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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #3

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General Comments

Nilsson and Kulmala present an overview of ground-based measurements of secondary aerosol formation at Hyytiala, Finland from 1996-2000. Previously publications have documented the same phenomenon [Kulmala et al., 2001; Nilsson et al., 2001a] and also attribute it to airmass character. The authors indicate that the current work is intended to "extend the analysis [of secondary aerosol formation] in time rather than space". Certainly time-series analysis of secondary aerosol formation is critical to our understanding of atmospheric aerosol processes. I am curious as to why this analysis

does not include time series data from 2001-2005. Is this data available? If so the authors should include it in this manuscript in order to address inter-annual variability. I feel that the current manuscript lacks a quantitative evaluation of the importance each variable, SO2, UVB, CS, Temp, and RH, has on the nucleation events. Including an analysis of this type and comparing it with existing nucleation theory would be an exciting contribution to our understanding of these events.

In its present form I do not think the manuscript is suitable for publication in AC&P without major revisions.

Specific Comments

Abstract

Don't, "speculate". Please quantify "strong emissions" and "strong boundary layer dynamics". What is your criteria for this statement?

The abstract states: "The critical factor that determined if aerosol formation would start on a day with Arctic air was the UV-B radiation". Can the UV-B (W mˆ-2) threshold for the onset of such an event be determined? How does it vary with available SO2, CS etc.?

With regard to the time series nature of the publication, Figure 2 shows that 1999 and 2000 were somewhat anomalous compared to the previous 3 years. What is the nature of this difference? Is the difference related to ENSO, the North Atlantic Oscillation, or inter-annual variability? If available, including data from 2001-2005 would double the length of the time series and may put these two anomalous years better into context.

I feel that the summary in Table 3 is not particularly quantitative. Since these conclusions stem from Tables 1 & 2 I will focus some discussion on their interpretation.

For Arctic air, certainly there is a large (-19%) reduction of CS in the late morning (9:00-12:00) compared to the early morning (6:00-9:00). But for P, and Ps are differences (-8% and -9%) significant compared to the CS reductions observed in non-nucleation

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airmasses (P=-5% and Ps=-7%)?

Arctic Air (A) -Here I agree with the authors, new particle production seems to be driven by clear conditions (UVB +40%) and a 23% drop in CS -it is not clear whether the 30% lower SO2 affects the probability of a nucleation event. Are there formation events where SO2 and CS are elevated or where UV-B is low? This might indicate conditions where SO2 was the controlling variable not CS or UVB.

Polar Air (P) -I understand “transition Polar” to mean maritime Polar air becoming continental in nature. Is this interpretation is correct? -Is there additional information on the thermodynamic character of the mixing airmasses during the nucleation events? For example is warm wet nocturnal boundary layer air mixing with cool dry free troposphere air as the boundary layer is eroded in the early morning for each case? Could a more quantitative assessment of the importance of temperature and relative humidity during nucleation events be included in the manuscript?

Can the interpretation of the Ps nucleation events be extended? In Table 2, CS during aerosol formation days for P and Ps air types are very close. SO2 is much higher +54% for Ps than P events, but UVB is also higher for Ps (0.58*0.90=0.53 Wm-2) than for P (0.33*1.44=0.47 Wm-2) events. Is SO2 or UV-B the controlling variable? I thought formation events were better correlated with the onset of turbulence than with increase in UV-B [Nilsson et al., 2001b]?

What does the variability between available SO2, UV-B and particle growth rates tell us about the intensity of the events in the different airmasses? What is their effect on the regional (global) aerosol fields, i.e. how much CS are they contributing? It seems that there is sufficient data to address some of these questions. An analysis of this type should be included.

Figure 10 should be removed as it does not accurately reflect the advection of airmasses into and out of the Arctic.

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The above comments lead to a larger point about the manuscript in general. While the authors have compiled an impressive amount of meteorological data that allows them to classify the events, I feel the manuscript lacks a quantitative assessment of the degree of control that each variable (SO2, UVB, CS, Temp, and RH) have on the events themselves.

While H2SO4 and NH3 were not measured continuously, can BIOFOR or other data be used to determine whether the events likely exceed binary and/or ternary nucleation thresholds such as [Napari et al., 2002; Vehkamäki et al., 2002]. Can the authors assess the degree to which the existing nucleation theory can explain the results?

Can condensation rates be estimated as a function of the controlling variables? If so are the, measured? (line 8-10, page 10443), predicted?, reservoirs of H2SO4, and NH3 sufficient to account for these growth rates or are seasonally higher VOC's a more likely candidate for growth beyond critical cluster sizes?

Are rates of growth in the summer, when there is a contribution from boreal VOC's, different than winter when regional SO2 (from fuel use) is higher? What about differences between controlling variables as a function of seasonal airmasses type?

While Figures 8 & 9 are informative and clearly show that the authors are moving towards predictability for this system, they have not addressed some larger, potentially more important issues with their data. For example:

What is the relative contribution of these newly formed secondary aerosols to the background population? Since pre-existing aerosol surface area is low how significant a source of new CCN are these secondary aerosols, 3%, 10%, 90% of total number.

To what extent are the events linked to anthropogenic emissions? i.e., new particle production and/or condensation growth in spring could be due to anthropogenic emissions. But in summer, if it is driven by boreal forest emissions then its impact with respect to the feedback shown in Fig. 11 is no different than pre-industrial conditions

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(ignoring deforestation).

Summary

While the authors have compiled an impressive time-series of aerosol data and linked it to the synoptic scale meteorology I do not feel that there is substantial new information that warrants publication in the manuscript's current form. If possible I suggest including data from 2001-2005 in order to address issues related to inter-annual variability. Second, while figures 8 and 9 and tables 1 & 2 are valuable I feel the remainder of the publication lacks a quantitative link between the frequency and intensity of the events. Obviously not all measurements will be available for the entire time series but the authors should endeavour to investigate and/or extrapolate based on the available data in order to address the following two items:

1) Is there a functional relationship between the new aerosol number being generated and the controlling variables, UVB, CS, SO2, Temp, RH etc. Including this even for the Arctic airmass type would be very informative. 2) For the varying intensities of newly formed secondary aerosol number in the airmasses (at least for the Arctic outbreaks), what is the effect on the regional airmass's total aerosol number, i.e. potential to provide new CCN?

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Interactive comment on Atmos. Chem. Phys. Discuss., 6, 10425, 2006.