

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 referee #1

First we would like to thank anonymous reviewer #1 for constructive help and suggestions that help us to improve our paper.

Two major concerns and “sub-quotations” (1-3) by the reviewer were about the chosen anthropogenic tracers and the calculations of the nucleation potential. Our answer is as follows:

Anthropogenic tracers and long-range transport and sub-question 2

It appears as if the reviewer interpreted our text as saying that the tested hypothesis was an explanation for the transition in the long-range transport (LRT) of particles. With this in mind our text and figures is perhaps a little misleading. Our intention was not

to solely think of LRT in association of moving particles, but rather as an indication of change in conditions, i.e. air mass characteristics. Here conditions may involve a change in condensational sink for instance, but also changes in precursor gases. Long-range transport of small Aitken particles to the Arctic is not plausible. This would involve very rapid transport from a source region at lower latitudes. Although the manuscript has two sections for this, one titled LRT and one titled “Anthropogenic tracers”, we try to emphasize this better in the text by writing the following after the second sentence in section 4.2:

“The tracers used in this analysis serve as proxy for air mass characteristics. They are not necessarily intended to address long-way transport of aerosol particles, but rather to indicate changes in the composition of the air with respect to aerosol precursors and pre-existing particles.”

The trace gases SO₂, CO and Pb-210 were used to address an explanation for the observed sudden transition in aerosol properties. Although, SO₂ has a natural source in the Arctic, particularly later in the summer after the transition occurs, the three tracers are closely related to anthropogenic emissions. Both SO₂ and CO are associated with combustion processes whereas Pb-210 are more a tracer for the continental influence on aerosols. Given the lack of local sources in the Arctic for these tracers must be transported from elsewhere and hence much of their properties are controlled by long-range transport.

With respect to the comment by the reviewer we have added in text section 5:

“The reduction in SO₂ is consistent with scavenging and reduced anthropogenic source strength.”

The source of precursor gases is of great interest as this will be important in assessing the anthropogenic impact on particle nucleation. Here we focused on the trigger for the transition and reason for the timing of formation of new particles. Despite the potential increase in biogenic with ice break up, sun and a warmer ocean, the level of SO₂

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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 suggested from literature.

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 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.

In order to clarify the text we have added following sentence in section 4.2 as follow:

“The biogenic source from the DMS is anticipated to be larger during summer (Ferek et al. 1995). Observed peaks in the DMS concentration occurred to show values up to 300 ppt, while the background concentrations stayed at a few tens of ppt (Ferek et al 1995). Even though the observed peaks of DMS concentration would be 100% converted to sulfur (S) it would give an background level of 0.04mgSm⁻³ and the highest peak correspond to be about 0.4 mgSm⁻³, which is still well below the peaks observed in the springtime at the Zeppelin station. However, there is nothing in the trend of SO₂ that would suggest that an increased biogenic source would explain the observed sudden transition in aerosol properties.”

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With respect to Pb-210 there are really no disagreement with our text and the comment by the reviewer. And as we highlight in the text, Pb-210 is really the only tracer that can be linked to the sudden transition with respect to the temporal evolution. With the clarification of our use of these tracers, we feel there is not very much to add or change regarding Pb-210 in the text. We however 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 governed 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).”

Nucleation potential and hygroscopic growth and sub-questions 1 and 3

Regarding the question of how much the particles are affected by the relative humidity and hygroscopic growth and the possible contribution by this to the sudden transition; 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 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

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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 all 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.

Our study considers the repeating change in the aerosol properties from spring to summer in the Arctic [Ström et al 2003]. This has been observed in many measurements sites over the Arctic region (Bodhaine et al 1981, Bodhaine 1989, Quinn et al. 2002, Ström et al 2003). We use a phenomenological model to investigate if it is possible to estimate this transition based on the equilibrium concentration of sulphuric acid and aerosol properties. To compare to the yearly repeating change that are observed at the Zeppelin station. This is very useful as a tool for future planning for example of aircraft campaigns or measurements programmes. Again this is not meant to be a detail study to explain the new particle formation. For this porous there are need for more

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detail studies and more sophisticated models. But again, we show with a simplified model for what parameters, besides the transport, that has an important role for the aerosol properties that are observed at the Zeppelin station. For this reason there the “detailed” analyse should be included in our study.

Minor comments

(p. 1219): Accepted and corrected.

(p. 1223): Authors conclude that both figures are necessary to show the stable repeating pattern of the aerosol properties.

(p. 1223): Fig 6a-b. “The ATI appear to provide a more distinct measure of when the atmosphere has reached summer conditions i.e. dominance in Aitken mode particles.” By study Fig 6a the ATI show to stay over a certain value around 0.4 in the second half of the period, i.e around DOY 150. To classify a threshold of $ATI=0.2$ to be the threshold for summer is not enough if we study figure 6b. From this figure $ATI=0.4$ is for some of the years already reached in the spring, i.e. $DOY < 150$. Even though it is not last for ten days that we had as a criteria for when summer is reached. As can be seen in Fig 6b, $ATI=0.2$ is reached in the early part of the period and also last for 10 days, and are therefore not representative to be classified as a summer condition.

(p. 1224): 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, the difference where the magnitude, which of course will be difference as the length of the trajectories are calculated for a more than a factor of two longer in time. So 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 tra-

jectories 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. 1226, section 4.1.2.): The comment has been taken into consideration and the text has partly been rewritten.

(p. 1229-1230, section 4.3.): The comment has been taken into consideration and the text has partly been rewritten.

(p. 1232Ě): Accepted and corrected.

(p. 1233Ě): The comment has been clarified in the text.

(p. 1238Ě): To study the nucleation in detail one should investigate daily nucleation peaks. In this study we investigate trends in the observed data and main driven mechanism for this observed trend shown in the study by Ström et al (2003).

(p. 1239Ě): This part we already have included in the text under point 3 in the Summary:

“With a simplified model, which delivers the nucleation potential for new particle formation in a form of vapour equilibrium concentration of H₂SO₄ we suggest that the aerosol microphysical properties are result of a delicate balance between incoming solar radiation, transport and condensational sink processes.”

Point 2 have been clarified with following sentence:

“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.”

(Reference list): Corrected after comment from the reviewer.

Figures

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

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Fig 3: Accepted and corrected; the y-label is corrected and the lines have been changed to colours.

Fig 4: Accepted and corrected.

Fig 5: Accepted and corrected.

Fig. 6: Accepted and the text have been clarified from recommendation from the reviewers.

Fig. 8: Corrected.

Fig 11: The comment from the reviewer to clarify the figure have been accepted and corrected.

Fig 12: We don't think that we should change the scale of the y-axis as the y-axis today are spanning over the range of data that.

References Ferek et al., JGR, Vol 100, 26093, 1995. Bodhaine et al., Atmos. Environ., vol 15, 1375, 1981 Bodhaine, Atmos. Environ., vol 23, 2357, 1989 Koepke 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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