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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

Interactive comment on "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" *by* A. Roth et al.

Anonymous Referee #1

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Roth and co-authors describe a set of single particle mass spectrometer measurements of ambient aerosol performed at a mountain site in Germany 2010. Ambient "out of cloud" particles and cloud residuals were alternately analysed by bypassing or flowing air through a counterflow virtual impactor, respectively. All data were combined and classified using a series of approaches. Mass spectra were first clustered using c-means "fuzzy clustering" and the resulting clusters were merged with similar types using distance metrics to create a more manangeable set of final particle classes. Differences between out of cloud particles and cloud residuals were then assessed.

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Interactive Discussion
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Interstitial aerosol was not investigated. The main findings are that cloud processed particles at the site are enriched in sulfate, nitrate and aminium ions relative to out of cloud particles. Larger, possibly aged, soot particles were also found to represent a higher fraction of the in-cloud than out of cloud population, because of internal mixing with hygroscopic inorganics. The manuscript is well written and scientifically sound, with some nice tweaks on existing single particle mass spectral classification. I have some minor comments only.

The article would benefit from a map of the site and surrounding region/topography to complement the local wind data. The proximity to local carbonaceous aerosol emission sources could be included here. Also on this point, Figure 4 would be better represented in 2 panels. Readers will be more used to seeing a windrose like this applied to wind speed and direction frequency. It is easy to miss that the radial axis is actually the fraction of the mass spectral population detected. A standard windrose showing windspeed and direction would be more useful. A second panel could show the dependence of particle hits on wind direction. This could be normalised by frequency of wind from each sector if suitable.

An expanded discussion of the merits of fuzzy clustering would be helpful. My understanding is that the advantage is that each spectrum can "belong" to several clusters to differing extents. However the classification approach here is exclusive, in that all spectra end up "belonging" to only one cluster or group. In this case, what is the advantage over traditional "hard" clustering techniques like neural network algorithms or k-means?

In section 2.4.2 it is not clear how the authors determined a false positive or false negative assignment of a spectrum to an "incorrect" cluster. How is the incorrect assignment identified? Hasn't the particle already been objectively assigned mathematically to the most "correct" cluster using Pearson's r? After the positive and negative mass spectra were normalised separately, they were combined and normalised again. Why are they normalised again? Isn't this redundant? Or is it simply to express everything as a

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fraction of 1 rather than 2?

In section 2.4.3, last line, what are the counting statistics that the authors refer to?

Section 3.2.4 contains only two sentences, but to me this is one of the most interesting findings in the article. The possibility of catalytic oxidation of SO2 (and other species like DMS) in by iron and vanadium has impacts for the atmospheric lifetime and climate impacts of thee particles. This is briefly referred towards the end of section 3.4 but the authors have good evidence here and should expand the discussion to consider their results in the context of other single particle studies that have focused on this topic, eg (Gaston et al., 2010; Ault et al., 2010).

Section 3.2.3 The difference in hygroscopic behaviour for smaller soot particles with little inorganic content and larger soot particles with higher inorganic content has been predicted in Western Europe using single particle mass spectrometry previously and found to agree well with HTDMA measurements (Healy et al., 2014).

Page 24420, line 2: replace “have been” with “were”

24421, line 20: replace “during” with “within”

24422, lines19-21: Rephrase, unclear

24423, line 16: replace “in southwesterly direction” with “facing southwest”

References

Ault, A. P., et al.: Characterization of the Single Particle Mixing State of Individual Ship Plume Events Measured at the Port of Los Angeles, Environmental Science & Technology, 44, 1954-1961, 10.1021/es902985h, 2010. Gaston, C. J., et al.: Real-Time Detection and Mixing State of Methanesulfonate in Single Particles at an Inland Urban Location during a Phytoplankton Bloom, Environmental Science & Technology, 44, 1566-1572, 10.1021/es902069d, 2010. Healy, R. M., et al.: Predicting hygroscopic growth using single particle chemical composition estimates, Journal of Geophysical

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