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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Changes in aerosol properties during spring-summer period in the Arctic troposphere" *by* A.-C. Engvall et al.

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Answers to anonymous Referee #2

First of all we would like to thank anonymous reviewer #2 for constructive comments and suggestions, which have been a help to improve our paper.

In this paper we have described the observed fast transition of the Arctic aerosol from spring to summer based on data from 2000 to 2005 from the Zeppelin station. The sudden changes in the aerosol properties observed at the Zeppelin, which are shown to mainly happen in the second part of May, happen within 5-10 days [Ström et al.]. We are aware that both long-term and short-term measurements of aerosols from the Arctic have been published since the beginning of the 80s [Bondhaine et al 1980, Bondhaine et al 1989, Quinn et al 2002]. These studies presents data of number densities

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(nuclei counter), aerosol chemistry (impactor) and light scattering (nephelometer) and mention that there is a annual cycle observed of the both the chemistry and number densities. What differentiate our study with earlier studies, is that we present data of number densities, size distributions, Aitken- and accumulation mode numbers, gases and trajectories from the Zeppelin station, in a comprehensive multi annual way, concentrated on the period of the year when the transition of the aerosols shown to happen. Using available multi annual data we also attempt to asses the controlling variable(s) for this repeating occurrence of fast transition. This has not been published before.

Regarding comment from the reviewer we cannot exclude that part of the Aitken mode particles are produced by biogenic sources, which are mention for example in the study by Ferek et al (1995) and Quinn et al (2002), but there is no clear evidence that Aitken mode dominates from biogenic sources in the summer. It can be transported from above and gaseous precursors do not have to be linked to the ocean. Eneroth (Atmospheric transport of carbon Dioxide And Other Trace Species In High Northern Latitudes, PhD Thesis 2003, Stockholm University) showed with data and trajectories from the Zeppelin station that high number concentrations were associated to transport from higher altitudes. Further the maximum of the biogenic precursor DMS have been observed to occur in July-August. This is after the transition that we study here.

(The answers follow the order of the comments from the reviewer)

Comments

Fig 2: We agree with the reviewer and omitted this figure.

Fig. 4-6: With respect to figures 4-6 we do not agree with the reviewer as figure 6 evolves from figures 4 and 5. It is of course the same underlying data, but whereas figure 4 describes the change in the number density absolute values, figure 6 refers to a change in frequency of occurrence (expressed as ratio). Hence, the figures are indeed there to illustrate the transition, but do so for two different characteristics of the data, amplitude and time.

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Fig 7: We agree with the reviewer that both figures 7a and 7b are not needed for the text. We omitted Fig. 7b as the majority of the other data used is from planetary boundary layer.

Section 2.1: The comment regarding evidence of aerosol layers between the altitude of Ny-Ålesund and the lowest arrival height of the trajectories, the answer is no. However, in our opinion it is almost impossible to address the question about effects of layers below the trajectory meaningfully. The aerosol properties along the trajectory are unknown to us. The issue could conceivably have been addressed with aircraft measurements through flights directed upstream of the station. Still any layering below the trajectory is below the trajectory.

The MPL data available from the internet is a level 1 data which means it is range corrected "Normalized Relative Backscatter" and our use of the data was to have some data to suggest the boundary layer (BL) height based on the cloud top height. Hence we did not try to analyse the LIDAR data in terms of aerosol scattering. Days with clouds (which is common especially in summer) are very likely associated with well mixed BL, due to the dynamics driving the clouds and the feed back on the vertical motion that diabatic processes in the clouds cause. During clear conditions the story could be another, but we still don't have the information highlighted by the reviewer. The micro pulse lidar (MPL) was in this study used to look on the trends of the cloud cover over Ny-Ålesund and to get a feeling of boundary layer height based on cloud tops. In order to clarify we have rewritten the text as follow:

"The Micro Pulse Lidar (MPL) operated by the National Institute for Polar Research (NPIR) often show cloud tops around 2000 m altitude (Shiobara et. al., 2003). We thus define this altitude as the border between the BL and FT. The MPL data available through the internet is so called level 1, which shows the normalized relative backscatter signals and the data is solely used to get a feeling for typical boundary layer heights based on cloud tops."

