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Atmospheric Chemistry and Physics Discussions

Interactive comment on "One year boundary layer aerosol size distribution data from five Nordic background stations" *by* P. Tunved et al.

P. Tunved et al.

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General comment by the authors

The referee has added several important comments increasing the readability of the article. The work put down by the referees is greatly appreciated and acknowledged in the manuscript. In most cases we agreed that the refereesŠ comments were justified. However, in a few occasions we did not agree with the referees Referee 2 suggested a number of additions to the manuscript, although interesting we find it difficult to include all of these in the current study and some of the suggestions better serve as starting points for subsequent studies. Main changes in the manuscript concern the nucleation part, which has been rewritten to meet with the referees comments. The obviously inconsistent terminology regarding different advection situations was also revised. Minor changes have been included besides those mentioned below. This mainly concerns typos. Our detailed answer is clearly outlined in the point-by-point response below. All

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changes significantly altering the content and/or conclusions in the manuscript is given in the following text. We believe that the revised version is an even more understandable and interesting manuscript thanks to the reviewers comments.

Anonymous Referee 1

This manuscript presents analysis of a large data set of aerosol number size distributions in Nordic countries. The data set is unique, there are very few long-term measurements available and the findings should be published. The analysis has been done carefully and much effort has been devoted. However, there are some aspects that the authors should consider (see below) and before publication in ACP revisions are required.

General comments

1. The Vavihill data is available only for shorter period and it is used and discussed very limitedly in the manuscript. The authors should consider either omitting this data or explaining clearly the measurement period and discussing the limitations for the use of this data set.

Response: The limitations when using the Vavihill dataset was only briefly addressed in the original manuscript (p.2796, line 8-9). This part is now more thorough.

"During the year of study, measurements at Vavihill only covers a portion of the winter and spring period (February-April 2001). Therefore no seasonal variation can be evaluated with this data. "

Was added following section 2.3.5, p. 2792. The Vavihill data should however not be omitted, since it may be used evaluate transport related changes during specific seasons (i.e. spring) for which data indeed are available.

2. On page 2787 line 27, the authors tell that three modes are used for fitting the size distributions. This has to be motivated clearly using this specific data, and not only using general statements on aerosol processes. It is conflicting to see

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four modes used in tables 3,5, and 6 and especially conflicting with statement p.2802 lines 5- 6, which indicates that 3 modes were not enough and actually 4 modes were used. In addition, the definition of the modes is rather unclear, especially as the authors introduce the concept of Aitken 1 mode (p.2799 last line and p.2800 first line). So what is the difference between nucleation mode and Aitken 1 mode? And also, looking at table 2, some values of Dg1 (Värriö) are same order of magnitude as Dg2 (Pallas, Värriö). The limitations in the fitting procedure need to be considered.

Response: Under 2.2 we re-wrote the part from p. 2788, l. 27 to p.2789, l.1 to instead include following information. This paragraph is found at the end of section 2.2:

"In order to understand this convention we have to be aware of the fact that the nature of the processes contributing to the different modes is not to be considered as discrete, but rather a continuous process. For example we can consider the growth of the nuclei mode due to condensation into the Aitken size range. If an Aitken mode already is present, which of course is the normal case, this growth would lead to appearance of two modes in the Aitken size range. Since smaller particles grow faster (by size) than larger, the two modes would in due time appear as one in terms of physical size distribution properties. In order to take similar situations into account it is necessary to include a term for this growing mode. In the following we thus include the term Aitken 1 and Aitken 2, where Aitken 2 is referred to as the semi-persistent Aitken mode and Aitken 1 denotes the growing mode. Only very seldom we were able to resolve more than one mode in the accumulation mode size range so this type of definition is limited to the Aitken mode only. Fitting of three modes to the aerosol size distribution is of course an oversimplification. Four or even five modes may also be present as reported by Birmili et al. (1998) and Birmili et al. (2001), and this is not considered in our analysis. However, as shown in section 3.4, three modes are sufficient to reproduce the measured size distribution in this dataset. "

The use of 3 modes for the fitting procedure was evaluated and proved to produce

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satisfying results. Parts of this test was also shown and discussed in the article under section 3.4 on page 2798, especially lines 19-24.

