlet for ~6 days total. During this time, the ATOFMS characterized 152 ice particles, and, if similar to the SPLAT instrument, many of these were detected over an ~8 hour period on March 1-2. This corresponds to only about 1 particle per hour detected. One could calculate how many hours of sampling would be needed to verify 'zero particle counts' with 95% confidence, e.g. assuming a Poisson process. My initial guess is that the ATOFMS was not connected to the ice-CVI out of cloud for a long enough time period to verify zero particle counts with any real confidence.

p. 17, lines 1-3 and p. 17, lines 19-22. Along these same lines, were any particles detected from the ice-CVI inlet when sampling out of cloud, either by the ATOFMS or the OPC? Can the authors provide an estimate of the fraction of unactivated aerosol particles that are carried in the minor flow of the Ice-CVI? Even if the Ice-CVI is 99.9% effective at removing aerosol particles, 10% of the 'ice' signal would come from aerosol that has passed through the minor flow of the Ice-CVI. This might help explain some of the unexpected results described later in the paper (e.g. ice residues of 25% sulfate from the SPLAT measurement).

p. 21, lines 16-20 and Figure 1. From figure 1, there is no clear change in the trend of the OPC signal when switching from the Ice-CVI to the total inlet on March 2, whereas

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most times the total inlet measurements are significantly greater than the Ice-CVI. This, combined with the relatively high ice concentrations (0.55 cm-3) compared to the rest of the project, the statement that there were 'strong westerly flow conditions' for that time period, and the possibility for 'mechanical fracturing of falling or windblown ice crystals, especially during snow fall' [Mertes et al., 2007], suggest that the site was likely influenced by blowing snow at this time. How strong were local winds during this period? Given that nearly half of all of the ice residue data come from this period, it would be worthwhile to discuss differences in ice residue chemical composition from this period with the remainder of the project.

p. 21, line 27- p. 22, line 1. The fact that class 5, representative of biomass burning particles, is enhanced in the ice residues is consistent with recent remote sensing [Lin et al., 2006; Sassen and Khvorostyanov, 2008] and laboratory studies [Petters et al., 2009] which suggest that smoke particles can impact ice nucleation in the atmosphere. In fact, many of the components listed in class 5, including potassium, nitrite, chloride and sodium, were shown to be correlated with the production of ice nuclei from fires in the Petters et al. study.

p. 22, line 28 – p. 23, line 1. The fact that classes 5 and 6 are not observed in the droplet residues may simply reflect the fact that the airmass that was not affected by biomass burning. The authors might compare the back trajectories to fire observations from MODIS during this time period (http://rapidfire.sci.gsfc.nasa.gov/).

Section 3.2.4. Although I understand the reasoning behind averaging the results for the ice residues to the background aerosol averaged over the entire project, the droplet residues were only collected during one day, and it would be useful to know how the DR compare to the background aerosol on this day (there should be sufficient particle counts for each category). As noted in the previous comment, it may simply be that the airmass did not contain any biomass burning particles. On the other hand, if there is a major contribution from class 5 in the background aerosol, but it is absent from the droplet residues, this would say something about the ability of smoke particles to act

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as CCN. In any case, it is a more useful comparison.

p. 23, lines 13-19 and Figure 4. The relative increases for coated versus uncoated mineral dust particles in the IR data compared to the BG data are surprising. Namely, class 1, thought to represent unprocessed mineral dusts, increase by a factor of <3 in the ice residues, whereas class 2, the processed mineral dust particles, increase by a factor of >6. This is somewhat unexpected, given that laboratory studies (e.g. [Eastwood et al., 2009]) suggest that unprocessed mineral dusts are more IN active than processed particles.

p. 23, line 28-p. 24, line 3. From the data presented, the results do not support the claim of the 'importance of sulfate particles acting as CCN at the JFJ.' First, as noted in the previous comment, the DR data need to be compared to the BG data from this day only. Second, significant cloud processing could be occurring in the droplets, adding sulfate mass to the particles after the particles have activated as droplets and causing a shift in their classification. In fact, the large size of the droplet residues (Fig 2) supports the notion that some cloud processing has occurred.

p. 27, lines7-9 and Figure 4. Although the authors note later in the text that the SPLAT and ATOFMS categories cannot be compared directly, it is surprising the dramatic differences in the biomass burning categories for BG (11% vs 79%) and IR (enhancement of biomass burning particles in the SPLAT instrument and depletion in the ATOFMS) compositions from Figure 4 for SPLAT and the ATOFMS. How do the data compare when considering identical sampling periods?

p. 28, lines 7-10. Again, the authors should compare back-trajectories to MODIS fire counts, rather than making the very vague statement that it's possible that there may have been differences in biomass burning contributions. Is there a CO measurement at the site?

p. 31, lines 2-4. Just looking at Figure 4, I come to very different conclusions about the chemical composition of the BG and IR based on the ATOFMS dataset compared

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to the SPLAT dataset. The authors clearly recognize the difficulty in interpreting the results from the two measurements, so much so that they deem it necessary to tell me (p. 29, lines 21-22) that the two datasets 'do not represent a contradiction.' With that said, although it was not the intent of the authors to do a comprehensive comparison of the 2 mass specs, it seems like a relatively straightforward exercise for Section 3.4 to compare data from the two instruments for the same sampling periods. Such a comparison would be much more useful than simply explaining why the two datasets (which should yield similar data) cannot be directly compared (do we need multiple single particle mass specs at every field campaign?). If after comparing measurements from identical time periods, the two instruments still yield different results, this would at least help the reader better appreciate the biases that each instrument has, so that (s)he can better judge data from these instruments in future field campaigns. I would go so far as to say that it would be most useful (although I'm not sure if it's possible) to analyze the data using the same classification schemes for the two instruments.

p. 33, lines 13-16. It seems that the signal for Na and Cl from CLACE 6 was likely from a different source than from the CRYSTAL-FACE study, given that class 5 for the current study is thought to be representative of biomass burning aerosol. So I don't really think these two studies are consistent in this regard.

Figure 4. I am unable to distinguish colors between class 5 and class 6 particles. Further, classes with <1% are not represented in the figure, although they are clearly important for the IR (e.g. classes 1, 8 and 10 in the ATOFMS background aerosol category). Figure 4 might be better represented as a Table.

Technical corrections

p. 20, line 2. Remove the word 'both.' As written, it implies that class 1 and class 2 each account for 17% of the particles.

p. 20, line 25. 'ratio'

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p. 22, line 25. ‘completely’

p. 22, line 28 and p. 34, line22. ‘extent’

p. 24, line 27. Figure numbering is not in order (i.e. fig 6 is referenced here, and fig 5 has not yet been called out).

p. 27, line 15. ‘these data’

References

Eastwood, M. L., et al. (2009), Effects of sulfuric acid and ammonium sulfate coatings on the ice nucleation properties of kaolinite particles, Geophys. Res. Lett., 36, 5.

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