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ACPD 6, S2274–S2283, 2006

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Interactive comment on "Scavenging of ultrafine particles by rainfall at a boreal site: observations and model estimations" by C. Andronache et al.

C. Andronache et al.

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We thank the reviewer for many useful comments and detailed helpful suggestions that contributed to a series of important clarifications and improved the revised paper. Based on reviewer's comments we made several changes to the revised paper, as described below.

Abbreviations used: A - Authors R - Reviewer

Major comments:

R: 1. What is the purpose of the model? If it is to interpret the data, then this should be stated in the abstract, introduction and conclusions. It is not completely new that removal rates of UF particles in the BL depends on mixing in-cloud scavenging etc., can this model be used to tell which process is most important to examine further? If the

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

model is to be used in air quality models or climate models, then its weaknesses should be clearly documented, the evaluation should be more careful and important processes such as turbulence and nucleation scavenging more physically parameterized.

A: The purpose of this model is to interpret the data from Laakso et al. (2003a) (denoted L2003a in the following discussion), specifically to see what microphysical processes can contribute to the scavenging coefficient determined from surface measurements of aerosol size distribution changes during rain events. We do not propose a new scavenging model to be used in air quality or aerosol transport models. Such models have the transport either represented in an explicit manner or based on assimilated meteorological data. In such cases, the aerosol scavenging module is coupled with the transport. The discussion presented in this paper is focused on interpretation of observational data and reveals the possible importance of various factors, which can be useful for other aerosol scavenging field studies.

R: 2. Parameterization of mixing: What about downward transport of air within the boundary layer? If the BL is assumed to be well mixed, how can there only be a one-way transport? How does this agree with more detailed models? Why assume a linear increase of w with altitude within the BL, I would assume that a logarithmic increase is more realistic? I think in general that the parameterization of mixing described on page 3812 is rather unrealistic, and hence the parameter f1 seems rather arbitrary. Why not try to estimate f1 using a more sophisticated model with turbulence included?

A: The existence of clouds over a mesoscale area is caused by low-level convergence of the mesoscale flow. This convergence forces air, moisture and CN to be lifted, enabling the cloud to develop and produce rain. For precipitating frontal systems, the mesoscale convergence creates an average upward vertical velocity which maintains the cloud system. The reviewer is right, that there are downdrafts as well but on average, the vertical velocity is upward. This feature is most evident in simulations from 3D cloud models or mesoscale models of frontal systems. The value of f1 used in our paper is not arbitrary. It is based on previous estimations of tracer mixing between

ACPD

6, S2274–S2283, 2006

Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

the BL and free troposphere. In the revised paper we provide better references and discussion. We have replaced the simple derivation of f1 with values from simulations with 3D cloud models reported in literature. For example, Niewiadomski (1986) finds f1=0.15 for one hour of mixing, using a 3D cloud model, and several other studies find comparable results (Tremblay 1987; Agusti-Panareda et al. 2005). The suggestion for detailed turbulence modeling is good and we consider it for future work, due to the great extent of such studies and since sufficient published material has been found in the present study to justify f1=0.1 in the reference run and the values used in the sensitivity analysis.

R: 3. I would like to see a more precise statement of what the authors mean by saying that the "model results are comparable with observations". Looking at the sensitivity simulations, especially for f1, I am not sure this is really true. Why is f1=0.1 chosen as a reference? It would also be interesting to see how the model performs for other rainfall rates than 1 mm h-1. It is stated on page 3818 that the observed fit of L0 is suitable for rainfall rates 0.4 - 10 mm/h, but how can you say that from the model simulations?

A: The statement has now been modified. We note that comparison between median values from observations and this model results should be considered when the model parameters (such as f1, EIC, Ta-Ts, etc) are varied in their typical range as shown in the sensitivity figures. We note also, that in previous published studies of aerosol scavenging by rain, comparisons between observations and models show large discrepancies. Such discrepancies were attributed to experimental errors and uncertainties in model parameters used. Value f1=0.1 is taken as reference value based on reported results from 3D models, and the sensitivity to other f1 values is shown. Also, in the revised paper we have made changes to clarify that the most representative value of R is about 1 mm/h for the present study (see for example the histogram in the new Figure 4a). Cases with R about 1 mm/h contributed statistically to the value of L0 reported by L2003a, and the most cases with larger values of R were in fact not included in their initial analysis (see L2003a for discussion).

