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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The summer aerosol in the Central Arctic 1991–2008: did it change or not?" *by* J. Heintzenberg and C. Leck

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We are most grateful for the extensive and constructive review.

Detailed Comments:

1) Typo corrected

2) In response to the criticism we weakened the sentence further to "Besides the limited extent of the data several causes may be responsible for these findings".

3) We do appreciate the unique value of data from station Alert. However, its latitude of 82.5 deg. North puts it south of the study area of the present analysis. Potential aerosol sources in the central Arctic thus cannot be studied in situ with Alert data. We maintain

that there are no surface aerosol data from the central pack ice. With our remark on future satellites we considered the upcoming combinations of active sensing systems with lidars and radars.

4) Answer: For no good reason, "Table 2" eliminated

5) The introduction has been shortened and rewritten in order to lead the reader from the ongoing changes in sea ice via a short discussion of potential causal mechanisms to the aerosol-cloud interaction as being one of the potential factors.

6) The obscure sentence has been rewritten to "Any ice cover raises the albedo of the surface whereas a scarcity of aerosol particles available for the condensation of water vapor reduce cloud albedo."

7) The obscure sentence was rewritten to" Together this complexity of the connection between sea ice, aerosol, clouds and radiation budget makes small changes in any of the components very important for the heat transfer to the ice and the subsequent summertime ice-melt."

8) Boundary conditions in the Chang et al. (2011) model are substantially different from the atmospheric conditions in the central summer Arctic. Here DMS concentrations are on the level of 40 pptv in August (Leck and Persson, 1996; Lunden et al., 2007; Karl et al., 2012). Although nucleation followed by growth continues to be the generally accepted understanding of the formation of particles below 25 nm diameter in continental and coastal regions, a direct demonstration that this is also the prevailing mechanism leading to new particles in the MBL of the central Arctic Ocean basin has yet to be given. Leck and Bigg (1999) reported on nucleation events that did not follow the typical banana-shaped growth curve (e.g., Heintzenberg et al., 2007). Instead, they observed enhanced levels of 3 - 5 nm diameter particles and simultaneous increases in particle number occurring in distinct size bands <50nm in diameter. Model simulations by Karl et al. (2011; 2012) indicate that growth of nucleation mode particles (3-25 nm diameter) to CCN sizes under typical Arctic conditions can only occur in the presence

of condensing organic vapors with sufficiently high concentrations in the gas phase (>7 pptv). This holds even for conditions of extremely condensational sink. Our main concern is that the DMS to SO2 conversion efficiency of the Chin et al scheme used in the Chang et al. model might be too high and is not constrained by observations. The Chan et al. model produces SO2 too efficiently by using a 75% SO2-yield from OH+DMS. Consequently SO2 levels in this model are an order of magnitude higher than in the Karl et al model. The latter values agree with measured data. Also the scheme in Chang et al. seems to produce an unrealistically efficient condensational growth through H2SO4, which stabilizes the initial embryos. The Chang et al. model uses constant [OH] during the simulation and the diurnal averages are definitely on the higher end of what can be expected in the central Arctic resulting in sulfuric acid available for nucleation being too high. Not even in a model case of the marginal ice zone with DMS = 410 ppt the Karl et al. (2012) model could produce sustained nucleation and growth based on sulfuric acid alone. Finally, the clear discrepancies in the temporal evolution between the modeled and observed number size distribution with respect to Aitken mode particles are indicative for missing processes, such as the fragmentation of larger particles (Leck and Bigg, 2010) a process not yet fully understood. In Leck and Bigg (2005b; 2005a; 2007) and near the end of our section 4 the processes leading to Aitken and accumulation mode particles are discussed. In the inner Arctic sulfur precursor concentrations appear to be too low to allow for substantial liquid phase oxidation. However, these processes may occur in air masses with higher DMS concentrations on their way from the open ocean via the marginal ice zone, which is mentioned in the revised text. In the revision we also clarify the uniqueness of the central Arctic in terms of nucleation and growth processes.

9) We agree with the reviewer about the inherent limitations of satellite sensors. However, we can imagine that future active (lidar) systems may be able to detect small aerosol amounts over the bright ice-covered central Arctic.

10) Typo corrected

11) The incriminated passage was eliminated.

12) We now explain what we mean by “structural” through the following introductory sentence of section 6: “The structural analysis presented in this section concerns the shape of the sub-micrometer particle size distributions and its potential connection to aerosol forming and transforming processes.”

13) Thank you for the suggested alternative, which we incorporated into the related sentence.

14) We see the difference in the different sorting procedures underlying figures 6 and 9. In Fig. 6 the data are simply sorted by total number yielding no obvious systematic change in the position of the concentration minimum between Aitken and accumulation tops as a function of N. If we, on the other hand, sort the data as a function of Hoppel diameter DHO we do indeed see a systematic variation of median N. So, it is the different independent variable that we build our argument on.

15) At total number concentrations of one per cubic centimeter and below as reported in the cited literature cloud formation will be limited by the number of available CCN. We would gladly send the reviewer a short video taken during the 2008 expedition, which demonstrates this limitation with a small (unscientific) experiment onboard the icebreaker.

16) See our response to comment eight.

17) We are not sure what the reviewer criticizes here and hope to have eliminated the problem by reformulating the sentence to “The combined aerosol statistics summarized in Table 5 and discussed in the present paper provide comprehensive physical data on the summer aerosol in the central Arctic with some connections to its formation and transformation processes. These data are the only surface aerosol information from this region and will probably remain so for some time because orbiting satellites do not cover the area close to the North Pole.”

Reviewer 3 (Stephen Warren)

We are grateful for the citing corrections and for the literature suggestions made by Stephen Warren.

Rererences

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 887, 2012.