3. Nucleation is discussed extensively in the manuscript. However, the authors do not show any example of how nucleation is detected at various stations (e.g. during northern air masses at all stations). The data shown and examples selected should be more balanced with the manuscript text.

Response: Nucleation is an important phenomenon in the boundary layer and has been discussed in several publications during recent years (E.g. Kulmala et al., 2001, Aalto et al., 2001, Nilsson et al., 2000, Nilsson et al., 2001a, Nilsson et al., 2001b). Considering the nature of this article as well as recent focus on the subject of nucleation it is natural to include somewhat detailed information of the issue in this manuscript. It is however agreed that the former manuscript suffered from a slight imbalance regarding text vs. figures dealing with the issue of nucleation. The revised manuscript includes general statements concerning important features of the nucleation phenomena along with a figure showing typical nucleation events as observed at the different stations.

"During this year of study, the frequency of the nucleation events has been shown to be largest around springtime, between March-May. This seasonal variation in nucleation frequency has been observed at Hyytiälä earlier (e.g. Kulmala et al 2001, Mäkelä et al., 2000).

Figure 2 shows example of typical events at the measurement stations Aspvreten and Värriö, revealing the commonly observed characteristics of the nucleation phenomenon. The fact that we are able to follow the growth for several hours indicates that the phenomenon occurs on a large spatial scale. Nucleation has earlier been reported to occur during sunny days (Kulmala et al., 2001). Further it was in the present study concluded that nucleation occur in air arriving from N/NW in most of the cases. Nucleation as shown in figure 2 is only very seldom observed in air advected from S/SE. Earlier reports stretch the importance of cold air advection and boundary layer

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height (e.g. Nilsson et al 2001). Also, high concentration of pre-existing aerosol is believed to quench the nucleation due to large condensational and coagulation sink, due to removal of the precursor gases and initially formed cluster respectively. . In table 2 statistics of the observed nucleation events during the year of study is summarized. In this statistics we only include the most pronounced events following a definition by Mäkelä et al. (2000b). The growth rate was found to be largest at Aspvreten, by 2.3nm/h. Peak concentrations observed during the events was highest at Aspvreten (9600 cm^{-3}on average) and lowest at Värriö and Pallas. Thus both growth rate and number concentration of newly formed particles is highest at Aspvreten. ”

Further, on l. 24, p. 2794 - l.3, p.2793, following section was excluded since the information is contained above:

“Often, but not always, the growth of these freshly nucleated particles can be followed for several hours until they reach the Aitken size range. A mean horizontal wind speed of 10 ms^{-1}in conjunction with a phenomena that may be tracked for three hours, yields a horizontal extent of the phenomenon which is at least 100km”

4. Discussion related to trajectories is somewhat confusing. The authors should e.g. clearly explain what means that "trajectories oriented NE" (page 2799 line 23). Similarly, p. 2800 line 4: how is SW airflow defined (i.e. what is the wind direction)? And p. 2803 lines 3-5: NE-transport path / NE oriented clusters? And more critically: p. 2804 last paragraph: What is northerly-southerly oriented airflow and how it is related to NE transport (title of the section)?

Response: We agree with referee 1 that the definitions of the trajectory clusters and directions of transport were inadequate. This has now been changed to follow general conventions on how wind directions are defined. That is, all clusters and trajectories are defined from the direction from where they arrive, e.g. SW-cluster, NE-cluster.

Specific comments

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1. Page 2788 lines 11-17: This part needs to be more clearly explained. Now it is difficult to follow which mode is coagulating with what and which mode number concentrations the authors refer to.

Response: This part was re-written in order to clarify the behaviour of the nuclei mode particles:

"Coagulation between nucleation mode particles and condensation of gases onto nuclei mode particles constitute growth mechanisms of nuclei mode into the Aitken mode size range. While this growth by condensation and self-coagulation of nuclei mode particles leads to increased number concentration in the Aitken mode, coagulation with larger particles might serve as a significant sink for newly formed nuclei mode particles. Transformation of nuclei mode particle mass into Aitken mode mass seems to largely be controlled by direct coagulation between nuclei mode particles and Aitken mode particles without observed growth of the actual nuclei mode. "

