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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.

Anonymous Referee #2

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The paper describes a rapid, repeatable change in the size distribution of aerosol measured at Ny-Alesund during the spring to summer transition. Within a 10 day window during the transition between seasons, the aerosol goes from being dominated by the accumulation mode to being dominated by the Aitken mode. Several factors are investigated to determine the extent to which they contribute to the rapid transition including.transport patterns, aerosol source and sink processes, and solar radiation. As a result of this lengthy analysis, it is concluded that the transition is driven by incoming solar radiation as transport and condensational sink processes compensate for one another. Much of what is presented in the paper has been previously published over the past 20+ years (decrease of long range transport of pollutions during the spring to summer transition, transition between spring and summer aerosol properties, in-

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crease of Aitken mode particles due to scavenging of larger particles, biogenic source of aerosol precursor gases during the late spring/summer). More specifics on this previous published information follow below. The new piece of information here is how rapid the transition is at Ny Alesund each year. The paper briefly mentions the change in aerosol size distribution as described by Quinn et al. (2002) and Strom et al. (2003). There are, however, many other papers that provide further information relevant to the one at hand. For example, reports of the difference in the spring and summertime aerosol size distributions in the Arctic aerosol date back at least to the early 1980s (e.g., Bodhaine et al., 1981; Bodhaine, 1989). In addition, the correlation of dimethylsulfide release from the ocean as the ice recedes followed by a maximum in the Aitken mode number concentration was reported by Ferek et al. (1995). Finally, a correlation between methanesulfonic acid and particle number concentration with both increasing during the summer months was reported by Quinn et al. (2002). Hence, the shift from an accumulation to Aitken mode dominated aerosol during the spring to summer transition has long been recognized. In addition, it has been attributed to several changes that occur on roughly the same time scale during this time of year including the release of biogenic precursor gases as the ice melts, reduction in particle surface area as deposition increases, and increased solar radiation for the oxidation of precursor gases. A more thorough discussion of these past periods, particularly the evidence for the biogenic source of the summertime Aitken mode particles would enhance the paper. It should be stated that the spring to summer aerosol transition has been previously attributed to a combination of factors (changes in transport, deposition and resulting aerosol scavenging, precursor gases, solar radiation) but that this paper attempts to determine the most influential factor(s).

Comments:

There are a several instances of redundant figures or an over-reliance on figures to tell the story. Either Figure 1 or 2 would suffice. Both are not needed as they give the same information. Figures 4, 5, and 6 also convey very similar information, i.e., that

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the change occurs around days 140 to 150. One or more of these figures could be omitted with the information still described in the text. Both figures 7a and 7b are not needed. One could be omitted with information from the second given in the text.

Section 2.1.: Was there any evidence of aerosol layers between the altitude of Ny Alesund and the lowest arrival height of the trajectories? Was the lowest useful height of the micropulse lidar signal low enough to reveal whether the aerosol was well mixed between the altitude of Ny Alesund and 1000 m?

p. 1220, line 24: Can the micropulse lidar be used to verify the presence of clouds as determined by the accumulation mode number density? Can evidence be provided that the removal of 22% of the data where the number density of the accumulation mode is less than 35 1/cc did not affect or bias the analysis?

Figure 4: There is about a factor of 2 decrease in the accumulation mode number concentration over the period shown. Though not as large as the increase in Aitken mode number concentration, it still appears significant and, obviously, is related to the decrease in a condensational sink. The figure could be improved by plotting the number concentration for the two different size ranges on different y-axes so the trend in accumulation mode particles could be seen more clearly.

p. 1223: How does the timing of this two week period for the transition from spring to summer correspond to break up and melting of the snow and ice and the potential release of biogenic precursor gases from the ocean to the atmosphere?

Section 4.1.1: How useful are 4 day back trajectories for this analysis? What fraction of the trajectories showed air masses coming from outside the Arctic 4 days back in time? If it is a small fraction, then it will be difficult to determine the influence of long range transport on the spring to summer transition.

p. 1227, line 8: Were measured SO2 concentrations less than the detection limit at times? If so, they should be reported as $<$ 0.01 ugS/m3.

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Figure 14. Labels on the y-axis should be the ratio, not the concentration of H2SO4.

p. 1236, last paragraph: Changing transport patterns and less transport from polluted regions may also contribute to the decrease in monthly mean SO2 between April and May.

Throughout paper: Pb210 is not an independent tracer of anthropogenic activity since it is condensed onto and transported by the aerosol. In many of the accumulation mode - Pb210 comparisons, it might be more revealing to consider the ratio of Pb210 to number density of accumulation mode particles. For example, in Figure 15 and in the discussion at the top of p. 1227.

p. 1230, lines 24 - 28: It is not clear why it is "Ěagainst the expectations air mass transport alone cannot explain at all the spring-summer transition in aerosol properties." Many previously published papers (see references above) have cited the spring sunrise in conjunction with availability of biogenic precursor gases as an explanation for the nucleation of new particles during the spring-summer transition.

Sections 4.5.1 and 4.5.2.: The calculations of the source and sink terms seem highly uncertain - especially the lack of consideration of the water uptake by the accumulation mode aerosol. A range of values for the equilibrium H2SO4 concentration should be given that reflects these uncertainties. This information could be conveyed in Figures 12 and/or 13.

p. 1237, lines 10 - 22: Previously published papers that demonstrate a link between biogenic precursor gases in the late spring and summer and nucleation of new particles should be mentioned (see references above).

p. 1238. line 6: But CS could be underestimated due to not considering the humidification of the aerosol. State how this affects the balance between CS, SO2, and radiation.

p. 1239, conclusion 2: This conclusion downplays the role of decreased transport of pollutant aerosols. Without the decreased transport of pollutant aerosols and aerosol

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precursors, the surface area of the aerosol would not decrease, and nucleation would not be enhanced. Therefore, it is a significant step in the transition from spring to summer.

p. 1239, last paragraph: Please provide a more complete description in support of the statement that :Ěremote sensing data show the whole troposphere to be involved in a similar transition."

Technical comments:

p. 1217, line 9: Change to "Several years later SCIENTISTS showedĚ"

p. 1219, lines 4 and 5: Incomplete sentence.

p. 1220, line 17: Change to "As the number concentration RANGES overĚ"

p. 1222, line 22: Change to "Ěmode particles are PRIMARILY A result ofĚ.."

p. 1223, line 14 - 15: Change to "Ěwhen the index REACHES AND REMAINS above 0.4 and LASTS for at least 10 days."

p. 1224, line 23 - 24: Delete sentence starting with "Handling of thisĚ" and change next sentence to "A variable called "trajectory vector" was derived to relate the large numbers of trajectories to other tracer data."

p. 1225, lines 18 - 19: Incomplete sentence.

References

Bodhaine et al., Atmos. Environ., vol. 15, 1375, 1981. Bodhaine et al., Atmos. Environ., vol. 23, 2357, 1989. Ferek et al., JGR, Vol. 100, 26093, 1995. Quinn et al., JGR, Vol. 107, 2002.

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

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