

Interactive comment on “Aerosol formation over the Boreal forest in Hyytiälä, Finland: monthly frequency and annual cycles – the roles of air mass characteristics and synoptic scale meteorology” by E. D. Nilsson and M. Kulmala

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1 General comments

The paper addresses a relevant subject of atmospheric chemistry and physics, i. e., the influence of air mass characteristics and synoptic scale meteorology on new particle formation over a boreal forest. Compared to long-term observations of meteorological parameters, corresponding observations of aerosol properties are more expensive

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and hence, still relatively scarce. Therefore, investigations like the present one provide a valuable contribution to fill existing gaps in data conditioning and evaluation to generalise the physical interrelation behind the fragmentary observations.

The present paper is a continuation of former synoptical investigations and long-term statistical analyses of aerosol parameters, among others performed by both authors. Because of the impossibility to extract full system information from a single dataset (“random sample”) the evaluation of different datasets and the comparison of more recent results with those obtained from former studies is of scientific use. With respect to the present study it might be instructive to have a “metareview” of relevant previous long-term analyses to see, where we are.

The Hyttiälä time series is one of the most comprehensive aerosol datasets, but some of the definitely interesting conclusions drawn in the paper can not be derived from the presented data and must necessarily remain “speculative”. However, as long as these speculations are not in contradiction with the observations, they should be accepted as justifiable working hypotheses. The authors discussed some potential climate feedback effects, in which synoptic waves play a key role. It should require much effort to identify corresponding signals from field observations. This task is interesting and relevant enough to be tackled in future studies.

Andreas Stohl gave a conclusive comment, in which he promotes the new conveyor belt model at the expense of the traditional cyclone model shown in Fig. 10, which referee 3 recommended to completely remove. Considering the scale and question of interest I will give some arguments for retaining the classical Norwegian cyclone model, regardless of its known deficits. However, it seems to me, that the problem is not the adopted cyclone model, but the low probability for European (!) air pollution to enter the polar dome by transport along isentropic surfaces. In this context, the Arctic Haze formation deserves a reconsideration with modification of Fig.10. Some hints will be given. The idea to link aerosol formation processes with synoptic scale waves, however, is sound and worth to be published.

I recommend the editor to accept the manuscript for publication in ACP after the revision recommended below.

2 Specific comments

- With respect to former papers of Nilsson et al. (2001), Boy et al. (2002, 2003, 2004) and Hyvönen et al. (2005) addressing a similar subject, the authors should consider to include a **table** to briefly characterise the datasets used in these studies (e.g., location, measurement period, measured values), synthetic parameters (e.g., nucleation probability) and the main parameter correlations (e.g., $NPF \sim 1/CS$, $NPF \sim 1/RH$). A corresponding “matrix” would certainly allow a better overview and classification/subsumption of the results of the present and future studies on this subject. In this way, the present study could be better marked off from the former ones, and perhaps, some of the requirements raised by referee 3 could be met.

Boy and Kulmala (2002, Eq.(1)) and Hyvönen et al. (2005, Eq.(1)) presented an instructive empirical nucleation parameter P_{nuc} as a function of UV-A radiation, temperature, condensation sink and relative humidity. Along the dependence of the nucleation parameter on water vapour contains a very important information, from which basic conclusions regarding the nucleation scenario (“inorganic” vs. “organic” nucleation mechanism) could be drawn. Hence, the verification/falsification of the anticorrelation $P_{nuc} \sim 1/RH$ by means of the present dataset, even in a qualitative manner, would be of high benefit.

However, for principle reasons I would vote for not to “overload” this empirical study on the **mesoscale** by very specific questions, e.g., regarding the nucleation theory or the underlying mechanism, which are related to processes on the **Angstrom scale**. It is commonly known, that there are several very tricky problems which must remain on the agenda for solution, some of them have been

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raised by referee 3. But having in mind the title of the paper, the focus of the study should remain restricted to the “role of air mass characteristics and synoptic scale meteorology” for new particle formation. Otherwise, I am afraid, the paper will get frazzled. I see the present paper as a further step to a future comprehensive synthesis on aerosol formation characteristics from long-term statistics.

- Subsection 4.2

1. Conveyor belt model vs. classical Norwegian model:

The cyclone model adopted in Fig.10 is essentially based on the polar front theory of Bjerknes (1920) and the subsequent air mass doctrine of Bergeron (1925), which itself was a comeback of the former air mass concept of Dove (1840). It was a milestone in weather prediction and Raethjen (1953) argued, that the “Norwegian meteorological school” can claim the merits to have importantly enhanced the wave theory and to have widened its practical application. Up to date, the kinematic wave approach is an useful means for front analysis in all weather services around the globe. Nevertheless, soon after the use of satellite data it became clear, that this idealised theory can not be observed in every step and detail in reality, e.g., the traditional perception of the occlusion by overtaking of the warm front by the cold front with very narrow warm sectors can not be observed. This was one motivation to come up with the semi-Lagrangian conveyor belt theory. It is not a completely new model, but allows a more general description and a larger variety of development possibilities. Several investigations of Stohl et al. demonstrated its capability, e.g., in explaining the transatlantic transport of air pollution. For example, atmospheric boundary layer (ABL) air exported by warm conveyor belts (WCBs) can perform long-range transport over continental scales (Stohl, 2001). In the analysis of Stohl (2001), the WCBs were considered to start **above 500 m** and to ascent of more than 8000 m.