ACPD

6, S2274–S2283, 2006

Interactive Comment

Full Screen / Esc

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Interactive Discussion

Minor Comments:

R: 1. Page 3802, lines 2-5. The measured scavenging rate data have already been presented in the paper by Laakso et al. (2003a). It is somewhat misleading to have it as a first statement in the abstract without saying that they have been measured and evaluated before.

A: We changed the abstract, clarifying that measurements and L0 values were reported in L2003a.

R: 2. Page 3802, line 5: The range is given for median values of the scavenging coefficient, this should be clarified. It is somewhat confusing to give this value when you in Figure 9 can see observed scavenging rates ranging from 7e-6 to 1e-4.

A: We changed this and now the observed and measured ranges are discussed. Generally the range of Leff from model evaluations is large due to possible variations of parameters, as is evident from the sensitivity studies.

R: 3. Page 3802, lines 10-11. It is stated that "the new model have values comparable with those obtained with observations". Looking at Figures 9-13, the model produces scavenging coefficients between 6e-6 and 2.5e-4, but the dependence on scavenging rate on aerosol size is different compared to the observations. A minimum scavenging rate occurs in the model at approx. 60-70 nm and then there is a local maximum at \ddot{Y} 300 nm which can not be seen in the observations. I think this should be mentioned.

A: The differences are discussed when the new reference run figure is presented.

R: 4. Page 3802, lines 24-26. What about biomass burning?

A: Biomass burning is added with a notable reference.

R: 5. Page 3803, line 7: Particle concentration is not only increased during pollution events.

A: This has been corrected.

ACPD

6, S2274–S2283, 2006

Interactive Comment

Full Screen / Esc

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Interactive Discussion

R: 6. Page 3805, line 5: Which estimates based "only on below-cloud collection removal" are you referring to? How big is the difference between the observations and these calculations?

A: Reference to L2003a is given here. They compared L0 with the below-cloud scavenging coefficients based on Slinn's model, and there are differences of about one order of magnitude.

7. Page 3805, lines 7-9: How is this presentation of data (goal 1) different compared to what is described in Laakso et al. 2003a?

A: The goal was re-phrased as we removed parts of the data description, with reference to L2003a.

8. Page 3805, lines 9-14: What is this model supposed to be used for? Analysis of data? As a parameterization to be used in other models?

A: The model is used to interpret data, specifically to try to understand the empirical scavenging coefficients, and the possible role of several microphysical processes involved in aerosol scavenging during rainfall. The model is not proposed as a parameterization to be used in other models.

9. Page 3806, line 20: Why is only 1998-2001 used and not the whole period?

A: We added all years 1996-2001 in the new revised figure.

10. Page 3807, line 4: Why is it only the particle concentration for particles smaller than 30 nm that increases? How can this be related to transport or mixing and not condensational growth or coagulation?

A: The cause for increase in number of particles smaller than 30 nm can include more factors, and we agree with the reviewer that condensational growth or coagulation can be important, and such factors are mentioned in the revised paper (see L2003a for a discussion).

ACPD 6, S2274–S2283, 2006

> Interactive Comment

Full Screen / Esc

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Interactive Discussion

11. Page 3808, lines 1-2. What about dry deposition? How does dry deposition change with changing surface characteristics (i.e. a wet/dry surface)?

A: Dry deposition was estimated to be about one order of magnitude smaller that the wet deposition during rain events as discussed by L2003a.

12. Page 3809, lines 1-5. I would like to see more clearly distinguished what is new in the present study and what has been presented before in Laakso et al. (2003a). I assume Figure 6 is the same as Figure 7 in Laakso et al.?

A: In the revised paper we remove Figure 6 (which is illustrating the fit of L0 from L2003a)

13. Page 3809, line 2: The observational fit/parameterization of L0 is missing in the Appendix.

A: The fit has now been added to the Appendix.

14. Page 3810, line 5-6. Which field data are used?

A: The discussion has been changed with a brief review of tracer mixing between BL and FT.

15. Page 3811, line 20. What is meant by "convective precipitation has a vertical velocity ? E?" You mean the vertical velocity in general within convective clouds? In that case, it can be clearly higher than 10 ms-1 (cf. eg. "A short course in cloud physics" by Rogers and Yau, 1989, Butterworth Heinmann, 290pp). Do you mean for what is usually observed at Hyytiälä?

