Review

Aerosol properties, source identification, and cloud processing in orographic clouds measured by single particle mass spectrometry on a Central European mountain site during HCCT-2010

Roth et al.,

This study presents the measurements from a field campaign where several on-line instruments were operating side by side. The focus of this paper is to characterize the chemical composition of aerosol particles measured by a single particle laser ablation instrument and determine how different aerosol species are activated into cloud droplets. Cloud droplet residues were analyzed using a counter flow virtual impactor (CVI). Aerosol chemical composition and size distribution are provided by a single particle laser ablation instrument (ALABAMA). This instrument is capable of generating both positive and negative spectra from individual particles and therefore has the unique feature of providing information of aerosol mixing state. Single particle spectra measured by the ALABAMA were classified using fuzzy clustering. This paper is well written and organized and I recommend it for publication. However, some additional information or discussion should be included.

- Was any attempt made to quantify the measurements from the ALABAMA using co-located instrumentation? How did the size distribution measured by ALABAMA compare with that of the OPC?
- CVI inlets can often lead to enrichment of aerosol particles. Has this CVI been validated to through comparisons with a whole air inlet (clouds particles +interstitial particles)/interstitial inlet set up? This information is not included in Mertes et al., 2005. More information on the flows and counter flows (and enrichment factors) of the CVI could be included in the discussion.
- Section 2.2: Can the authors provide more information on the types of clouds that were studied, and how the out of cloud periods were chosen? How are these out of cloud periods thought to be representative of aerosol particles activated into clouds, e.g include information on air mass trajectories and on wind direction?
- The authors mention that after cloud processing, aerosol particles contain higher amounts of nitrate and sulphate. They mention that the increases in nitrate and sulphate particles will increase particle hygroscopicity and their ability to act as CCN. It is true that higher fractions of inorganic ions will increase the hygroscopicity of the aerosol particle, however given the size of the particles studied (> 200 nm) it is likely that they will be good CCN independent of their aerosol composition.
- For the mineral particles (Section 3.2.4) why do the authors think that there is such a difference in activated aerosol composition? All particles of the sampled size should, according to theory, be activated into clouds. Chemistry of aerosol particles is not thought to play a role in the activation efficiency of aerosol particles with diameters > 200 nm (Duseck et al., 2006)

Minor comments:

Page 24420, Line 14 suggestion: “having a diurnal variation”

Page 24421, Line 1: On one hand, the presence…

Line 28: What is meant by individual cloud?

Page 24422, Line 20: The second part of this sentence needs to be rephrased.

Section 2: Experiments and data evaluation: Although mentioned later on in the manuscript, it would be useful to have information where the site is located with respect to the nearest city, Suhl in the methods section.

Page 24423, Line 24: I assume that the interstitial aerosol was not detected because of their small size. Can the authors include this information? Were any size distribution measurements made between the interstitial inlet and the CVI to calculate aerosol activation profiles?

Page 24424, Line 20: The authors mention that the HR-ToF-AMS and the MAAP were operating continuously alongside the ALABAMA. Were the fractions of soot measured by the ALABAMA comparable with the fractions of BC measured by the combined HR-ToF-AMS+MAAP.

Page 24425, Line 11: It would be useful to provide a summary of the criteria described by Tilgner.

Line 16: What is the number of spectra required for statistical evaluations?

Page 24426, Line 20: The abstract states that more than 170,000 bipolar mass spectra were obtained while sampling out of cloud aerosol and more than 14,000 bipolar mass spectra were measured from cloud residues.

Page 24427, Line 3: Define MPIC and TROPOS.

Page 24429, line 22: remove "actual"

Page 24430, Line 2: Reformulate sentence "Due to the fact that a reduced data set….."

Page 24430, Line 27: "more from fuel combustion than.."

Figure 3: Relative high fractions of soot particle were observed by the ALABAMA. How do these compare to the fractions of soot observed by the MAAP + HR-ToF-AMS instrument? Was there any attempt to compare absolute numbers/volume measured by the MAAP/HR-ToF-AMS with that of ALABAMA.

Page 24432, Line 9: Laser ablation techniques have a tendency to be sensitive to matrix effects that favor certain species with low ionization efficiencies, e.g. K. Do the authors consider that matrix effects influence the measured composition of the aerosol particles?

Page 24432, Line 14: Aerosol particles with diameters between 200 nm and 450 nm are relatively large. Freshly emitted (anthropogenic particles) or freshly formed aerosol particles are principally measured at diameters < 100 nm.

Page 24432, Line 15: "had no time to grow by condensation…"

Page 24432, Line 26: east north-east direction

Page 24434: The authors state that their measurements are different to those observed in other studies with other instruments (Aerodyne, AMS). The reason for this is explained as being a result of the short averaging time of the Aerodyne instrument compared with a single particle instrument.

Is there so much variation observed over the period of the cloud that would cause different averaging results? Are the size distributions measured by the two instruments in the two studies comparable? It would be better for the authors to focus on the comparison with Schneider et al., from the same study than from observations by Drewnick et al. (2005). In general how does the fraction of organic material observed by ALABAMA compare with that observed by the C-ToF-AMS instrument. How does the size

distribution of aerosol particles measured by the ALABAMA compare with that measured by the C-ToF-AMS instrument?

How much was the scavenging efficiency of organic species dependent on their mixing state?

Page 24434, Line 10: "data"…… "single particle analysis".

Page 24435, Line 4: "…mass spectra show peaks"

Page 24435, Line 9: the definition of TMA should be made earlier.

Figure 8: The AMS biomass burning axis could be change to be in the same range as the EBC axis.