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(p. 1220, line 24): As the MPL is placed a few kilometres from the Zeppelin station the direct link to the clouds at the Zeppelin station is not straight forward. Instead RH measurements from the Zeppelin station are more pertinent. By collecting available data of the relative humidity (RH) from the Zeppelin station for the years 2002 to 2005 we compared the data with the aerosol data, i.e. accumulation mode number density, from the Zeppelin station for the same period. Due to the cold temperatures and that aerosol might be affected by clouds above the station via precipitation the RH threshold for cloud affected aerosol data is not a RH=100%. Calculation of the median and quartiles for the number densities for RH=85%, RH=90%, RH=92% and RH=95% respective medians were Nacc85=60cm-3, Nacc90=39cm-3, Nacc92=35cm-3 and Nacc95=30cm-3. Our attempt is to reduce the influence of periods when clouds were at the station or in near vicinity. As such, the choice of 35cm-3 is subjective. We will include this analyse into the text to clarify the issue.

Fig 4: Comment accepted and accumulation- and Aitken mode particles are presented with different axes.

(p. 1223Ě): The melting of snow for any land areas of significance occur much later in the summer and do not affect the time period of this study. It is however of interest. Especially the thawing in July and August of the Siberian tundra is a process that should be looked into more detail (see Ström et al 2003). Most of the water around Svalbard is free from ice. The ice cover on the west side of Svalbard is very limited by the Gulf Stream that reaches these regions. The pack ice edge typically reaches only the most northern part of Svalbard. Any biogenic processes associated with ice brake up can only be connected to air masses from the north. Our air mass analyses do not reveal any such systematic differences.

Section 4.1.1: The four-days trajectories where in an early stage of the study compared to 10-days trajectories to investigate if there where any significant differences in main direction to the site. This study showed that four-days and 10-days mainly have the same direction as the four-day trajectories. The difference where the magnitude,

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which of course will be difference as the length of the trajectories are calculated for a more than twice the time. We concluded that four-days trajectories are enough for our purpose, i.e. study of the main direction to the reception point, Ny-Ålesund. We add following sentence in section 4.1.1:

“Note that we in an early stage compared the four-day with 10-days air mass back trajectories and they did not show significant difference with respect to our approach of the air mass origin used in the paper. Therefore we continue to use four-days trajectories thorough out the study.

(p. 1227, line 8): The detecting limit is based on how much air that are collected through the sample. This does not vary so much as they have daily routines to collect the filters, so the typical detection limit is around 0.01mg S m-3. Analysed results below the detection limits are reported as the half of the detection limit.

Figure 14: Comment accepted and corrected.

(p. 1236 last paragraph): We do not see any clear relation between SO2 levels, aerosol and transport patterns. It is however, true that the reduced anthropogenic source strength is also reflected in the decrease of SO2, which is obviously not compensated by biogenic processes. We have added to the text (section 5) that:

“Ěthe reduction in SO2 is consistent with scavenging and reduced anthropogenic source strength in the summer period.”

With respect to the reviewer’s comment about Pb-210 ratio with aerosol number density, the information’s essentially available in figure 10b where Pb-210 and aerosol accumulation surface area are plotted in same graph. Besides some excursions the two variables follow each other well. There are no indications of a change between these two that would explain the transition. We emphasis the main characteristics of Pb-210 and aerosol particles as given below.

“With its long lifetime 210Pb (half-life 22 years) the atmospheric lifetime is mainly gov-

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erned by the lifetime of aerosols (Paatero et al. 2003). Most of the atmospheric 210Pb is attached to accumulation mode aerosols."

And:

"Lead-210 is one of the tracers investigated in this study that shows a more pronounced change around DOY 140. Together with a decrease in the mean activity, the variability also drops notably. It is worthy to note again that 210Pb is associated with accumulation mode particles and thus could be a proxy of relative increase or decrease of the aerosol surface area, which also are dominated by accumulation mode aerosol (Fig. 10b)."

(p. 1230, lines 24-28): The source of the precursor gases is of great interest as this will be important in assessing the anthropogenic impact on particle nucleation. Here we focus on the trigger for the transition and reason for the timing of formation of new particles. Despite the potential increase of biogenic precursor gases (with ice brake up, sun and warmer ocean) the level of SO2 decrease with time. Hence we cannot easily argue that the transition is due to an increase in precursor gases unless we include some nucleating vapour not observed. To address this issue we made some simple estimates based on possible DMS levels.

