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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Analysis of particle size distribution changes between three measurement sites in Northern Scandinavia" *by* R. Väänänen et al.

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The authors thank the reviewer for the constructive remarks and suggestions. Below are our answers to the comments. Review's comments are marked by italics.

General comments
In the introduction part of the paper the author state that they aim to address the four following questions:
(i) are there fundamental aerosol dynamical differences between air masses entering the different stations, or in air transported between the different station pairs?

(ii) does the west-to-east air mass transport differ from the east-to-west transport in any observable way?
(iii) how fast do particles grow effectively in size during air mass transport and how does this differ from the growth observed during nucleation events at ïnËĞAËŻxed measurement sites?
(iv) what is the net-effect of aerosol source and sink processes on particle number concentrations during atmospheric transport?

First of all, are there any fundamental differences present (bullet(i)), and if so, please explain what they are.

We found no fundamental differences, as stated in the first paragraph of section 4. The main observed subtle differences are briefly summarized in the same paragraph.

Secondly, the authors state that the overall goal is to provide new insight in aerosol dynamical processes. This is a worthy undertaking, and certainly a relevant one. Unfortunately, in the current manuscript, the only processes that are discussed in any detail are in fact gas-to-particle conversion and nucleation. The role of primary emissions is completely left out of the discussion as is the importance of wet/dry deposition.

Aerosol growth and nucleation have been extensively discussed in the literature, and if "new insight in aerosol dynamical processes" is to be provided, the range of processes and sources presented in this paper needs to be extended. To reach these goals, the authors need to as a minimum at least provide a discussion of the importance of these processes, but ideally I recommend trying to find ways to use the data-set to extract relevant estimates of the same. e.g.: Why does the different approaches yield different results? What does it imply in terms of other sources and sinks?

I also urge the authors to complete the study of aerosol number size evolution relative time spent over land with also the winter time data. Currently, it seems illogical to present only summer time data for one method (i.e. time over land), and winter + summer time data for the other method ("Lagrangian type" approach). The argument

that winter time is deficient of biogenic emissions is insufficient. Perhaps a winter time analysis of time spent over land in fact will provide information of other sources than secondary biogenics which (as acknowledged by the authors) is at a minimum during winter time, bringing the study closer to its original goal.

As suggested by the reviewer, we have extended our analysis for the winter period (see our more detailed response below). The winter analysis will provide us with some insight into primary particle sources, and this will be added to the manuscript. The problem remains, however, that the primary sources will likely to be different between the summer and winter. Our analysis gives the net effect of aerosol sources and sinks during the air mass transport, so it is almost impossible to get any separate information on aerosol deposition processes. We will, however, discuss these issues in more detail as suggested by the reviewer.

When comparing transport between the stations, the authors needs to show that the air flow in fact is connected, not only in terms of Lat-Long, but also wrt altitude. If this is not done already, I do suggest that the authors confirm that the trajectory is within the mixing layer (or at least below some reasonable altitude) at the upstream station. If the air resides at a very high altitude over the upstream station, the data from downstream and upstream stations may in fact not be connected at all, which could explain some of the behaviour.

We checked this out. The fractions of considered trajectories that are within the mixing layer at the upwind station (during the transport from upwind to downwind station) are the following: Abisko -> Pallas: 97 (96) %, Pallas -> Abisko: 87 (81) %, Pallas -> Värriö: 94 (94) %, Värriö -> Pallas: 98 (97) %. We therefore conclude in the vast majority of cases, the air flow was connected also vertically.

Furthermore, do the authors take into account the altitude of the trajectories when estimating time over land? Must trajectories start over ocean in order to qualify for further analysis? How big fraction of trajectories (=transport events) meet the criteria

above?

We required that trajectories must start from over the ocean, but their initial altitude over the ocean was not limited because the only initial requirement was that the original air masses should unlikely to be polluted. The total fractions of air mass trajectories that passed our requirements were 81% for Abisko, 75% for Pallas and 58% for Värriö.

