Reviewer 1

First, we would like to thank this reviewer for an unusually insightful review of our paper with many constructive comments and for showing a great deal of understanding, not only on the scientific issues but also on the challenge of writing an overview paper like this.

The problem is not to write the technical overview of instruments and the deployment, which can be used as a reference when later using the data; the problem is to write a paper that "tells a story" which is be enjoyable to read and to publish this in a science journal where some degree of scientific novelty and discussion is also usually required. There is no point these days to write a technical report; very few read those. At the same time, this type of paper is an opportunity to take a step back and take a look at the "why" and the "how" in a perspective that transcends this one expedition. This is what we started out trying, and hence we did make the choice the reviewer finds difficult; not "one or the other" but "both". Reading this reviewers comments, we feel like we have – in principle – been able to walk this balance and we are truly encouraged by the positive response this received.

But nothing is ever perfect and this reviewer also has several critical points, mostly concerning Section 7 and 8, where results from ASCOS are summarized and discussed. Below we will respond in detail to these; this will be done so that what we perceive as key comments from the review will be copied in *gray italic*, followed by our comments and suggestions for revisions; the latter will be written **in bold text**. We hope in this way it will be clear what are responses to review comments and what are suggestions for the revisions.

But before going into the details, we want to provide a background to Sections 7 and 8; why they are there and the problems this brings, and how we reasoned about these sections when they were conceived. These last two sections are intended to be the "icing on the cake" and also what lifts this paper from a technical report to a scientific paper. This requires "result", which is Section 7, and "discussion", which is Section 8.

This raises (at least) two main concerns:

i) Most ASCOS PIs for natural reasons want to first report their main findings in first-authored topical papers, where they get full credit, and not in an overview paper where they are "hidden" amongst numerous authors in alphabetical order;
ii) Differing views on the dominant sources of aerosol particles over the central Arctic Ocean, and how they vary with season and meteorological condition, is evident in the literature and remain also after ASCOS. Based on this observation, and what was found during ASCOS, we feel that the final answer, if one exists, still lies beyond our reach. This reviewer doesn't hide his/her preferences; this is fine and everyone is entitled to an opinion. However, in Section 7 we wanted to relate what we found in ASCOS, in relation to the objectives identified in section 2.

Hence the strategy we came up with was to base these sections, and in particular section 7, *only* on *published* results from ASCOS. The intention is therefore *not* to provide a balanced review of Arctic aerosol science in general; only to report what has come out on this from ASCOS, and only after it has already been published in other papers. The encompassing nature of an overview paper also means we wanted to avoid "evaluating" those results.

This approach naturally also poses the problem that lead authors of this paper will be in the hands, in a manner of speaking, of the co-PIs. We would like to stress that every coauthor of this paper has been given ample opportunity to weigh in on what should be included in Section 7, and in no case has a suggestion to include material been denied, as long as it is derived from ASCOS and is already published in a peer review journal. Therefore there may be scientific pieces that are missing, and will remain missing, in this paper. That is *not* to suggest they

are unimportant but simply and only because nothing has come out and been published of the analysis from ASCOS (yet) that speaks to those issues.

Finally there is one more point to be made in this context. Contrary to what someone may think ASCOS is *not* only an aerosol project. This is implied already in the first paragraph of Section 1, is implicit throughout Section 3 and also stated explicitly in the first paragraph of sections 4. Its uniqueness relates to its cross-disciplinary approach, involving a large number of disciplines. While the issue concerning the dominant sources of the high Arctic aerosol, that this reviewer has constructive comments on, is indeed very important, the objectives of ASCOS do not allow us to discuss this in more detail than many other findings. Indeed, a pure meteorologist could argue that issues on aerosol sources for Arctic clouds is secondary when confronted with the challenge to understand the meteorological processes that form clouds in the central Arctic; this is another issue that we also do not want to dominate this paper.

Main comments

However, I find the paper does a patchy job of summarizing the science. While the meteorological (7.3) and aerosol-cloud interaction sections (7.4) were nicely written, the entire discussion of the aerosol chemistry and physics (7.2) is focused on primary oceanic particles whereas no attention is given to an overall discussion of the aerosol present at the site. Likewise, there is little discussion given in the ocean chemistry measurements to the biogeochemistry of the system, and instead it is only bubble size spectra and turbulent mixing that are discussed in the water column section (7.1).

