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

Anonymous Referee #1

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Summary of article

This paper explores how observed aerosol number size distributions changes both as a function of time over land as well as changes in the aerosol number size distributions that can be observed during air mass transport events that connects two different stations. The study utilizes number size distribution observations from 3 different sites (Värriö and Pallas in Finland and Abisko in Sweden). Trajectory analysis is the main tool in identifying inter-connecting transport events as well as in deriving statistics with respect to time spent over land.

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Particle growth rates and particle formation rates derived using three different methods (“time over land”, “lagrangian”, and “event analysis” are compared and differences are discussed.

The paper is generally well written, although language needs to be improved. Scientifically, the content of the paper is sufficient to merit publication, but only after the authors have addressed the issues/questions raised below.

Recommendation

Conditionally accepted after revisions and language improvements.

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 fixed 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.

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. Un-

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fortunately, 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.

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.

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

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above?

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.

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

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. Therefor 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.

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

Page 9411, line 18: dLogDp to dlogDp

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

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.?

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

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 land-use types dominates in this corridor? Are there any centers of population that could affect aerosol properties during transport?

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.

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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.

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?

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?

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?

Page 9422, line 11-13: The highest decreases in the accumulation mode concentration were seen between Pallas and Varrío for both directions during the summer time. Only one of the four clusters associated with transport between Varrío and Pallas (in either

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direction) show a decrease in accumulation mode concentration that can be considered significant.

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

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