In an Eulerian reference system, the change of air mass properties at a certain location **near the surface** is mainly controlled by horizontal translation of the cyclonic system. As the present analysis is essentially based on **surface measurements** and air mass characteristics from routine weather observations unlike middle/upper tropospheric air flows, satellite-observed warm front cloudiness etc., the Norwegian cyclone model is certainly a good enough approximation to describe the feedback outlined in subsection 4.2. The conveyor belt model is the more advanced one, but the Norwegian model is sufficient with respect to the question of interest. It can be considered as a special case of the conveyor belt theory (by the way, a similar perception was adopted by Tunved et al. (2005)).

A short discussion/reference in the text might be helpful to solve this “conflict”.

2. Arctic Haze phenomenon:

Following Stohl (2006), surfaces of constant potential temperature (isentropics) form closed domes over the Arctic, with minimum values in the Arctic boundary layer. This leads to an isolation of the Arctic lower troposphere from the rest of the atmosphere by a transport barrier called “Arctic front”. In the Arctic in winter and spring surface temperatures become extremely low, stratification becomes thermally very stable with frequent and persistent near-surface inversions. The strong reduction of turbulent exchange and wet deposition owing to dryness favour very long aerosol lifetimes in the Arctic winter. To realise a transport of air pollution along isentropics toward the Arctic, the source region must have the same low potential temperatures as the Arctic air mass. Potentially warmer air can not penetrate into the dome, but will be lifted. Therefore, as Stohl argued, northern Eurasia remains the main source region for Arctic Haze (location of the Arctic front over Eurasia at 40°N, diabatic-cooling of air traveling over snow-covered land, transport into the dome).

Later on, Stohl (2006) proposed three different pathways along which air pollution can be transported into the Arctic: (1) rapid low-level transport into the Arctic followed by uplift at the Arctic front when and where it is located far north (possible transport from densely populated regions in Europe and from boreal forest fire regions, deposition of aerosols and water-soluble pollutants in the Arctic), (2) low-level transport of already cold air into the polar dome, associated with further diabatic surface cooling (transport of highly polluted air masses to the Arctic, low moisture content of the flow, stable stratification, pathway from European and high-latitude Asian sources over snow-covered high-latitude Siberia, absent in summer), (3) ascent of air masses south of the Arctic followed by high-altitude transport or by several upward/downward transport cycles, slow descent into the polar dome due to radiational cooling (frequent transport route from North America/ east Asia, less frequent from Europe).

However, Stohl (2006) argued that in the literature the role of transport for causing winter-time Arctic Haze is discussed ambiguously. In view of the complicated picture, this is not surprising. In any case, the Arctic Haze phenomenon is controlled by both transport and removal processes. With respect to the present analysis it follows, that – even not being favoured – air pollution transport from European source regions to the Arctic is possible. The authors should refine the picture given in Fig. 10 by considering these findings on Arctic Haze (e.g., overlay of conveyor belts in the figure). Nonetheless, in view of still existing deficits in process understanding, any picture must be accepted to remain a preliminary one for the time being.

- Subsection 4.3:

Potential climate feedback: Cold air outbreak together with enhanced UV radiation (corresponding to low cloudiness) \Rightarrow new particle formation (NPF) \Rightarrow

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enhancement of CCN formation \Rightarrow enhancement of cloud albedo \Rightarrow conservation of air mass characteristics toward more southern latitudes

Positive feedback: Global warming \Rightarrow decrease of the frequency of cold air mass outbreaks \Rightarrow decrease of NPF frequency \Rightarrow decrease of CCN formation \Rightarrow decrease of cold air mass conservation \Rightarrow further warming

If such a signal would exist, on which scale do you expect it to be soonest detectable? I suppose, the challenge to identify such a signal is comparable to the search for, e.g., the impact of cosmic galactic rays on the cloud evolution via ion-induced nucleation etc. Are you aware of corresponding datasets, which would allow such an analysis? Does your dataset allow such an investigation on the regional scale? Can you sketch a “design” of a corresponding data evaluation study or long-term observation program (what is urgently needed to bring the answer a step forward)? These questions are not deserved to be discussed in the manuscript.