The authors also needs to better harmonize the agreement between figures and text. This is especially true for section 3.2, which I occasionally find contradicting. The language needs, at least in parts, improvement and proof reading by native speaker is recommended.

We will rewrite part of the analysis in section 3.2 to make the text more consistent with figures. The language will be checked out carefully.

Specific comments Page 9410, line 2-3: The authors need to specify what they, mean with "continent" in this context. Are all transport directions considered in the analysis? According to figure 1 it seems so that also southerly air flow is accepted in the analysis (sector defined by the red lines). The authors need to better explain how the trajectories are selected; should the whole trajectory be within these sectors? Should the trajectory always start over the ocean? etc. What I want to have explained is whether or not trajectories coming from areas south of the horizontal red line is included in the analysis as well

We will clarify the selection of trajectories in the new subsection 2.2.4. In short, each 96-h trajectory had to start from over the ocean, it hads to spend >90 percent of its transport time in the sector defined in Figure 1, and "time spent over the continent" is the total time that this trajectory spends over the continental area (not over the ocean) during its last 96 hours of transport time.

Page 9410, line 8-10: The authors state that only the (extended) summer period is investigated since this is the time of year when biogenic emissions are highest. This is

true if the authors want to study biogenic alone. However, as stated in the introduction, the goal of the study is somewhat broader and according to bullets i-iv it seems to be oriented towards growth, transformation and deposition in general. Therefore I find it surprising that the winter period is excluded from the analysis. I do strongly suggest that the authors also comment on the statistics that can be derived for the winter period as well. If a growth pattern is evident or if number concentration is increasing during this period as well, the conclusions presented has to be somewhat modified. Similarly, if the winter period is lacking the features of the summer period, the current conclusions can be strengthened.

As suggested by the reviewer, we have extended our analysis for the winter period. We will divide section 3.1 into two parts: "3.1.1 Summer period" and "3.1.2 Winter period". Our focus will remain in the summer period. The main the findings from the winter period will be discussed shortly in section 3.1.2 with couple of additional figures, and the potential consequences of these findings will be taken into account elsewhere in the text.

Page 9411, line 16: ". . .into logarithmically spaced size bin basis. . ." needs rephrasing.

Page 9411, line 18: dLogDp to dlogDp

We will rephrase this sentence into the following form: "Measured particle number size distributions were mapped into the size bins dN/dlogDp, where dN is the particle number concentration in each size bin and dlogDp is the logarithm of the width of the size bin."

Page 9412, line 1: ". . .method suited. . ." to ". . .method is suited. . ."

OK, corrected.

Page 9412, line 3: ". . .3-6 clusters. . .", Is it so that the number of clusters was selected a priori; if so, on what is this selection based?

Page 9411, Section 2.2.3: Whole section need to be improved language-wise. It is not completely clear how the trajectory/transport selection was performed here. Is the data clustered prior identification of connecting trajectories or v.v.?

We will rewrite part of the text in section 2.2.3. More specifically, we will clarify how the clustering was made and which clusters were selected for the more detailed analysis presented in section 3.2. The language will be checked out throughout the paper.

Page 9412, line 19: Please expand on how the terrain affects the emissions. Do the authors perhaps mean land use type?

The terrain, i.e. land use type certainly affects emissions, but since this information was not used in our analyses, we will remove the sentence from the text.

Page 9412, line 20: ". . .67.1–69.0 degrees. . ." Why this corridor? 67.1 seems specific. . .is it based on actual variability of land use type in the corridor? What landuse types dominates in this corridor? Are there any centers of population that could affect aerosol properties during transport?

The corridor was chosen to exclude the airmasses that do very wide loops twhen travelling between two measurement sites. The towns inside the corridor include Sodankylä and mine town Kiruna. The distance from Kiruna to Abisko is around 90 km. We will calculate the fraction of trajectories between Abisko and Pallas of which the air masses travelled also above Kiruna to see if it can affect the aerosol properties. The main land use type in this section is boreal forest.

