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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Explicit simulation of aerosol physics in a cloud-resolving model" *by* A. M. L. Ekman et al.

Anonymous Referee #2

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I) General Comments:

The paper is focused on the role of aerosols in a convective cloud system. The impacts of key parameters and key processes of the aerosol cycles on the cloud macro- and microphysical properties are studied. Therefor, a cloud resolving model was interactively coupled with detailed aerosol and chemistry modules. The study also covers the convective transport of aerosols and trace gases into the upper troposphere.

Since aerosol-cloud-chemistry interactions as well as the aerosol budget of the upper troposphere and lowermost stratosphere are still unlcear in many concerns, the paper covers a highly relevant field of atmospheric research.

The interactive coupling of a cloud resolving microphysical model with a detailed aerosol module and a comprehensive chemistry scheme is progressive and forward-

looking.

The concept of the study has been well elaborated. The simulations were performed and analysed carefully. Many mechanisms of potential effects of aerosols on convective cloud systems were identified.

Nevertheless, there are several specific points which have to be further discussed. Some open questions as well as suggestions to improve the manuscript are listed below.

II) Specific Comments:

1. The prescribed initial aerosol number concentrations of 50 cm^{-3} and 500 cm^{-3} in the Aitken- and accumulation mode, respectively, are very small for continental urban air. As reported also by Seinfeld and Pandis (1998), the concentration of fine particles can be in the range of 10^3 to 10^4 cm^{-3} in urban air. Typical concentration of accumulation mode particles amount to 10^4 cm^{-3}. The choice of only small number concentrations implies the following questions:

- i) Why are the number concentrations chosen that small?
- ii) The chemistry module is initialized with SO_4 taken from the EMEP data base. Since this SO_4 has to be interpreted as particulate matter, it should be identified with the inititial mass of the SO_4 modes. How is the SO_4 mass distributed on the Aitken and accumulation modes? Is the SO_4 mass concentration generally conform with the modes size ranges and typical sizes of accumulation mode particles when small initial particle number concentrations are considered? These points should be dicussed in more detail. (see also III.18).
- iii) The sensitivity experiments indicate a major role of aerosol nucleation and coagulation in the development of the cloud. New aerosol particles are formed and are transformed to CCN which than initiate cloud nucleation. However, the

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small particle number concentrations chosen may promote the nucleation of new aerosol particles rather than condensation of H_2SO_4. This may promote the role of aerosol nucleation and coagulation in the simulations. In the simulation D7 larger initial particle numbers were considered and the impact on the aerosol budget seems to be small compared to the reference simulation. However, it might be possible that aerosol nucleation and coagulation are much less important in D7 as in the reference simulation. This would change some of the conclusions.

The authors should comment on these points. Especially on the question: How important are processes like aerosol nucleation and coagulation when much larger aerosol number concentration are initialized? An appropriate set of simulations (at least the R, A and F simulations) has to be repeated considering realistic initial aerosol number concentrations. If the conclusions of the study significantly change if these additional simulations are considered it would be necessary to repeat also the other simulations.

2. There are two manuscripts in preparation which are used as references (Ekman et al. on the evaluation of the model, Wang et al. on heterogeneous chemistry in the model). If it is unclear whether the papers will be published before the present study, the present manuscript should contain some more discussions on the performance of the model compared to observations or the heterogeneous reactions, respectively. Just to compare the maximum values (listed in the Figures) does not sufficiently prove the model quality. The discussion on the evaluation (page 765) could be extended, for instance, by an evaluation of the spatial and temporal development of the cloud. It might be appropriate for the present study just to demonstrate that the cloud shows typical characteristics of a convective cloud as reported by observations.

3. The model seems not to include prognostic ammonium/ammonia or aerosol nitrate which can be, together with sulfate, important compounds of liquid aerosols impacting the aerosol water content and size, especially in polluted air. The paper should include at least a brief discussion on the potential of these compounds to change the main

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conclusions of the study. The experiment E5 partly covers this point. Such a discussion could also advert or motivate future studies on the role of secondary organic aerosols in cloud formation.