As discussed above, this section is not intended to be a broad review of on aerosol science in the central Arctic summer; it is intended as an overview of results from ASCOS and includes only a selection of results that has already been published by the respective primary PIs. Also, this section should not be read in isolation; it should be contrasted to the earlier sections, to judge if ASCOS brought answers to the questions posed; questions on the surface micro-layer and upper ocean bubbles were key questions providing the momentum – and a great deal of the funding – that made ASCOS happen and it would be remiss to not give results relating to these issues proper space. **This strategy was mentioned briefly in the introductory remarks to the section, and this text will now be expanded and revised to make this point even clearer.**

That being said, Section 7 can of course be much improved, **and we suggest that the way to do that is to include more results, much of which we must point out, was not published yet at the time of submission**; we are in a sense shooting at a moving target here.

On the other hand, some of the issues brought out by this reviewer, for example ocean chemistry and biochemistry, cannot be covered in the revised paper either, simply because this work has not yet lead to published results from ASCOS that we can refer to. Once we cross that border, every version of Section 7 that we can write will be challenged by the fraction of the science community that feel they were not given sufficient balance and in the end, the paper may be never published.

Hence in summary, published ASCOS papers, that have appeared after the original submission of this paper and that weigh in on this discussion, will be added and covered shortly in the revised paper. We believe this will provide an improved balance, still focusing on the ASCOS objectives. Moreover, we will also review the text carefully to avoid and remove overly certain statements of things we in fact do not know.

A short list of additional work that will be cited is included at the end of this response.

The Discussion section is similarly weighted, with much discussion about the primary organic particles. While I do not dismiss the novelty of the bubble measurements and detection of EPS substances in aerosol and cloud water, the paper does not justice to the field by ignoring the other aspects of the science. What is particularly bothersome is that there are some results from this study that point to the primary particles of oceanic origin NOT being the only source of CCN material.

Respectfully, we feel we have to at least partly disagree on this comment. Given that the discussion is still limited to ASCOS results and objectives, we do discuss two apparent paradoxes at some length: i) That the same EPS (polymer gel) substances are found in the ocean surface microlayer and in cloud water, while the direct flux observations cannot account for the variability in atmospheric aerosol number concentration; ii) The fact that essentially zero IN was observed near the surface while frozen drizzle from low clouds was persistently observed, thus indicating the presence of IN.

In this discussion, the importance of local as well as remote sources is discussed, as well as some hypotheses that could explain some of the paradoxes, including problems with the observations and other hypotheses. This discussion ends with the statement: "The relative importance of this local or regional biological source compared to advection from lower latitudes, at the MIZ and over the ice-free ocean south thereof where influences from man-made sources are still limited, remains an open question." We don't see how that can be interpreted as advocating the surface source above the long-range transport.

Undoubtedly this can be formulated even more carefully and we will go over this text again, to eliminate conclusions that are too uncertain and make clear what is speculation and what is known. There will also be the additions to the discussion necessary from the additions in Section 7. But we cannot provide answers that are not there; only acknowledge that this is the case, and we will not stray away too much from ASCOS results, although the discussion can be somewhat more general that the results in Section 7.

i The aerosol flux studies of Held et al. (2011) described in the paper indicate only a small source from open leads

Agreed; this is also mentioned in the text in Section 7 and is one of the primary discussion items in Section 8. **We will go over this text again and make sure it is made clear what is speculation and what is fact.**

ii. The work of Leck et al. (Size resolved airborne particulate polysaccharides in summer high Arctic C. Leck, Q. Gao, F. Mashayekhy Rad, and U. Nilsson Atmos. Chem. Phys. Discuss., 13, 9801-9847, 2013) has indicated that measured polysaccharide levels do not fully match the observed levels of organic aerosol measured during ASCOS).

