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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 referees' 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 2

This paper contains interesting material about aerosol size distribution measurements at various background stations in Scandinavia. The paper's data are unique, and it merits consideration for publication in ACP. Before publication, however, some more work needs to be done to make this an attractive paper.

The major criticisms are:

1. The data set spans 1 year, and also includes significant gaps for some stations. I believe it is not sound to suggest that these data are representative enough do discuss an annual variability unless supported by climatological information.

Response: One of the major criticisms is that the authors discuss the data in terms of annual cycles. A suggested solution would be inclusion of more data or supportive climatological information in order to determine if the year of study is representative or not. It should however be noted that the title of the paper clearly indicate that the study is limited to one year. It is not the authors' intention to suggest that this year is representative for a longer time period. The use of word season in this context might therefore prove misleading to a reader. In order to clarify and prevent any forthcoming misunderstandings the title of section 3.2, p. 2795 l.1 have been changed from

"Seasonal variation of aerosol properties"

to

"Variation of aerosol properties between the seasons of the year of measurement"

Similarly, title of section 3.3, p.2796, l.22

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“Seasonal variation of trajectory orientation”

was changed to

“Variation of trajectory orientation between the seasons of the year of measurement”

Further changes was made in the introduction p. 2785, l. 26

“i.e. find the seasonal variation for the whole Nordic region”

was replaced by

“i.e. investigate how the aerosol properties varies over a full year in the Nordic region”

Small additional changes were also included throughout the text in order to avoid misinterpretations of the purpose with this investigation.

We have further stated the limitations of the Vavihill dataset. The Pallas data set do admittedly contain some gaps. However, the closeness between Värriö and Pallas allows the Värriö data to support the Pallas data during the periods of missing data. Both stations are located on almost the same latitude and experience similar surroundings.

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

Was added on page 2796, line 14.

According to suggestions from referee 1, following text below was added to the manuscript:

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. ”

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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. Besides the presentation of various data, the paper is poor in actual scientific findings. The authors recognize the obvious importance of air mass types, pre- cipitation, solar radiation, and more meteorological factors, but fall short of meeting the many expectations aroused in a reader by the introduction.

Response: One of intentions of the authors is to give a description on how aerosol properties varies over the Nordic countries over one year of measurements and determine how long-distance transport affect the different receptor sites. According to the authors these goals where met in the article;

since this study is unique in the approach of linking different stations over a large geographical area over one year of measurements,

since this approach has not been used before, for aerosol size distribution data collected in the Nordic region, the article gain scientific qualities even though specific studies of different meteorological parameters are lacking.

On the issue of the clusters found in justifications below we have evaluated the typical surface pressure situation forming the different clusters and added an example of this.

Following paragraph was added under 3.4.1 in conjunction with a figure (17).

“As discussed in section 2.1, clusters will represent a typical synoptic weather situation. A comparison of similar clusters arriving different stations presents similar average pressure fields. In figure 17 average pressure fields are presented for the four stations involved in this case study. The averages were calculated over the four day duration of the individual trajectories over the DJF-period (figure 16b). The different pressure fields present striking similarities as can be seen in the strong gradient over the North Sea

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and British Isles. The slight differences between the stations arise from the fact that the averages are calculated based on different days due to moving weather systems. This put confidence in the clustering procedure and actually confirms the idea that we will experience a reduced bias from station specific meteorology when comparing the different locations. Sea level pressure data was obtained from the NCEP/NCAR archives. ”

We further evaluated the precipitation associated with different types of advections situations.

Section 3.4.2 “An analysis of the precipitation rate along the trajectories indicates up to four to five times more precipitation associated with SW clusters as compared with NE cluster, on average. In absolute numbers, the accumulated precipitation along SW trajectories between Hyytiälä and Värriö was found to be 2.6 mm on average. On the other hand, the accumulated precipitation along NE trajectories as in case 1 (3.4.1), was as low as 0.75. Wet deposition is well known to be a strong removal mechanism for especially accumulation mode, but also for Aitken mode particles. ”

It is naturally agreed with referee 2 that additional meteorological analysis would be interesting to a reader, but it is not realistic to include further information in this article since the material is quite extensive as it is.