For clarity, we also inlcuded the size ranges used in this study for the definition of the nuclei, Aitken and accumulation mode particles under section 2.2:

"In this study the aerosol size distribution has been interpreted in terms of three modes, nuclei (<30nm), Aitken (30-110nm) and accumulation mode (110-1000nm). "

2. Page 2788 line 27: The authors should mention also the role of coarse mode.

Response: On line 26, p 2788, of the original manuscript following text was added: "Coarse mode particles may, if high enough concentration is present, serve as a significant sink of nucleation mode and Aitken mode particles. However, since coarse mode includes particles > 1μm (Seinfeldt and Pandis, 1998), no evaluation of this mode can be done due to instrumental limitations. The instruments used typically cover a size range from <10nm to 500nm "

3. All the symbols need to be defined. E.g. Dgs on page 2789.

Response: This have been changed accordingly

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4. Page 2793 lines 22-23: I disagree. There is substantial variation in number concentration especially during DJF and also MAM.

Response: The variation is at maximum 10% (p.2793, line 23). We do not agree with referee that 10% diurnal variation is to be considered substantial. Please note that the y-scale only cover number concentration from 1500-3000 cm^{-3}. In the figure caption of figure 45 (in revised manuscript) we added:

"Note that the axis scale for the number density begins at 1500 cm-3 for the large frames."

and

"Note that the axis scale for the number density begins at 500 cm^{-3} for the large frames."

5. Page 2794 lines 18-20: It is not obvious why diurnal variation results from local sources.

Response: On page 2794, line 19 we added:

"The nature of industry and household activities in the vicinity of the stations typically follows a diurnal pattern (e.g. lack of large industries with 24h production activity). Thus if local anthropogenic activities do affect the aerosol size distribution, this would be evident as diurnal variation of the aerosol number concentration. "

6. Page 2794 line 23: The authors state that the nucleation events occur at other stations as often as in Hyytiälä. This statement needs supporting data or reference.

Response: The statement refers to the other measurement stations involved in this study during the year of investigation. This fact is displayed by table 2 and also denoted in figure 7-10. It was not the the authors intention to make a general statement on this issue, neither extendable over a longer time-period nor representative for other stations

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than those included in the study.

"....e.g. Hyytiälä during this year of study. " Was added following line 24, p. 2794

8. Page 2795 line 10: Jansson -> Janson. By the way, is this the central reference to the nucleation classification?

Response: Reference changed to Mäkelä et al., 2000

9. Page 2795 lines 21-22 and Page 2796 lines 12-14: The nucleation in Aspvreten is highest in winter!

Response: We do not agree. Check with table 2 and figure 7. It is clearly stated that the frequency of nucleation events is largest during spring (MAM-period, 14%) as compared with wintertime (DJF-period, 2%). In order to clarify this even more we added: "At Aspvreten we observed the lowest frequency of nucleation events during winter and autumn (See table 3, DJF period) "

Further, on line 14, we refer to table 3 in the revised manuscript.

10. Page 2796 lines 1-3: I understood that nucleation is a natural phenomenon. If that is correct, how do the smelters in Kola Peninsula relate to nucleation? What do the authors mean by nucleation? How is nucleation defined?

Response: Nucleation as encountered at the different stations is described on p.2792, lines18-21. Further information was added under same section (3.1) in this revised copy of the manuscript. The industry located around Kola Peninsula was suggested to occasionally give contributions to nucleating gases or precursors thereof (e.g. SO2)

We clarify this on page 2796: "Easterly winds may bring elevated concentrations of nucleating gases or precursors thereof to the station from the smelters contributing to the observed nucleation. "

In defining nucleation we do not distinguish between precursors of natural or anthropogenic origin.

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11. Page 2796 lines 8-9: At minimum, the measurement period in Vavihill needs to be given (see also the general comment). Page 2796 line 12: Is the Vavihill data complete enough for such a statement?

Response: We have changed the discussion and excluded Vavihill from this general statement and instead introduced following text (l.7, p. 2796):

"Although sparse in data, Vavihill tends to show equally high or higher aerosol concentrations as compared with Aspvreten. "

Additional changes were included, highlighting the limitations of the dataset from Vavihill (l. 9, p. 2796):

"Data available from the stations only spans from February-April 2001. "

12. Page 2796 lines 18-20: There are several other potential explanations as well. E.g.role of temperature or height of the mixed layer.

