Atmos. Chem. Phys. Discuss., 7, S259–S264, 2007
www.atmos-chem-phys-discuss.net/7/S259/2007/

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.

Anonymous Referee #1

Received and published: 22 February 2007

Review of Engvall et al. "Changes in aerosol properties during spring-summer period in the Arctic troposphere"

The manuscript presents an analysis of aerosol size distribution transition taking place in the Arctic in late spring/ early summer. The observations at Svalbard over six years show a consistent and quite dramatic shift from accumulation mode dominated to Aitken mode dominated distribution within a two-week time window at the end of May/beginning of June. This work aims at finding the controlling factor behind this shift by using measurements of gas and particle phase properties as well as trajectory modelling.

All in all, the manuscript addresses an interesting scientific topic which is currently

not well understood. The presentation is well structured and easy to follow, although it would benefit from an additional round of editing (grammar, spelling, missing references etc.). The methodology is outlined clearly and assumptions necessary for the analysis have been made explicit. The many figures included are for the most part informative and relevant for the study.

However, I do not find all the results behind the main conclusions very convincing. My two major concerns are:

First, it is unclear to me, how the analysis of the chosen anthropogenic tracers shows that long-range transport is important for the shift from spring to summer distributions but cannot alone explain it. Concentrations of SO2 and CO are influenced also by sources and sinks not directly linked to (aerosol) transport (e.g. DMS emissions and clouds for SO2, OH concentration for CO). It is therefore not easy to isolate the effect of transport from other effects for these tracer concentrations, let alone to use them as indicators of particle transport. Furthermore, if SO2 and CO are despite this used as tracers for long-range transport of particles, why do the results conflict with the analysis of Pb-210, perhaps the least disputable indicator of continental particle influence? On the other hand, if the authors find that SO2 and CO cannot be reliably used as indicators of long-range transport of particles, how can they rule out that changes in transport patterns are not solely responsible for the observed size distribution changes? After all, there is a clear drop in Pb-210 concentration (associated with accumulation mode of continental origin) at around DOY 140.

These questions need more elaboration in the text. If one or more of the tracers are found not to be good indicators of particle transport, discussion on them could be significantly condensed - the analysis is quite lengthy as it already is.

Second, I find the calculation and analysis of nucleation potential shaky for several reasons:

1) As atmospheric humidity in the Arctic typically increases towards summer, neglect-

ACPD
7, S259–S264, 2007

ing hygroscopic growth of particles in the analysis causes more bias in summer than in spring. Because of this, the equilibrium concentration of sulphuric acid is overestimated more in summer than in spring, partly favouring the conclusion made by the authors. If relative humidity measurements are available at Zeppelin, the authors could make an educated guess on the composition of the particles and recalculate CS using particle wet sizes (however, see points 2 and 3).

2) New particle formation typically takes place over a limited period (observable formation in BL ~ couple of hours) of a day and, according to our current understanding, is most likely when the sulphuric acid concentration reaches its peak values. Therefore, daily averages of SO2, CS and diffuse solar radiation S do not necessarily represent the actually conditions in which nucleation and subsequent growth happen. In particular, using daily mean of S is misleading as the difference in midday radiation (i.e. peak H2SO4 concentration) in the Arctic is probably not as radical between spring and summer as the difference in daily averaged radiation.

3) As the authors themselves acknowledge, new particle formation events observed at 10 nm particle size at Zeppelin have initially taken place far away from the station. In fact, earlier studies suggest that new particle formation in the marine atmosphere most likely happens in the free troposphere and is entrained downwards to the boundary layer. It is therefore doubtful whether the (daily average) conditions at Zeppelin station resemble the conditions at the actual nucleation site in any meaningful way.

For these reasons, the authors need to be extremely careful with the conclusions they draw based on their phenomenological model. I would even recommend excluding the current detailed analysis and keeping to general observed features in the Arctic atmosphere (including free troposphere!): decrease in SO2 and CS, increase in OH etc. and the possible indications to new particle formation. In any case, the method used is far too crude to give any guess of a critical value of H2SO4. It is also important to remember that factors other than H2SO4 concentration and CS affect the new particle formation rates. For example binary nucleation in FT is thought to be favoured by low

ACPD
7, S259–S264, 2007

EGU

temperatures - it is therefore possible that the cold atmosphere and high SO2 concentrations in spring lead to significant particle production in FT although OH concentration is not very high.