Nucleation phenomena are a well-treated subject throughout recent publications. Especially the BIOFOR (Biogenic Aerosol Formation in the Boreal Forest) carried out in Hyytiälä 1998-1999 sprung out with several publications on this issue (Eg Kulmala et al., 2001, Aalto et al., 2001, Nilsson et al., 2000, Nilsson et al., 2001a, Nilsson et al., 2001b). It is not within the limits of this article to present investigation equally thorough as e.g. those given above. An interested reader would find more detailed information in the above given publications compared with a condensed material implemented in this article. Nucleation is an important phenomenon, but the ambition with this article is not to present a detailed study of the phenomena.

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However, in order to meet with referee 2 some statistics on the issue was added under section 3.1 as well as changes in the text as below:.

“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 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

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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"

Although the suggestion to perform a isolated study of the Kola Peninsula certainly would be interesting this is out of the scope of the present manuscript

3. I would like to see a more thorough motivation, i.e., why this kind of analysis is done and what can be accomplished later with the authorŠs data.

Response: We utilize a semi-Lagrangian approach for a large number of stations and for a long period of time. This has not been done before for the size distribution measurements in the Nordic countries. This is argued to be a necessary step for the development of a thorough understanding of the different processes affecting the aerosol size distribution. Especially the development of regional models such as the EMEP model is in great need of parameterized aerosol processes. This investigation serves as a first step in this direction. Further, the usefulness of the measurement network for this purpose is demonstrated in the article.

The conclusions were therefore extended to include these important aspects of the investigation:

"The measurement network seems ideal for evaluating the transport of suspended particulate matter. In the present study we have explored the possibilities of linking different measurement sites in space and time. We conclude from the study that a constant field cannot describe the aerosol over the Nordic countries. Instead, we revealed typical differences between the stations. These differences were argued to be transport related. We did not performed an in depth study of the influence from different meteorological situations associated with the different types of advection. However, the influence from precipitation was argued important for the lifetime of the aerosol. In the future this work of course has to be extended and a more detailed investigation of the role of meteorology and sources are called for. This investigation serves as a first step in this process of finding important factors affecting the aerosol over the Nordic coun-

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tries and to find and supply parameterizations of aerosol processes to be included in regional models. ”

Individual points:

p. 2792, l. 17 “No indications of local anthropogenic influence was found”. Please explain in more detail how this judgement was made.

Response: “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 anthropogenic activities do affect the aerosol size distribution, this would manifest as diurnal variation of the aerosol. ”

Was added under section 3.1

p. 2792, l. 20 “The frequency of the nucleation events has been found... ”. Does this statement refer to the data from this study? This is not clear.

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. However, the statement is valid for other investigations as well, e.g. Mäkelä et al., 2000. This was clarified by adding text as below on page 2792:

“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. Mäkelä et al., 2000, Nilsson et al., 2001). ”

Fig. 2: “Time dependent evolution”: It would be more illustrative to have nucleation days and non-nucleation days plotted separately.

Response: Changes have been added according to the comment by referee 2 In order to conserve the interesting features from this change we had to withdraw a paragraph from the text, namely:

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" The Hyytiälä size distribution data is clearly associated with a diurnal cycle concerning the shape of the size distribution. An elevated concentration of nuclei mode particles starts to appear during the morning hours (8-12) reaching a maximum concentration around noon. During the following time intervals on can observe a gradual increase in the number concentration of Aitken mode particles (16:00-20:00 and 20:00-24:00). The presence of a maximum of Aitken mode particles during night time and a minimum during morning hours till midday is behaviour found at all stations during springtime. The diurnal variability is most pronounced during the spring months, reflecting the larger frequency of new particle formation events as compared with other seasons. These features apply to all stations investigated. Smallest temporal variability in the shape of the size distribution is found for the winter period and no obvious pattern can be recognized at any station for this period. "

was replace with:

