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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Concentrations and fluxes of aerosol particles during the LAPBIAT measurement campaign in Värriö field station" *by* T. M. Ruuskanen et al.

T. M. Ruuskanen et al.

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General comments

However, while the individual measurements are good, their reporting in this paper remains rather descriptive....

We agree with referee's comment and we have changed our conclusions correspondingly. Now we have written the section" Summary and conclusions" in integrative way showing how different observational techniques contribute to our main results.

However, I have some conceptual problems with notion that more polluted air masses

switched off the nucleation event...

We have changed the parts where we are talking about switching off. It is true that the event does not break in reality. We just don't see the particles any more at the site.

In abstract: "In this paper we describe the measurement campaign, concentrations and fluxes of aerosol particles, air ions and trace gases, paying special attention to an aerosol particle formation event broken by a air mass change from a clean Arctic air mass with new particle formation to polluted one approaching from industrial areas of Kola Peninsula, Russia, without new particle formation."

In Particle formation events: "This sudden change happened because of the change in the air mass demonstrated in the chapter 3.7"

Conclusions: "As the route changed, a strong anthropogenic influence was observed, and the observation of the new particle formation event at the site was interrupted due to the change of the air mass with high coarse aerosol particle and inorganic trace gas concentrations."

Scientific Comments

p714, l12. Could the authors please add references to the PHAUCPC and LICPC which are non-standard instruments?

We have added three references

O'Dowd C. D., Aalto P. P., Yoon Y. J. and Hämeri, K.: The use of the pulse height analyser ultrafine condensation particle counter (PHA-UCPC) technique applied to sizing of nucleation mode particles of differing chemical composition, Journal of Aerosol Science, 35, 205–216, 2004.

Marti, J. J., Weber, R. J., Saros, M. T. and McMurry, P H.: Modification of the TSI

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3025 condensation particle counter for pulse height analysis, Aerosal Science and Technology, 25, 214-218, 1996.

Mordas, M., Kulmala, M., Petäjä, T., Aalto, P. P., Matulevičius, V., Grigoraitis, V., Ulevičius, V., Grauslys, V., Ukkonen, A. and Hämeri, K.: Design and performance characteristics of a condensation particle counter UF-02proto, Boreal Env. Res., 10, 543-552, 2005.

P715. l15-l21. A laminar flow through a 4.4 m long tube should induce significant flux loss, which should be estimated. Clearly, this has no effect on the direction of the flux, but on the magnitude.

The eddy covariance aerosol fluxes are in this manuscript only used to demonstrate periods of downward flux caused by either pollution plumes or nucleation events. Because the exact magnitude of the flux is not of interest, we have not corrected the fluxes for flux losses or particle losses in the sampling tube.

P718. l10. The under-counting of the APS 3320 for large particles is well documented. However, if the APS was calibrated against the MOUDI gravimetric measurements, the disagreement between calibrated MOUDI measurements and EAS (Fig. 4) would imply that either the MOUDI is losing material or that the EAS is over-counting?

In figure 4 uncorrected APS data was accidentally used. New figure is submitted where APS data is really corrected against Moudi data. The comparison between different instruments is however difficult because of the totally different operational principles dependent on particle composition and humidity just to mention a few.

P718, l22 and P725, l11. Which process causes the high NO3- deposition during the 2nd sampling period, which is not mirrored by high NH4+? Is it likely due to (i) deposition and washout of NO3- particles that are not NH4NO3, (ii) deposition and washout of HNO3 or (iii) NOx snow chemistry? It seems the ions were not balanced during this period. Is there any ancillary information in the overall dataset? Could the

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combination of deposition and concentration measurements be used to derive effective deposition rates or scavenging ratios?

