

We are grateful to the reviewer for her/his positive comments and careful reading of the manuscript. Below we address the comments with our answers in blue. The numbering of pages and lines in our answers refer to the new version of the manuscript. Changes in the manuscript are written in red.

Anonymous Referee #2

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The paper presents a hard-earned data set of single particle analysis of samples collected in the high Arctic during the summer of 2008. The analysis is used to categorize particles according to their morphological and chemical properties. The most significant result is that “the particles to be activated into cloud droplets over the Arctic pack ice areas can not be seen as simply organic salts”. This result has implications for the modeling of CCN activity and cloud drop formation over the high Arctic. The paper should be published once the concerns outlined below are addressed.

p. 2, line 14: I’m not sure how particles can increase planetary albedo by absorbing sunlight.

We rephrased the sentence to

P2L13. “They alter the planetary albedo both directly by absorbing and scattering sunlight and indirectly ...”

p. 4, line 5: Is this meant to be Arctic SURFACE OCEAN DOM?

Corrected

p. 5, line 21: Should be SVALBARD.

Corrected

p. 11, line 25: Why is the size distribution characterized as “bimodal” when, on average, it contains 3+ peaks?

P11, line 24-28. Thank you for pointing out this unclarity. The sentence was rephrased to:

“The number size distribution of all imaged aerosol particles exhibited a maximum in the Aitken mode region at 32 nm in diameter and a double peak above 70 nm in the accumulation mode region with maxima at 89 nm and 147 nm with a shoulder to larger diameters at around 335 nm (see Fig. 3, red line).”

Figure 6 caption: I think it should be “unstable”, not instable. Also the labeling of the two lines is confusing. In the lower panel there are central particles of HP (red line) and unstable central particles (green line). What line represents the aggregate, aggregate with film and skeletal particles? Also – in this figure and throughout, it would help the reader immensely if legends were added to the plots.

Labels were added to the plot for further clarification.

Figure 9: It is very difficult to tell the orange and red line apart.

The color of the orange line was changed to blue.

p. 15, lines 13 – 20: It is unclear if the descriptions of bonds holding the marine polymer gels together are referring to gels in atmospheric aerosol or in seawater. Assuming seawater (given the Verdugo reference), the size range of nanogels (100 – 200 nm) and microgels (> 1000 nm) are larger than the airborne aggregate particles shown in Figure 9. How can this difference be reconciled?

The paragraph has been rewritten to:

p. 15, lines 16-32: “In seawater the observed size range of gel particles ranges from solvated nanogels (100-200 nm; Bigg et al., 2004) that can further anneal into microgels (> 1000 nm) by interpenetration and entanglement of neighboring nanogels or hydrophobic interaction. Changes in environmental factors like UV-B radiation (Orellana and Verdugo, 2003) or physico-chemical parameters like pH and temperature (Tanaka et al., 1980) lead to

inhibition/dispersion or volume change of the gel polymer assemblies. The transport from the ocean water into the atmosphere results in an enhanced exposure of the gel particles to solar UV-B radiation. Together with changes in the physico-chemical environment of the gel particles due to e.g. condensation of acidic gases onto the aerosol droplets the transport into the atmosphere thus might lead to fragmentation and/or shrinking of the gel matter and result in a reduced diameter of atmospheric gel particles compared to gel matter in the ocean (Leck and Bigg, 2005b; Orellana et al., 2011). Embedded in the polymer network is the high content of water (99%) that prevents the network from collapsing (Chin et al., 1998). The biopolymer networks of marine gels are highly surface active and show refractory properties are therefore not expected to evaporate under the electron beam.”

p. 17, line 10: Should be IMAGING.
Corrected

p. 17, lines 30 – 33: The dominant particle type was those containing both Na/K and Ca/Mg. Stating a “dominating content of Na/K” or Ca/Mg is not accurate.
Thank you for pointing this out, we corrected a mistake in the figure legend.

Figures 5, 6, and 9: Each of these figures contains some version of the aggregate particle type but each shows different size ranges for that particle type. What is the definition of “airborne aggregate particle” shown in Figure 9 (which resides in the 10 to 100 nm size range) compared to the aggregate particle type shown in Figure 9 (which is in a larger size range)?

Fig. 5 (middle panel) shows the number size distribution of **all gel-type particles** observed with SEM. The SEM images do not allow a differentiation into the subgroups of gel-type particles (“aggregate” particles, “aggregate with film” particles etc.). Gel-type particles were observed above 45 nm and up to 800 nm in diameter.

Fig. 6 (lower panel) shows the number size distributions for **central particles of halo particles**, with the green line representing stable central particles (comprising “aggregate” particles, “aggregate with film” particles and skeletal particles) imaged with TEM, and the red line unstable central particles of halo particles.

Fig. 9 shows the number size distributions of **airborne aggregate particles and their building blocks** at different locations as obtained with TEM. Sizing of “*aggregate*” particles in our study revealed the 10 to 100 nm size range for the building blocks of these particles.

Therefore Figures 5, 6 and 9 show number size distributions of different types of particles that cover different size ranges and thus cannot be readily compared.

How is this reconciled with the statement on p. 19, lines 30 – 33 that says “organic marine gel matter contributes to the particle number concentration . . . especially at diameters below 60 nm”?

It should be “below 75 nm” instead of “below 60 nm”.

Organic marine gel matter (in single particles and gel-type particles) contributes mainly to particle numbers below 75 nm because all other types of particles appear at higher diameters: unstable single particles appear mainly in the accumulation mode (Fig. 6, upper panel) and halo particles (with sulphur-containing satellites) appear above 75 nm (Fig. 5, lower panel).

p. 20, lines 12 – 29: The discussion of the fragmentation of larger particles into smaller particles in the atmosphere due to UV radiation exposure is highly speculative and not supported by direct evidence. The papers cited appear to be based on studies of seawater.

What thermodynamically viable mechanism can break apart particles in the 100 - 200 nm size range in the atmosphere? As far as I know, there are no reported observations of such events. (By the way, the Karl et al. (2013) and Tanaka et al. (1980) references, which may provide some insight here, are missing in the list of citations.)

It is correct that UV induced fragmentation of larger gel particles into smaller particles has been studied in seawater, as discussed in chapter 3.3 (p15, line 21-29). According to the current understanding the transport of gel matter from the ocean into the atmosphere occurs through bubble bursting and subsequent formation of small droplets that comprise seawater and e.g. marine gels (see Introduction, p.4, line 13 ff.). In the atmosphere these droplets can be subject to further condensation of water vapor or other substances, coalescence and incloud processing which all potentially change the physico-chemical conditions within the droplet (Tanaka et al., 1980). The latter in combination with an increased exposure to UV-B radiation in the atmosphere makes it in our view reasonable to discuss fragmentation and/or skinking as a possible mechanism to produce smaller sized fragments of marine gels (Leck and Bigg, 2010; Karl et al., 2013). This argument is further supported by the finding that spherical subunits in “aggregate”/“aggregate with film” particles and the dense inclusions in “mucus-like” particles appear at diameters down to 10 nm not only in the Arctic but also at different locations at lower latitudes (Fig. 9).

Tanaka et al. (1980) and Karl et al. (2013) were added to the reference list.