Leck et al. (2013) is one of the newer papers that will be included in the revised. Chang et al., (2011) measured with an aerosol mass spectrometer (AMS), quantifying chemical composition of the *non-refractory* components of the aerosol. The AMS is thus not able to detect constituents that are chemically and physically stable at high temperatures, i.e., they have *refractory properties*. The AMS sampled ambient particles from 100 to 500 nm aerodynamic diameter. Leck et al. (2013) report on polysaccharides in size-resolved atmospheric particles from 35 nm to 10µm aerodynamic diameter. Phytoplankton and sea-ice algae biological secretions produce polymer gels: marine gels that are water insoluble, thermally stable (*refractory properties*), highly surface-active and highly hydrated. The polymer molecules also form three-dimensional networks to which other organic compounds (proteins and lipids) are readily bound. The marine gels span the whole size spectrum from 2−10 nm diameter (colloidal nanogels) up to micrometer-sized gels (colloidal microgels) that can aggregate to several hun-

dred micrometers. Hence Chang et al. was unable to detect the marine gels reported in Leck et al. and a direct comparison on the quantitative determination of the polymer gels cannot be performed.

The estimated fraction of the *non-refractory dissolved organic matter* in seawater characterized as neutral sugars was about 2.7 % on average (Gao et al. 2012) which would, based on estimates of sub-micrometer organics (*non-refractory*) in Chang et al. (2011), correspond to the order of 10 pmol m^{-3} of polymer gels (*refractory*). This back of the envelope calculation is comparable to result from Leck et al. (2013); measured mass concentration of polymer gels between 15.8 to 42.5 pmol m^{-3} (25^{th} to 75^{th} percentiles).

iii. The work of Chang et al. (Aerosol composition and sources in the Central Arctic Ocean during ASCOS, R. Y.-W. Chang, C. Leck, M. Graus, M. Müller, J. Paatero, J. F. Burkhart, A. Stohl, L. H. Orr, K. Hayden, S.-M. Li, A. Hansel, M. Tjernström, W. R. Leaitch, and J. P. D. Abbatt, Atmos. Chem. Phys., 11, 10619-10636, 2011) has evidence for a continental signature in the aerosol sampled during ASCOS, in addition to a long-range biogenic aerosol source with MSA as a major component. This paper should be discussed more in the overview as it is the only paper that I am aware of that assesses the source of air to the site during the campaign.

The study of Chang et al. (2011) was not able to detect the marine gels reported in Leck et al. (2013). The following is a quote from their paper: "Although the degree of oxygenation of the organic component of the marine biogenic aerosol would normally suggest that it had been processed in the atmosphere, we cannot rule out that local sources emitted these primary oxygenated organic aerosols, as suggested by previous findings over the pack ice (Leck and Bigg, 2005)." The primary oxygenated organic aerosols refer to the polymer gels. Related to the refractory limitation of the AMS the following was stated: "However, there also appears to be aerosol that was almost purely organic (31% by mass), although its source is unclear at this time."

Hence, while there are results in the Chang et al. paper that speak to a possible continental influence, it is a far cry from what can be called "evidence". The authors finally conclude: "… although it was not possible to conclusively identify a single source for this aerosol at this time, with primary marine and/or aged continental origins possible. The combination of poor correlations to typical continental tracers and the similarity of the spectrum to primary organic aerosol measured at Mace Head (Ovadnevaite et al., 2011) suggest a marine origin, although contributions from aged continental origins cannot be ruled out."

As written, the overview implies there is no transport of aerosol to the site (for example, top of page 13579) but that is apparently not the case, given the results from Chang et al.

This is a misunderstanding; what is argued here is that pollution or biomass-burning plumes, which we do see aloft on occasion, does not make it down into the boundary layer as far as we have been able to determine in any of the tracers or aerosol observations available in ASCOS. This, however, does not mean that aerosols, for example marine or terrestrial natural aerosols, were not transported to the ASCOS site, for example from the MIZ or from the open ocean beyond the ice edge.

This text will be revised to make clear that advection is a possible source of aerosols. Leck et al., 2013 and Kupiszewski et al., 2013 are two new papers that discuss in detail the source of airborne particles to the ASCOS site that will be included in the revised summary. The results from Chang et al. will also be considered in this context. Other original material suggesting remote sources and long-range transport, for example the paper by Shupe et al (2013), will also be cited in this discussion; interestingly the sub-

section of the ice-drift, about one week out of the 40 days of the expedition, for which Chang et al indicates a continental source is the same as analyzed by Shupe et al.

iv. Nucleation events were observed during ASCOS, when aerosol particle concentrations were low (page 13580, line 3). This is clear evidence that atmospheric nucleation of new particles was occurring, i.e. a primary source from the ocean would not be dependent on pre-existing aerosol surface numbers.