Page 9414, line 14: "After that, there was a small drop. . ." The authors must mean drop in "rate of increase" as no drop in number concentration is evident. Can the authors also comment on the fact that there seem to be two distinct rates of number increase; one for low time over land, which is fast, and secondly one slower rate of increase that for air-masses that spent longer time over land. At the same time, the mass increase is linear throughout the full range of time spent over land. Can the

authors comment/hypothesize on the reasons for this behavior? Why does the rapid increase stop at 25-30h over land. Of course the increasing CS will quench further new particle formation, but I do not see any obvious reasons that this should generate two distinct slopes. Could in fact be so that the slow rate of number increase reflect primary emissions and the rapid slope reflect the composite of nucleation+primaries??

Page 9415, line 4-5: ". . . a time period when the number concentration dropped although the particle mode diameter was growing. . ." Again, I can't see this drop in figure 5.

We agree with the reviewer in that claiming that there is a drop in the particle number concentration at certain transport time is an overstatement without sound statistical basis. We also agree that one should rather claim that the rate at which particle number concentrations increase with the transport time clearly decreases at some point. We will discuss this issue in more detail in the revised text and modify the two paragraphs (line 11 on page 9414 to line line 7 on page 9415) accordingly.

Page 9418, line 15-17: This statement seems highly subjective. What is "best clustering" in this context?? The authors must do a better job in rationalizing the selected number of clusters. I also recommend the authors to present the total number of transport cases between the different stations in relation to the number of cases used.

As mentioned above, we will clarify how the clustering was made and which clusters were selected for the more detailed analysis.

Page 9419, line 19-20: "During wintertime, the accumulation mode was very weak already in Pallas and did not change much." I do not agree. For cluster 2 the accumulation mode is definitely not weak and there is a substantial increase in aerosol mass during transport. For cluster 1 the accumulation mode might be considered weak, but its concentration during transport more than doubles. I also ask the authors to calculate the change in mass and from this change in mass estimate how much condensable gases would be required to sustain this growth. How does derived values relate

to observations? Furthermore, could the increase suggest something with respect to primary emissions in the sector? Other sources of condensable gases than biogenics? (Given that the photochemical production would be low during winter)

Page 9419, line 20-22: "In summer, the general modal dynamics was quite similar to that associated with the Abisko-Pallas transport route" This is true for cluster 1, but what about cluster 2? What can be the cause of differences?

In the original text, we were a bit unclear in separating between the most frequent cluster (cluster 1) and next most frequent cluster (cluster 2). This seems to have caused confusion and will be corrected in the revised manuscript. Currently, the only way to estimate the total amount of condensable gases in a field experiment is to derive it from the observed particle growth rates. As a result, we cannot make the suggested comparison. We will, however, add some text in which we will compare the shifts in mode diameter to the apparent particle growth rates observed at each station (reported in section 3.1) and to the real growth rates observed during new particle formation events. We will add a similar comparison for particle formation rates and discuss this issue in terms of primary and secondary particle formation.

Page 9420, line 5-6: "Also, although the summer data were best fitted using four clusters.." Again, I do not understand the reasoning behind the selection of number of clusters. Sum of errors? Or what?

Again, we will clarify how the clustering was made and which clusters were selected for the more detailed analysis.

Page 9420, line 10-12: "On average, these peak diameters remained relatively unchanged when the air masses travelled to Abisko, and the total concentrations on all size ranges increased. For all the modes, particle number concentrations increased at the rate of 0.002cm". What processes could be responsible of such behavior?

There in no single process that would cause this kind of observation (dilution would do

the opposite), so we cannot really propose a plausible explanation.

Page 9422, line 11-13: "The highest decreases in the accumulation mode concentration were seen between Pallas and Varrio for both directions during the summer time." Only one of the four clusters associated with transport between Varrio and Pallas (in either direction) show a decrease in accumulation mode concentration that can be considered significant.

We will rewrite this paragraph and reformulate the point raised by the reviewer.

Page 9422, line 14: "place" is probably not the best word. Size or mode is better.

We will use the term "mean diameter of the mode" instead of "place of the mode".

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 9401, 2013.