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***Interactive comment on* “Size resolved airborne particulate polysaccharides in summer high Arctic” by C. Leck et al.**

Anonymous Referee #1

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Review of Leck et al. paper “Size resolved airborne particulate polysaccharides in summer high Arctic”

The paper presents the important information of previously unquantified particulate polysaccharides in marine environment with size resolved information being an important asset. The paper comes from a productive group specializing in Arctic environment and producing series of papers on different aspects of particulate organic matter in the Arctic. In this paper, however, the authors include too much of the secondary information which often does not help to clarify the processes, but instead defocuses and confuses the whole story. Overall, the paper is very detailed and would be suitable for publication in ACP after removing the most speculative statements, restructuring the paper and addressing other important issues.

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The English of the manuscript needs a particular attention as at the moment the paper contains too many bad style sentences. There are many instances suggesting that large sections were written by an inexperienced researcher and was not carefully read by the lead author before submission.

The major comments are the following:

1. Abstract should be restructured as at the moment it is loosely connected with size resolved information interspersed by the bulk chemical composition. I am not convinced that the relative abundance of submicrometer polysaccharides is closely related to length of time that the air mass spent over ice (see comments below). It is not a speculation that any significant chemical species could potentially alter CCN activation, but that is a more general claim.
2. The introduction of the paper looks rather comprehensive, but I wonder if the authors missed the paper of Ovadevaite et al. (2011) who presented highly relevant phenomenon of dual behaviour of sea spray OM in sub and supersaturated conditions. In my view, evidence of Orellana et al. (2011) about the existence of exopolymer gels in cloud droplets is very consistent with the ground measurements of Ovadnevaite et al. in the North Atlantic. While not directly relevant in the context of the paper it would add contextual clarity to the implicated effects of marine organics to the cloud processes.
3. I am very surprised at the reasoning of the turbulent flow in the inlet. The laminar flow conditions not only ensure lossless (or as close as possible) sampling, but also favour isokinetic split sampling of different instruments. Lossless sampling is crucial while turbulent flow enhances losses. The authors claim that turbulent flow helps mixing of air in the inlet, but there is nothing to mix when surface friction and turbulent eddies have already done that job. There is no gain with turbulent flow – only the loss.
4. The entire section 4 is assembled out of context, it's like putting cart in front of the horse. A lot of the information presented in this section is indeed important, but would only become relevant during the discussion. At the moment the reader is forced to

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believe upfront that all the presented information will have to be somehow related to the author's cause – which is against a scientific method proving everything point by point. Moreover, while the authors present all possible conditions potentially influencing the characteristics of aerosol samples, only selected conditions reiterated further in the discussion. Hence, major restructuring is required distributing portions of section 4 where appropriate.

5. The phenomenon of Days Over Ice (DOI) is inflated out of proportion. It is natural to expect that the supermicron particles will be present in decreasing numbers further from the primary source (open water) due to dry and wet deposition, thereby enhancing relative contribution of submicron ones which are typically removed only by wet deposition or in-cloud scavenging. Figure 2 can be easily replaced by a single sentence, stating the average time the air mass spent over the ice. The section 6.2 is quite misleading as well. Aren't the results broadly demonstrating the effect of deposition, especially considering accuracy and analytical uncertainties which make some of the claims hardly statistically significant? To me the Figure 9 suggests just that.

6. The authors present highly speculative idea of primary particles produced through the ice pores. There is no physical quantitative evidence presented in the paper or elsewhere in support of the above process leading to significant primary production. The differences in the polysaccharide content can equally be well explained by the air masses of different region/location, laden with varying amounts of primary polysaccharides. Different regions/locations of open water not only can experience different wind speeds, but more importantly different biological activity (trophic levels).

7. Another highly speculative idea not supported quantitatively is the production of particles by the breakup of fog droplets. While the idea was presented back in 1999, no quantitative support was provided to strengthen the idea since then. Can the authors present a physical/mechanistic explanation of the process, never mind the chemical one? Possibly some production may occur via the proposed pathway (especially considering nanosized particles - 5nm as in the referenced paper), but authors should at

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least consider conservation of mass in the context of this paper. Making more particles out of the bigger fog or cloud droplets does not yield mass, only number. The only gain in mass, which is needed to match primary fluxes with the measurements of polysaccharides, can come from oxidation or secondary production. The oxidation process can only contribute limited mass (doubling the primary OM at best), while invoking secondary production process invalidates the authors claim of primary origin of particulate polysaccharides. Actually, secondary production sounds very plausible considering VOCs emanating through the ice pores if production over ice is indeed proven in a more robust fashion.

8. The use of the term “mass” is very loose throughout the text. The authors should make it crystal clear what was the percentage of polysaccharides contributing to the total sea spray mass if primary or total particulate mass if secondary processes are involved. That can be done by calculating enrichment factor ($EF = THNS / (THNS + SS)$) or just referring to the total mass. At the moment the reader is confused about the significance of THNS contribution as there are numerous references to “the full mass median size distribution” or “total mass of polysaccharides determined”. The author perhaps could instead use the word “combined mass of polysaccharides” since “total” has the meaning of ALL mass including all chemical species.

9. The authors implicate wind speed explaining the differences in polysaccharide content between submicron and supermicron particles. However, all the existing sea spray source functions (Monahan, Gong, Martensson, Fuentes) include wind speed only as an overarching parameter. Even if there was some evidence that wind speed affects production of different size particles in a slightly different way it is far premature trying to corroborate the observed changes in chemical composition when existing primary organic matter source functions consider sea spray OM in bulk (typically submicron mode only). As it stands in the paper, it is another speculative idea.

10. Comparison with bubble bursting experiment is valuable, but authors should consider not only similarity in chemical composition, but also the enrichment.

11. Conclusions are not concise. Some of the quantitative information is missing, namely percentage contribution of THNS to total sea spray mass, while speculative comments should remain in the discussion only, not conclusions.

Minor comments:

1. How RH was of the impactor flow was controlled at 50%?
2. What was it meant for “1 μ m nylon filters” used for sampling artificially produced sea spray? Was it filter pore size? Was there any size cut applied or there were total suspended particulates sampled? If TSP, what would be the implication for the comparison with impactor samples?
3. Impactor concentrations refer to standard temperature and pressure. What was the range of actual temperatures and pressures?
4. Why the accuracy of LC/MS/MS was so poor as presented in Table 1 (close to and above 100%)?
5. Section 4.2 is unclear. The use of the term “recoupling” is confusing. If back trajectories suggested that air in the upper boundary layer had come from different region than in the lowest 100m that would suggest de-coupling, wouldn't it?
6. P9821, line 2. “jet drop sized aerosol particles” must be rephrased. Same applies to “similarly active film drop mode” on P9823, line 17.
7. I disagree with the statement that film drops would produce chemically different particles than fresh jet drops. Please rephrase or explain clearly. P9823, line 14.
8. Figure 6. Y axis is THNS concentration, where “2/3 of the mass within Aitken mode” come from. Possibly confusion over the “mass” term as previously noted.
9. How hexose implicated anthropogenic pollution affects the overall results?
10. Figure 3. The ice drift period is not denoted by DOY.

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11. Figure 5. The caption needs clarity with presented 6 graphs. Use “top”, “middle”, “bottom” graphs or name a,b,c,d,e,f,g.

12. Figure 8. What is the reason of presenting and comparing different size fractions: Aitken, Acc+Aitken and TSP. How the reader should interpret them?

Few examples of bad English:

P9812, lines 20-21.

P9813, lines 3-4.

P9815, lines 18-21.

P9820, lines 9-11.

P9823, lines 14-16

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