Response: This is agreed with the referee, and further description of the nucleation phenomena was added under section 3.1. See general comment 3

13. Page 2797 line 5: Where does the continental influence come from to Värriö/ Aspvreten?

Response: We changed this statement as below:

"During the autumn period a large number trajectories arrive from S/SW. Thus, both stations are likely to experience the sources on the continent, changing the aerosol properties accordingly. "

14. Page 2797 line 8: What means decoupled in this context?

Response: Decoupled was a bad use of words and was replaced with:

"does not share the same advection pattern"

15. Page 2798 line 28: Does the size of mode refer to (mean) diameter or number

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(concentration) of the mode?

Response: This is clarified.

“The Aitken mode shows largest differences when comparing the geometrical mean diameter, Dg ”

16. Page 2800 line 5: What means "good statistical representation"? What is the total number? Are the situations simultaneous?

Response: We skipped this sentence since it did not contribute to the discussion, but instead appeared confusing.

17. Page 2801 line 1: Is Kulmala et al, 2001, the central reference for this?

Response: The issue is treated by Kulmala et al., 2001, but maybe Virkkula et al., 1995 is a better reference.

Both references are provided in the revised manuscript.

18. Page 2801 line 27: What are "anthropogenic properties"?

Response: The use of word anthropogenic in this context reflects our intention to clarify that we believe that we during this specific case resolve the heavy industries located at the Kola Peninsula. This since the type of size distribution encountered during this typical advection situation lacks counterparts in the rest of the data-set. It is however agreed that the use of the word anthropogenic properties probably is associated with more questions than answers. We thus replace this line with:

“likely affected by industries on Kola Peninsula. ”

19. Page 2802 lines 14-16: It is confusing that the authors speak about Aitken mode and Aitken mode2 right after each other. How is Aitken mode defined here in relation to Aitken mode2?

Response: See general comment 2.

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20. Page 2803 last paragraph: What is the time scale for transport between stations? Is deposition out of question in shaping aerosol size distributions?

Response: Deposition is not excluded, but by mistake left out in the discussion. The time scale for transport is 2-3 days. Changes have been incorporated accordingly.

"This should be related to the actual transport time between the southerly and northerly stations, on average 2-3 days derived from the trajectory analysis. "

And,

"The influence from dilution and dry deposition can neither be neglected. "

21. Page 2804 lines 10-11: Hyytiälä is actually bigger than Aspvreten.

Response: This is true, and discussion has bee changed accordingly.

22. Table 1: Why are these size ranges used? Is that based on literature or on actual data?

Response: A definition based on size for the nuclei-, Aitken-, accumulation-, or coarse-mode concept is not available. Differences can be seen in different studies and not least between disciplines. In contemporary literature on atmospheric aerosols the mode for coarse particles are typically larger than 1 micron, accumulation mode larger than 100 nm, Aitken mode larger than 20 nm, and Nuclei mode smaller than 20 nm. Strictly speaking, there are an infinite number of solutions for the automatic fitting routine unless the procedure is constrained. Thus, to initiate the calculations the routine is seeded with values within the ranges presented in Table 1. The choice of the ranges comes from the commonly used ranges as listed above, but also from the hands-on experience of manually fit thousands of distributions. The selected ranges have proven to give the best blend of subjective constraints and fast convergence to solutions by the fitting routine.

23. Table 2: Why is the comment "...and this does not necessarily cover for the

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whole season"?

Response: This is agreed to be confusing and we changed the text accordingly:

"Here given as % days of the data set exhibiting nucleation as described under section 3.1. "

24. Figure 14: What do the authors mean by "model" in figure caption? These figures would be better with expanded x-scale.

Response: " Model" is maybe not appropriate here. It is replaced by "fitting routine"

"The red crosses show results derived from the fitting routine. "

25. Figures 15 and 17: What means: "Each endpoint corresponds to approximately 5h. "?

Response: We try to clarify this by replacing the above sentence with:

"The time spacing between each pair of endpoints (dots) in the figure is 5h. "

Interactive comment on Atmos. Chem. Phys. Discuss., 3, 2783, 2003.