Ferek et al 1995, discuss in their study the natural source of SO2 from oxidation of DMS. Their study shows how the concentration of DMS increase from spring to summer (July) with an average concentration of a few tens ppt and in the summer (August) values of 300ppt occurred. If we assume that 100% of the observed DMS are converted to SO2, in this estimating we use an average concentration of 30ppt DMS which will then give an SO2 concentration of 0.04mgSm-3. This is according to our observation of 0.07mgSm-3 below, what we call background concentration of SO2 at the Zeppelin station. Converting 300ppt to SO2 gives a concentration of 0.4mgSm-3, which is well below the highest observed concentration in our study (˜2mgm-3). Note that we assume that 100% of the DMS are converted which is an over estimation. Ferek et

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al. state that about 70% of the DMS are able to oxidise to the form of SO2. Given the discussion above and the decreasing trend in SO2, it is unlikely that a biogenic source would explain the transition.

If the transport alone would explain the transition from spring to summer aerosol, we should expect that the main transport should change from being “anthropogenic” to clean i.e. a change from sources at lower latitudes to come from the north and the biogenic source of precursor gases, which in our study is not the case. The transport does not show any systematic change from spring to summer, which means that the repeating trend of the transition in aerosol must be explained of something more than the transport and biogenic sources.

We have added following text in section 5:

“Regarding the biogenic sources to explain the production of the summer Aitken mode distribution (Ferek et al. 1995, Quinn et al 2002). Estimating DMS levels from observations by Ferek et al (1995) to SO2 levels show that base line of ˜0.04mgSm-3 and a maximum level of 0.4mgSm-3 that occur in the summer Arctic (July-August). If the transport alone would explain the transition from spring to summer aerosol, we should expect that the main transport should change from being “anthropogenic” to clean i.e. a change from sources at lower latitudes change and arrive from the north where the biogenic source of precursor gases is, which in our study is not the case. The transport does not show any systematic change from spring to summer, which means that the repeating trend of the transition in aerosol must be explained of something more than the transport and biogenic sources.“

Section 4.5.1 and 4.5.2: Regarding the question of how much the particles are affected by the relative humidity and hygroscopic growth and the possible change of this from spring to summer; we have evaluated the relative humidity (RH) data from the Zeppelin station. The influence of RH will affect the calculated condensation sink, CS that is used for estimation of equilibrium concentration of sulphuric acid. The relevant issue

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here is if including RH makes CS change with time differently than not including RH.

RH data are available only for years 2002 to 2005 i.e. not for the entire data set. We have used the dry aerosol size to calculate the evolution of the equilibrium concentration of H2SO4 vapour. To assess the impact of RH on CS we have calculated CS for dry condition as well as when including growth by aerosols using observed RH at the Zeppelin station. We consider a hygroscopic growth for an H2SO4 aerosol, following the approximation from Köpke et al (1997). The result shows that the hygroscopic growth affect the aerosol al most the same over the whole period.

Comparing CS for these two conditions, with or without including hygroscopic growth, show a CS increase with a factor 1.8 in April, 1.7 in May and a factor of 2 in June.

Hence the equilibrium concentration of H2SO4 vapour will decrease about a factor of two over the entire 3-month period, but the difference between the months are not large enough to explain the transition observed in equilibrium concentration of H2SO4. If anything, the slightly higher CS in summer compared to spring would tend to make the transition less pronounced.

This gives further, which we also comment in the manuscript, that the CS could be underestimated with a factor of 2-3. That means that our estimated value of the equilibrium concentration of H2SO4 vapours given the proxy OH values could be over estimated in spring and summer. However, it would not change the trend and hence our conclusion. Changing the assumed scaling value of 5e6 for OH to 2.5e6 easily modifies the increase in absolute value by a factor of two. This value was simply used to get the right order of magnitude for OH.

(p. 1237, lines 10-22): We will add some of the suggested references from the reviewer in the text. (See previous answer (p. 1230, lines 24-28))

(p. 1238, line 6): See answer above (Section 4.5.1 and 4.5.2).

(p. 1238, conclusion 2): To clarify this we add the following text:

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“The reduction of the over all anthropogenic influence is more gradual in nature (see for instance CO) and cannot explain the sudden change in aerosol properties.”

(p. 1239, last paragraph): To clarify this we add the following text:

“Ěremote sensing show similar sudden change in aerosol properties which indicates that the whole troposphere is involved in a similar rapid transition in aerosol properties Ě ”

Technical comments are accepted and corrected throughout the text.

References Eneroth, PhD thesis, Stockholm University, 2003. Ferek et al., JGR, Vol 100, 26093, 1995. Köpke et al. E. MPI-Rep., 243, 44pp, Hamburg, 1997. Quinn et al., JGR Vol 107, 2002. Ström et al. Phys. and Chem. Of Earth,28, 1181-1190, 2003

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