"The diurnal variability in the shape of the size distribution was investigated. In figure 3 the composite time dependent median size distributions for Hyytiälä during March-May are depicted. The data have been divided into 4 h intervals and calculated as the median of the integral of scans during the period of the year. The data was further divided into one sub-set with days with typical nucleation events and another with days when we did not observe nucleation events. Two features become obvious. First, a systematic diurnal variation is only observed for the sub-set with nucleation. In the case with nucleation events we experience this as an initial increase of small particle in the time interval between 08:00-12:00, even more pronounced during 12:00-16:00. Hereafter the size is shifted towards larger particles during time-steps 16:00-20:00 and 20:00-24:00. This behaviour could be interpreted as growth of the small, initially formed, particles. This since we believe that the nucleation phenomena can be observed over large areas simultaneously. We cannot observe a systematic diurnal variation in the sub-set with no nucleation. The second feature comparing the behaviour of the two data sets is the fact that the nucleation sub-set has obviously much less mass asso-

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ciated with the aerosol as compared with the non-nucleating cases (e.g. much larger concentration in the accumulation mode in non-nucleation cases as compared with nucleation cases). This probably reflects the fact that we observe nucleation when we have northerly advection, which is likely to bring rather clean air to the measurement sites. These typical features comparing statistics concerning non-nucleation days and nucleation days are found at all stations. That is, nucleation events do affect the size distribution in a diurnal fashion. This feature is lacking for non-nucleation days. ”

Further, we limit the picture to describe one station in the revised manuscript.

The conclusions were of course changed accordingly;

“An investigation of the diurnal variation of the size distribution shape was performed. It was found that during nucleation days we observed a characteristic diurnal variation in the nuclei and Aitken size range, with nuclei mode particles appearing in the morning hours. The size distribution is later during the day shifted towards larger size classes. We interpret this as a growth of the freshly formed nuclei mode particles. No diurnal variation in the shape of the size distribution was found for non-nucleation days. ”

Fig. 11: How is it possible to calculate running means during some periods of missing data?

Response: This is a justified point and has been changed accordingly.

Figs. 15-16: Change color coding, so that trajectory clusters and their corresponding size distributions bear the same color.

Response: This mistake has been adjusted

Fig. 16, 18: What is the justification of normalizing size distributions at different stations? This feature appears irritating to a reader, rather than illustrative because it suggests concentrations ratios between distributions that are not real.

Response: The purpose of the normalization was to highlight the differences in actual

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shape of the size distribution when evaluating transport related changes of the aerosol.

“Size distribution normalized to 1 are given in the right frame in order to highlight the changes in size distribution properties between the stations ”

Was added to the figure caption of figure 16 18.

Fig. 17: The nomenclature “NE oriented clusters” is contrary to convention, and intuition. The practical meteorologist labels winds after the direction from which they arrive. The clusters woule be better termed “Clusters arriving from SW”, or shortly “SW clusters”. For clarity, please highlight the measurement sites with an additional symbol in Fig. 17.

Response: This has been changed according to the refereeŠs comment. We now use a consistent terminology as example given by referee 2. E.g. SW-clusters.

Word choice: I find the term “Nordic” in the title unappropriate. The authors may correct me if they wish. “Nordic” (“Nordic combination”, “Nordic war”, “Nordic council”, “Nordic literature”) refers to the more cultural issues. Why not use “Scandinavian”, a proper geographic term, globally understood, which may also be used to include Finland?

Response: After consulting various reference literatures on the issue above, we maintain our stand point, that Scandinavia is not a proper term for the area of investigation since it not necessarily include Finland. Scandinavia may be used to include also Finland, but Nordic by definition includes Finland. That the “Nordic countries” is an accepted and established terminology is not least evident from the fact that this study is partly funded by The Nordic Council of Ministers (NMR). We therefore choose to keep the word Nordic countries in the title.

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