Our measurements cannot give a sure answer, but HNO3 is rather probable, because an industrial plume was observed at evening of April 30, during that sampling period (that broke the observation of the first nucleation event). Results of atmospheric transport modelling (to be published soon in a separate paper) show that this plume originated from Nikel metallurgy factory about 200 km North from Värriö. According to the measured aerosol concentrations (a few ug/m3) and observation time of that plume (about 15 hours), the deposition velocity must be in order of 1 cm/s to produce the observed deposition loads (assuming that HNO3 constitutes a half or so of the aerosol mass). That seems realistic. Even higher concentrations of aerosol were observed at April 28 and 29, but origin of these is not correctly identified, thus to talk about their composition would be highly speculative.

P719, l4. The product of particle number concentration and gravitational settling velocity will provide a lower estimate of the deposition flux. At 5 μm there is still a strong contribution of non-gravitational processes to the overall deposition velocity, especially over aerodynamically rough surfaces such as forest.

We agree with the referee. Using just the gravitational settling is a first stage approximation. In the early version of the paper we included discussion about bulk resistances, but as deposition velocities below tree crowns are very difficult to calculate and would require a detailed deposition model meant for forests, we left the discussion out.

P719, l19 and Fig. 6. Text and figure caption suggest that the graph should show mass fluxes. In fact it appears to show number fluxes!

We have changed the caption to number flux and added some text to explain that the mass deposition is calculated from the number flux.

P720, l6. As described in the introduction above, the word 'interrupted' reflects, in my

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opinion, not the right concept. At the very least it is not the event that was 'interrupted' but the observation of this event.

As in general comments

P721, l6. No indication appears to be given as to how these growth rates were derived / calculated. Is the dependence of growth rate on particle size consistent with other measurements? What is controlling it?

The paper by Hirsikko et al. (2005) mentioned in the discussion part of the paper gives more detailed description of the model. The growth rates are similar than described in the other papers mentioned in the discussion. The growth of nanometer size aerosol particles is supposed to be controlled by the content of organic vapors in the atmosphere, as well as by the thermodynamical properties of the seed particle and the environment.

P721, l14. Have the fluxes been filtered for unsuitable micrometeorological conditions, such as (i) low turbulence, (ii) non-stationarities and obstructed wind sectors (if relevant)? What are deposition velocities are implied by the deposition periods and how do they compare with the maximum theoretical limit of 1/Ra?

The answer on the first question is no. Low turbulence applies to a very small fraction of the data set, and not during any of the periods of special interest. There is no special wind sector that is obstructed. Non-stationarity is a possible problem in the beginning and end of the pollution periods and in the beginning of the nucleation events, but in both cases a clear downward flux is established after the original transition. The answer on the second question is that such factors are being analysed for a separate parameterisation. A detailed analyse of the deposition processes in the size range dominated by the CPC is out of the scope of the current manuscript.

P721, l28. References are needed to back up the statement that the direction of the flux during nucleation events is consistent with previous observations.

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These references were already in the paper in the discussion part.

Nilsson, E. D., Ü. Rannik, G. Buzorius, M. Kulmala and C. O'Dowd, 2001, Effects of the continental boundary layer evolution, convection, turbulence and entrainment on aerosol formation, Tellus 53B, 441-461.

Buzorius, G., Ü. Rannik, E. D. Nilsson and M. Kulmala, 2001, Vertical fluxes and micrometeorology during the new aerosol particle formation, Tellus 53B, 394-405.

P722, l5. According to Fig. 1, wind speed (and thus turbulence as further supported by the small values of |L| in Fig. 2) is very low on the morning of 30 April, and this is an alternative reason why fluxes during this period are small. It could be argued that fluxes cannot be measured in the morning and the time trace of Fig. 12 is therefore unconvincing. It does, however, show that fluxes are downwards during nucleation events.

In the morning of April 30 we have a very fast transition from stable to unstable conditions (from positive values of 1/L to negative). So there isn't really much "small values" to talk about. Furthermore, we do not understand why the referee doesn't think that fluxes could be measured this morning. Yes, the measured fluxes were small while we had stable conditions and low wind speed, but that is because the fluxes most likely were small. In any case, the main purpose is to demonstrate downward fluxes during the nucleation events, which takes place during the unstable period with more turbulence

P722, l10. The hypothesis that circulation patterns associated with cloud streets is responsible for the fluctuations in the measured aerosol flux is highly speculative. If this were the case, the concentration should show similar fluctuation. Does it? The variability in the flux could equally be due to advection and storage errors during the changing conditions of the nucleation events, which are by nature non-perfect conditions for flux measurements.