A: Such intense vertical velocities are only found in deep convective clouds (in the updraft core). They do not represent average values as we discussed in determination of f1 using simple arguments.

16. Page 3812, lines 10-11. Please insert a reference for the fact that wb on average is positive during a whole rain event.

ACPD

6, S2274–S2283, 2006

Interactive Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

A: Average vertical velocity is positive over a cloudy area due to low-level convergence of the wind field, which creates the vertical flux of water and aerosol needed to maintain the clouds. Meso-scale models show a positive correlation between the rain area and the upward vertical velocity at 850 or 700 mb.

17. Page 3813, line 11. Does chemical composition really not matter that much? How can it then be, as mentioned on lines 26-28 on the same page, that some particles < 50 nm are activated and some with 200nm remain inactivated for the same supersaturation?

A: The statement has been corrected.

18. Page 3813-3814, why not estimate the number of nucleated aerosols based on general Koehler theory, assuming a certain composition of the aerosol?

A: We do not include estimations of the number of nucleated aerosols based on theory because of lack of detailed chemical composition and supersaturation data. We feel that field data cited are quite representative for this calculation and for the purpose of this study. The suggestion is considered for future studies.

19. Page 3815, lines 22-23. Please specify what is meant by "Model predictions of Leff are comparable with L0 from observations". It would also be interesting to know what the differences mean in terms of UF particle concentration.

A: We revised this section and added a discussion on impacts on UFP concentration.

20. Page 3818, lines 15-26. I think this discussion is a bit out of place, as the L0 scavenging coefficient already has been published in Laakso et al. (2003a). It would be more interesting to see a discussion around Leff. And how do you know that L0 obtained from the data from Hyytiälä is representative for other locations?

A: Discussion on L0 concerning material already presented in L2003a is eliminated and we have added some new discussion about Leff.

ACPD

6, S2274-S2283, 2006

Interactive Comment

Full Screen / Esc

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Interactive Discussion

21. Page 3819, lines 1-5. Again, I think it should be clarified that these results are from Laakso et al. (2003a).

A: This has been corrected.

R: Page 3803, line 6: ultrafine particles have already been defined as UP.

A: This has been corrected.

R: Page 3802, line 21: Change "atmospheric particles removal" to "atmospheric particle removal".

A: This has been corrected.

R: Page 3802, line 29: Insert "e.g" before the reference Komppula et al (2005).

A: This has been corrected.

R: Page 3810, line 6: Insert "the" before "..cloud, where super ? E"

A: This has been corrected.

R: Page 3810, line 8: Insert "e.g" before the reference Komppula et al (2005).

A: This has been added.

R: Page 3811, line 32: Change "that" to "than".

A: This has been added.

Figures and Tables:

R: Figure 2: I am not sure I understand what is showed on the x-axis in this figure. Is this the average concentration for a rain event with duration of a certain length? Are events shorter than 0.5h removed from this figure?

A: This figure was removed in the revised paper. Due to removal of some figures, the numbers are changed in the revised manuscript.

ACPD

6, S2274–S2283, 2006

Interactive Comment

Full Screen / Esc

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Interactive Discussion

R: Figures 3: Are rainfall rates smaller than 0.4 mm h-1 removed from this figure?

A: The data for R< 0.4 mm/h are not removed from this figure, as it is meant to contain all of the data.

R: Figure 5a: Same as for figure 2, what does the x-axis mean? Is 6000x15 min the longest rain event?

A: The figure 5a was removed. A new figure is shown to illustrate the frequency of various rain intensities.

R: Figure 5b: Are rain events <0.5 h not removed from this figure (and the others)?

A: The figure contains all data.

R: Figure 9. Why is only RH=60% and RH=99% showed and not the reference 90%? Figures 10-13: Is the black curve supposed to be the reference simulation in all figures? Then why is it then not the same?

A: We use now RH =60% and RH=95% in the Figure 9 for sensitivity to relative humidity. Then, we use RH =95% in all the other figures. This way, RH is consistent in all figures.

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ACPD

6, S2274–S2283, 2006

Interactive Comment

Full Screen / Esc

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Interactive Discussion

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ACPD

6, S2274–S2283, 2006

Interactive Comment

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