Of course nucleation events occurred and no one has ever claimed otherwise! However, there is no single answer to this question and we do point that out in Section 8. **One nucleation event is in fact analyzed in Kupiszewski et al. (2013), discussing the helicopter profiles and now that this paper is available, it will be include Section 7, as well as a modeling paper by Karl et al. (2013) based on ASCOS data, which offers more insight into this issue.**

v. The CCN closure experiments indicate that the organics present in the particles are insoluble (section 7.2), i.e. if primary organics are present then they are not CCN active, counter to the general emphasis of much of the paper. In fact, the belief is that the organics are likely sugars, which would be expected to be highly soluble.

The Arctic gels (water insoluble) during ASCOS have been shown to consist of hydrophilic and hydrophobic segments (Orellana et al. 2011), in agreement with their chemical behavior modeled by Xin et al. (2013; *J. Phys. Chem. Lett., 4, 2637−2642*). The interaction of the hydrophilic and hydrophobic entities on the behavior of the three-dimensional polysaccharide structures during the cloud droplet activation strongly suggests a dichotomous behavior for polymer gels (Ovadnevaite et al., 2011; *Geophys. Res. Lett., 38, L21806*); only partial wetting character is shown below 100% relative humidity and thus only weak hygroscopic growth but at the same time a high CCN activation efficiency is shown, which is promoted by the hydrophilicity or surface-active properties of the gels. The chemical and physical behavior of the polymer gels is in good agreement with the CCN closure experiments, which indicate that the organics present in the particles are water insoluble.

Instead, I recommend a balanced presentation of the results for primary oceanic particles, alongside a discussion of the evidence for in situ production of sulfate and MSA from DMS and the role of aerosol transport from lower latitudes.

MSA and DMS are already discussed in the context of regional sources mainly in the MIZ. With inclusion of the Kupiszewski paper, with an analysis of the helicopter profiles (see above), this discussion will now be expanded; further discussion of that would have to await more published ASCOS results on this topic.

We also do feel that we have a balanced discussion, as long as the reader understands that this is not an aerosol review paper. Recall, ASCOS is not an aerosol project; it is a "cloud project" and many processes contribute to how and where clouds are formed, besides the aerosols.

If this is not possible within the scope of the overview, then I recommend pulling out the science conclusions entirely and leaving the paper as a technical overview of the campaign. A second paper could be written on the sources of aerosol to the ASCOS field site.

Respectfully, that would be to admit defeat in the face of this discussion; we are convinced that this will gain nothing. The only thing that would happen is that the paper would likely be rejected because there is not enough new science or results discussed. We would encourage an Arctic aerosol synthesis paper to be written, based on ASCOS and possibly other data; however, that is not this paper.

More discussion and arguing sharpen everyone's arguments and promote further science; that must be the basic aim of all scientific publications. We hope to bring new results, without trespassing on the truth. The task of the authors is to not make firm statements were the evidence is not there. The responsibility of the reader is to not interpret things into the text that is not clearly stated there.

Finally, I found it exciting to read through the Appendix about all the novel instrumental approaches that will have information on the nature of the aerosol present, such as the radioactive tracers, single particle studies, thermal analysis of aerosol volatility, ATOFMS, etc. However, scant results from these instrumental approaches were presented. Why not?

If the PI responsible for a certain observations or instrument has not provided input in the form of a published paper; the policy we have adopted, described at the beginning of this response, hence precludes inclusion of results from that instrument.

Certainly, we would strongly encourage everyone that has results from ASCOS to publish those; this was the purpose of creating this ASCOS Special Issue.

Additional Specific Points

1. Introduction – very nicely written. Indeed, the writing style in the entire paper is excellent.

Thanks!

2. In Section 3, where the types of measurements required is discussed, more emphasis should be given to gas phase studies. Indeed most aerosol scientists (and I put myself into this category) often don't give the gas phase the emphasis it deserves. If the goal is to really determine the balance between primary oceanic particles and secondary atmospheric particles formed from DMS (or isoprene, e.g.) oxidation, then the gas phase precursors AND oxidants have to be measured in situ. If there is a weakness to the instrument suite on the Oden, it was the lack of emphasis given to these gas phase measurements.