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Buzorius et al. (2001) and Nilsson et al. (2001) examined this in more details, including not only periodicities in concentration and cloud cover (radiation), but more importantly, fluxes calculated on a shorter time basis as well as turbulent spectra and cospectra. For this detailed study, the answer was yes, for the current study, it is out of the scoop of the manuscript to go into such details. Finally, in response to the last suggestion of the referee, it should be noted that this behaviour usually continues several hours after the explosive and transient conditions of the initial nucleation event.

- P723, l23. Which ions (presumably not measured) do the authors propose balances the large ammonium concentration in the accumulation mode?

We have added the following sentences in line 21:

A significant lack of negative ions in the accumulation mode is thought to be due to not analyzed organic carbon species, e.g. organic acids or humic-like substances. Organic acid ions are likely contributing to lacking negative ions and owing to their large molecular mass to significantly unresolved particulate mass in the first week sample. Another common anion not measured in this study is carbonate, but it is not quite common in accumulation mode and considering it's relatively low molecular mass not quite capable of balancing the mass.

- P724, l24-26. How was this coarse deposition flux derived from Table 3 and why is the agreement with the deposition estimate based on the EAS data so much better than suggested in Section 3.4?

Derivation of deposition flux from Table 3 is straightforward: deposited mass divided to deposition time. It is assumed that coarse particles constitute the main deposited mass. The mass of spheroidal particles dealt in section 3.4 is usually only a small part of total mass flux (reference is given in 3.4). If the major part of mass was in the insoluble form of spheroidal particles, the deposition fluxes of ions in solution were considerably smaller.

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EGU

- P725, l1. If values of 32 and 0 mg m-2 are judged to be in 'rather good agreement' with each other, one might wonder what the authors would have judged to have been poor agreement. But I do agree that a lot of assumptions went into the comparison.

"rather good agreement" is replaced with "fair agreement".

- P728, l4. "facility's goal is to"

Corrected

Technical Corrections

Title: better: "at Varrio field station"

changed

p711, l2: better: "in the free troposphere"

changed

p711, l15: "as a possible mechanism"

changed

p711, l17: "However, in some special cases ? E"

changed

p712, l16: better: " to identify, the contribution from local diffuse ? E"

changed

p714, l7: "measurements are shown in Table 1."

changed

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p714, l9: "with an Air Ion Spectrometer"

changed

p715, l1: "continuous spectra."

changed

P715, l5: "treated as a real sample"

changed

P715, l18. "Hyytiala" does not appear to have been introduced in the text and not all readers may be familiar with the SMEAR II station and the earlier flux work performed there. Maybe better introduce this as: " ?E to that used in previous flux studies in Southern Finland (e.g. Buzorius et al., 2001), except ? E"

Changed "at SMEAR II station in Hyytiälä, Southern Finland"

P716, l19: better: "temperatures above zero"

changed

P717, l3: "arriving at the SMEAR I station"

changed

P721, l28: "during the pollution episode"

changed

P723, l3: "The first week of the ? E"

changed

P723, l7: Please improve English of "while coarse mode around 2 μm."

"and the coarse mode mean diameter was around 2"

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P723, l8 (and two occurrences l12): "of the Aitken mode"

added the-words

P723, l13: "presents the detailed contribution of each of the inorganic" - There are further omissions of the article throughout the text, that should be corrected.

added the-words (some might be missing)

P726, l14. "similar to the recent"

changed

Table 2. Some additional spacing between rows would make the table easier to read.

Added 4pt between every table row

Fig. 6. The caption and y-axis label are inconsistent (see above).

Corrected

Fig. 15. Presumably this composition is based on mass loading (rather than mole or charge). Please specify in the legend.

Corrected to caption

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 709, 2007.