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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

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This paper aims at presenting a new model for estimating total boundary layer (BL) wet scavenging rates of ultra fine (UF) particles (10<d<510nm). Apart from direct below-cloud scavenging of particles by rain, the model also considers mixing/transport of particles, subsequent uptake of particles by cloud droplets and removal by in-cloud precipitation processes. The model is compared with observations conducted over southern Finland 1996-2001. The topic of the paper is interesting. However, I do have some reservations on the presentation and the methodology.

The model presented is aimed at dealing with aerosol removal by rain in a more sophisticated way than just looking at aerosol concentration vs. rainfall rate, but processes such as mixing by turbulence and droplet activation are dealt with in a very simplistic way. The question is then, is it really better than just assuming a removal rate dependent on precipitation intensity? More precisely, I am not sure for what purposes the model can be used for. Air quality models and climate models usually already have some kind of turbulent transport/mixing of particles within the BL and nucleation scavenging should also be considered.

If the proposed removal module should be used for modeling purposes, I would like to see a more careful evaluation against observations to show that the proposed model is more useful than a “standard” model. As it is now, the uncertainty in the calculated removal rates is large and the dependency of the removal rate with particle does not even show the same distribution (cf. Figure 9). I would also like to see an evaluation of (especially) the parameter f1 that is deduced, how does this compare with more detailed calculations using a turbulence model?

If the proposed model is just a tool for interpreting the data and showing that measured UF particle rates in the BL are not only dependent on rainfall rates, I think this should be clearly stated in the abstract, introduction and conclusions of the paper.

Major comments

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 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.

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

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3812 is rather unrealistic, and hence the parameter f1 seems rather arbitrary. Why not try estimate f1 using a more sophisticated model with turbulence included?

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 L0 is suitable for rainfall rates 0.4≤R≤10, but how can you say that from the model simulations?

Minor Comments

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.

2. Page 3802, line 5: The range given is for median values of 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.

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 ˜300nm which can not be seen in the observations. I think this should be mentioned.

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

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

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?

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?

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?

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

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?

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)?

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.?

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

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

15. Page 3811, line 20. What is meant by “convective precipitation has a vertical velocityĔ?” 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?

16. Page 3812, lines 10-11. Please insert reference for the fact that wb on average is

positive during a whole rain event.

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 <50nm are activated and some with 200nm remain inactivated for the same supersaturation?

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

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

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

Spelling etc.

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

Page 3802, line 21: Change “atmospheric particles removal” to “atmospheric particle removal”.

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

Page 3810, line 6: Insert “the” before “..cloud, where superĚ”

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

Page 3811, line 32: Change “that” to “than”.

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Figures and Tables

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?

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

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

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

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?

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 3801, 2006.

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