Minor comments:

Please check and revise language throughout the text (sentences missing predicate verbs, missing full stops and blanks between words, typos etc.)

p. 1219, lines 4-5: Is this title for subsection 2.2.1? As there are no other subsections, either delete or change title for 2.2.

p. 1223: First full paragraph is unnecessary as figures 4 and 5 contain in practise the same information. For the same reason figure 5 can be deleted.

p. 1223, second paragraph: The definition of ATI and the motivation behind its use are not very clearly presented in the text. Why does ATI 'provide a more distinct measure of when the atmosphere has reached summer conditions'? Is the threshold value 0.4 chosen to provide best fit to the available data at around DOY 150? If so, why is ATI a better measure of reaching summer conditions than earlier presented analysis of changes in Aitken mode concentrations? If ATI is to provide an independent measure of summer conditions, why doesn't the clear jump after DOY 130 (e.g. ATI=0.2) represent transition between spring and summer?

p. 1224, section 4.1.1: Is 4 days long enough time to capture all the relevant characteristics of the transport? Why?

p. 1225-1226, section 4.1.2: This section should be partly rewritten to focus better on the question at hand, i.e. do the transport patterns explain the transition in aerosol properties. Explain how e.g. the fraction of N-component or correlation between the two altitudes are relevant to this question.

p. 1229-1230, section 4.3: I find it difficult to see the need for such a long discussion of individual days. The section should be motivated better or written in a more general,

ACPD
7, S259–S264, 2007

condensed form to support the conclusions. Again, how does this analysis show that transport is important but not enough for the transition?

p. 1232, lines 4-6: These two sentences contradict. OH is not observed.

p. 1233, line : It is slightly misleading to say that equilibrium concentrations vary from close to zero to up to 1d8 when figs 12 and 13 indicate that only single points (most likely bad data) fall to these extremes. The vast majority of points fall between 8d5 and 3d7, which are more or less realistic values.

p. 1238, first full paragraph: As already pointed out: weekly median radiation is not a good indicator of nucleation potential. Daily peak values are much more likely to affect nucleation rates.

p. 1239, main conclusions: Based on the results of the analysis, I find the formulation of the conclusions too strong. See comments above. It seems also likely that there is no one main process that controls the transition but that change in transport patterns as well as enhanced removal mechanisms play an important role in addition to aerosol microphysics (conclusions 2 and 4).

Reference list: missing (at least) Rahn 1981 and Paatero 2000

Figures:

Fig 2. Due to log scale on x axis it is very difficult to compare different months to each other. The figure could be even omitted as the same information is contained in Fig. 1.

Fig. 3: The dN/dlogDp values are very low. E.g. Strom et al. (2003) reported approx. an order of magnitude higher values for Svalbard (in cm-3; what are the units here?). As the figure is quite busy, it would be good to plot different months with different colours (but keep different line styles for black-and-white prints). Explanation of line styles needs to be given only in caption or (preferably) in legend.

Fig. 4: Mention that thin lines are standard deviations.

ACPD

7, S259–S264, 2007

Fig. 5: y-axis should be labelled Rait/Acc

Fig. 6: The caption is formulated in a very complicated way. Do the authors mean 'the ratio of frequency of R>1 and frequency of R<1'? y axis label should be ATI. Horizontal line for 'threshold value' (currently 0.4) could be added.

Fig. 8: Check caption. There are no blue and red lines in the figure.

Fig. 11: E-W contribution would be easier to see if particles/SO2/Pb-210 were plotted also on lower panel of each subplot.

Fig. 12: The scale on y axis should be changed as most data points fall between 1d6 and 1d7 /cc. On current axis the temporal variation (beyond increase in variation towards summer) is impossible to see.

Fig. 14: Correct y axis labels.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 1215, 2007.