One limitation in a paper such as this is the risk of it becoming so long that no one would care to read it. Yet being an interdisciplinary project, there are requests for all disciplines to be properly represented. Expanding on one section requires either shrinking something else, changing the interdisciplinary balance or increasing the length of an already very long paper.

With respect to the gas phase observations, in general gas phase chemistry was not the main subject of ASCOS; its aerosol program focused on formation and effects of marine particles and program limitations excluded comprehensive observations of all known and/or suspected atmospheric particle formation mechanisms. The DMS/MSA particle formation was covered as much as program limitations allowed which excluded the coverage of the complete complex photochemically active chemical process chain, albeit key components such as SO_2, DMS, and VOCs were measured.

DMS and acetonitrile sampling combined with sampling of atmospheric radioactive isotopes (e.g. ^{210}Pb, ^{222}Rn and ^{7}Be) were also performed as tracers for marine and continental sources respectively, to provide information on air mass origin and therefore on potential source regions contributing to the atmospheric composition.

While the reviewer is free to disagree with this strategy, this paper is about ASCOS and that is the priorities that were made. **That being said, we will reexamine this part of the paper very carefully in the revision, to make sure the need for gas phase observations is properly represented in a well-balanced fashion, but not at the cost of something else equally important.**

3. In Section 4, the measurements from the helicopter could be downplayed as it turns out there were not so many made.

On the contrary, we feel that the helicopter profiles, the only one of its kind, were in fact quite numerous, and contrasting to what other such more southerly measurements are available in the central Arctic, this facility and the data that comes out of it (now published) is a truly unique component of ASCOS that we are rather proud of. Rather than reducing this, the fact that results here are now published makes this a necessary component of the paper.

Also, I found the final few paragraphs in this section about pollution from the ship and the operating conditions to be too lengthy.

As for the text on pollution from the ship, sampling anything from a stationary ship in this very clean and pristine environment is an enormous challenge that maybe this reviewer has not entirely contemplated; it is a main and major factor in the entire experimental design and has ramifications far beyond the actual sampling on board. It is therefore very important for us to describe this so that the readers are reassured that our sampling is not contaminated. **In revision we will revisit this description to make sure this is written in a balanced way.**

4. It is said in Section 5 that fog was frequently encountered. Given the aerosol processing that fog will do, I would have been interested to read about the potential effects that it could have on aerosol processing, but this was not discussed.

This is discussed briefly in Section 8; **further discussion will be included in Section 7 with the results from the Karl et al. (2013) paper.**

5. I really liked the work of Mauritsen et al. described in section 7.4. It is nicely summarized here and this might be one of the major findings from the ASCOS study.

Thanks you; we think this is a key original result from ASCOS.

Additional published papers that will be added to the results/discussion:

Leck, C., Gao1, Q., Mashayekhy Rad, F., and Nilsson, U.: Size resolved airborne particulate polysaccharides in summer high Arctic, Atmos. Chem. Phys. Discuss., 13, 9801–9847, www.atmos-chem-phys-discuss.net/13/9801/2013/, doi:10.5194/acpd-13-9801-2013, 2013.

Kupiszewski, P., Leck, C., Tjernström, M., Sjogren, S., Sedlar, J., Graus, M., Müller, M., Brooks, B., Swietlicki, E., Norris, S., and Hansel, A.: Vertical profiling of aerosol particles and trace gases over the central Arctic Ocean during summer**,** Atmos. Chem. Phys. Discuss., 13, 10395 10461, www.atmos-chem-phys-discuss.net/13/10395/2013/, doi:10.5194/acpd-13-10395-2013, 2013.

Shupe, M. D., Persson, P. O. G., Brooks, I. M., Tjernström, M., Sedlar, J., Mauritsen, T., Sjogren, S., and Leck, C.: Cloud and boundary layer interactions over the Arctic sea-ice in late summer, Atmos. Chem. Phys. Discuss., 13, 13191–13244, www.atmos-chem-phys-discuss.net/13/13191/2013/ doi:10.5194/acpd-13-13191-2013, 2013.

Karl, M., Leck, C., Coz, E., and Heintzenberg, J.: Marine nanogels as a source of atmospheric nanoparticles in the high Arctic, Geophys., Res. Let., 40, 3738–3743, doi:10.1002/grl.50661, 2013.