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Interactive comment on “Atmospheric organic matter in clouds: exact masses and molecular formula identification using ultrahigh resolution FT-ICR mass spectrometry” by Y. Zhao et al.

Anonymous Referee #1

Received and published: 24 September 2013

This manuscript describes FT-ICR-MS results from 2 cloud events at Storm Peak Laboratory in Colorado. The manuscript presents novel data as no application of FT-ICR-MS has been performed on wintertime supercooled droplets. Any insight on organic matter in these conditions is novel and a worthwhile contribution to the literature. The study also shows interesting new results on organic nitrogen and organosulfur compounds. Therefor the manuscript is suitable for ACP and I would support publication of the results if attention has been given to the issues below.

A main problem of this method as well as most organic matter characterizations is that the results reflect only a part of the organic matter. In the case here, peaks will

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only be obtained for species that have been extracted, are being ionized and present a negative ion species. The authors in different places acknowledge parts of this. Still the reader should be more reminded of this in the discussions and conclusions. While it is fine to not have any organic carbon recovery data, some results need to be put in context as the losses are potentially substantial. First at no point the pH of the samples is given or if available this would be crucial as it would give insights on the potential ionization of species and hence their retention (or not) in the reverse phase extraction step. Second the authors say that they could lose low (<100Da) MW species, or the most abundant organic species in clouds are typically small carboxylic acids and small carbonyls which could be missed. However these species all have very high O/C ratios and so it might be good indicating how this then impacts the comparison of O/C ratios with other studies. I do not say anything is wrong, I just think the limitations and possible artifacts could be better detailed.

Fogs and clouds are very similar in terms of organic matter and organic matter processing. It is a little odd that in many instances obvious examples from the fog literature are not cited. e.g. the paper should reference existing ESI-MS work in fog like Capiello (2003).

In some instances, it would be helpful if the authors could be more quantitative e.g. conclusions: what you mean by “Large numbers” (L8) or “Higher numbers” (L9)

Experimental question: ESI parameters: why were these particular conditions chosen? Was this optimized using any kind of model compounds? Was there any attempt made to change the voltage and see if widely different results were obtained.

Large parts of the manuscript are excessively descriptive going on and on discussing details. e.g. CHO section has a BDE discussion then CHON has a BDE discussion. ... It would make the manuscript more readable if this could be condensed and the main points extracted (may be throw in a table of BDEs).

Details: p20563L17: a newer reference for WSOC % would be more appropriate than

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a 15+ year old review

p20564L2: “under” cloud relevant conditions rather than “with”

p20564L13-14: reformulate, this does not sound right

p20564L24: and other locations: rather "gas phase" than "gaseous phase"

p20565L2.. not really correct as some of these studies don't include the species that are mentioned and van Pinxteren has 21%, please reformulate

P20568L24: may be replace “done” by “performed”

P20568L24: can you be more specific what the set of naturally occurring cloud water analytes is: how many species? may be which ones ? (if a small “set”)

P20570L12: 82.2 and 82.6% is the decimal really significant?

P20573L5: formulaS

P20584L25: significant digits?

References: Please check for correct initials (e.g. Sun Y.L. or Collett J.L.) or dates (Herckes 2006?)

Marshall reference: odd text symbols?

McLafferty: is this not McLafferty and Tureek?

Pruppacher should be Pruppacher and Klett (also in refs cited) and there is a newer version of their textbook

Figure 1: It would be best if the labels would be outside of the pie

Reference cited: Cappiello, A., De Simoni, E., Fiorucci, C., Mangani, F., Palma, P., Trufelli, H., Decesari, S., Facchini, M.C., Fuzzi, S., 2003. Molecular characterization of the water-soluble organic compounds in fogwater by ESIMS/MS. Environ. Sci. Technol. 37, 1229–1240.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 20561, 2013.

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13, C7249–C7252, 2013

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