- p.10443, line 21: A semantical and philosophical problem (not to be responded by the authors):

It is not “unrealistic” to study the “whole system”, but to resolve all spatio-temporal scales of the system. As you know, a whole system can be described by a simple box model, e.g., by an equilibrium equation for some global mean-state parameters as in the “daisyworld” model of the Gaia hypothesis. The abstraction/generalisation of a complex system including the resolved scales and the scale interactions depend on the question of interest (a model is an abstraction with advising power to reality) . When we wish to know, how large the effects of your hypothesised feedback can be on the regional scale, a regional scale model must be upgraded with respect to the description of the nucleation/activation mechanisms etc. This will become possible, sooner or later. The search for the

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effect on the global scale is again another question. In general I think, the search for corresponding signals from model-generated datasets is not hopeless, but a promising endeavour, sooner or later. Such an approach might be instructive to answer the question, which quality and quantity of observations do we need in future, e.g., to complete existing datasets and/or to avoid expensive and gigantic data “graveyards”.

A hypothesis, such as presented here, is more than a “speculation” and provides always a good entry to push/initiate corresponding investigations in future (such as CLAW hypothesis, forest-climate feedback, activation theory etc.).

3 Technical corrections

3.1 Text

- p.10428, line 4: Boy et al. (2006) demonstrated a good agreement of their predicted on- and offset of new particle formation as well as the total aerosol number concentrations with observations. I would like you to add this reference.
- p.10438, line 4: “we believe we have found”. Perhaps, the situation is better captured by “From the current study we suspect/ suppose/ suggest/ hypothesise/ propose etc.”. It is less than “we have found” but more than “we believe”.
- Please unify air mass annotations in the text, in Tables 1, 2, 3 and Figs. 3, 5, e.g., middle latitude vs. mid-latitude
- “feedback” (10426/27, 10439/1, 10439/7, 10443/19, 10444/6), compare it with spelling on p.10440, line 1
- p.10427, line 5: “nanometer sized”

- p.10427, line 18: add closing bracket
- p.10429, line 3: complete institution name: “Institut für Meteorologie der Freien Universität Berlin”; please add the number of the weather chart from 1997 (date). The air mass classification scheme is not included in each number.
- “Heated sub-Polar air mass (Ps)” refers to the German “gemäßigte Polarluft”. Perhaps, it is more appropriately translated by “temperate”, “tempered” or “moderate sub-Polar air mass”.
- Capitalization vs. use of small letters: “sub-Polar” but “Subtropical” but again “tropical” (10429/7, several times)?
- p.10430, lines 5-11: Make the reference to your four categories of aerosol formation events compatible with your subsequent enumeration, i.e., 1) . . . 4).
- p.10432, line 8: The latitude must be 62° N (cf. location data on p. 10427, line 13)
- p.10432, line 27: Is there a more appropriate term instead of “disintegration” (terpene ozonolysis, photochemical dissociation)?
- p.10434, line 8-12: The second half sentence is hard to understand. Please clarify the message.
- Please unify: “Northern Europe” (10435/2), but “northern Europe” (10426/7, 10428/6, 10443/17, 10461/Fig.10), “northern Finland” (10428/8), “central Europe” (10428/14, 10434/22, 10435/10, 10435/14, 10443/22), “eastern Europe” (10434/22, 10435/14)
- p.10435, lines 19-20: “we go ... when going” (avoid double use of verb)
- p.10436, line 9: “in the West and North”

- p.10436, line 11: "... air favours nucleation"
- p.10436, line 14: "in the South and East"
- p.10436, line 18: "the key parameter appears"
- p.10443, line 7, first half sentence: Perhaps, you do not have intended a "double negation" here.
(a) "This does not exclude that other major factors are involved." (b) "This does not mean that other major factors are not involved."
- p.10443, line 14: "None", "no one"
- p.10443, line 22: "recent observations ... demonstrate/demonstrated"
- p.10443, line 23: "may even be able",
"actually ... even" instead of "even ... even"

3.2 References

- Please add the number of the "Berliner Wetterkarte" from 1997 you are referring to.

3.3 Figures

- Figs. 3, 4, 5: Please unify annotation/spelling throughout the text: sub-Polar (P), heated/moderate(?) Polar (Ps), mid-latitudes (Sp), Subtropic (S) ... (e.g., in the text sub-Polar (P), in the figures Polar (P))

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- To avoid possible misinterpretation of Figs. 4 and 5 I recommend to clarify the following: The frequency axis represents the “scale” for the fractional frequencies of the different air mass classes, given by the width of the coloured areas, but not the “ordinate” for the single graphs starting at the zero frequency. The sum of the different fractions must be lower/equal than 100 percent.
- Fig. 6: In the caption: The longitude of Hyytiälä must be 25°E (cf. p.10427, line 13)
- Fig. 9: Compare style (letter size) with Fig. 8

3.4 Tables

- ./.

References

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Boy, M., Rannik, Ü., Lehtinen, K. E. J., Tarvainen, V., Hakola, H. and M. Kulmala: Nucleation events in the continental boundary layer: Long-term statistical analyses of the aerosol relevant characteristics, *J. Geophys. Res.*, 108, D21, 4667, doi:10.1029/2003JD003838, 2003.

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