4. Emissions of primary particles are neglected (page 759, lines 12-15). Since primary particle emissions can be an important source of accumulation mode aerosols, which mostly can serve as CCN, the results may be sensitive to the inclusion of primary particle emissions. The study should include at least one sensitivity experiment including primary particle emissions. An experiment considering, for instance, a constant surface source of BC would be sufficient to evaluate the potential importance of particle emissions. Emission rates typical for Germany can be taken, for instance, from Cooke et al. (1999) [JGR, 104, 22137–22162].

5. In figures 6-10, relative changes are presented which were calculated from the differences between the respective sensitivity experiments and the reference simulation for specific altitudes. If the cloud shape changes in the sensitivity simulations, it might be possible that the calculated relative changes partly reflect the changed cloud shape (e.g., changed cloud base/'center'/top) rather than the microphysical properties which might be similar to the reference case at other altitudes. If this is the case, the authors should compare, for instance, maximum values or values at cloud base/'center'/top instead of changes at specific altitudes? The authors should either prove that changes in the cloud shape are not relevant or change Figures 6-10 in an appropriate way.

6. The model includes size dependent activation of CCN. The aerosol module uses fixed size limits separating the different modes. This implies that, due to the log-normal distributions assumed, the aerosol number concentration generally is small at the size limits. Hence if the size limit between the Aitken and accumulation mode is located in the size range where effective activation starts, the number of cloud droplets may be misrepresented. This effect can be seen in simulation H3 where the size limits were decreased and more cloud droplets are formed at the cloud base. It should be discussed in more detail that the modal concept of the aerosol module can cause

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problems in cloud simulations if size dependent activation is considered.

III) Minor and Technical Corrections:

1. Title:
The title should give the information that convective clouds are the focus.

2. Abstract:
'Nucleation mode aerosols (0<d<5.84...' change to ...(here defined as 0<d....); same for accumulation and Aitken mode.

3. Introduction, p755 line 17:
The list of References concerning upper troposphereic small particle concentrations is not complete. Say (e.g., Clarke ....) or include at least:

- Hermann et al., Meridional distributions of aerosol particle number concentrations in the upper troposphere and ...., JGR, 108(D3), 4114, doi:10.1029/2001JD001077, 2003.
- Petzold, A., Fiebig, M., Flentje, H., Keil, A., Leiterer, U., Schröder, F., Stifter, A., Wendisch, M., and Wendling, P., Vertical variability of aerosol properties observed at a continental site during LACE 98, JGR, 107(D21), 8128, doi:10.1029/2001JD001043, 2002.
- Schröder, F., Kärcher, B., Fiebig, M., and Petzold, A., Aerosol states in the free troposphere, JGR, 107(D21), 8126, doi:10.1029/2001JD000194, 2002.

4. page 759, line 9; and Table 1:
'deviations are pre prescribed': A reference should be given for the choice of sigmas listed in Table 1.

5. page 759, line 27:
'When 5% of the mass and number...' Since lognormal size distributions are assumed,

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mass and number show different size dependences. It should be discussed more clearly which thresholds for mass or/and number apply.

6. page 761, line 17ff.:
Nucleation scavenging and impact scavenging by precipitation are discussed as the aerosol scavenging mechanisms. Is the impact scavenging of the non-precipitating cloud water/ice not important in a convective cloud? A short discussion on that point should be included.

7. page 762, lines 2-3:
'Brownian diffusion .... is the most important removal process for small particles': the diffusion itself is not the removal process, but the particle-particle interactions triggered by diffusion. The formulation should be modified.

8. page 762, lines 12-13:
10 aerosol variables are included in the cloud module (5 modes, each with number and mass concentration). How is the composition of the mixed modes treated? Are the Mass of BC,OC and SO_4 not prognosed separately? This should be explained in more detail.

9. page 763, line 28:
Is the [rho(level)/rho(surface)]^3 dependence conform with the available measurements (by e.g., Clarke et al; Petzold, et al; Minikin et al, see point III.3) ?

10. page 765, lines 23-25; Figure 5:
The plume is hardly visible. The scale of the color bar should be modified.

11. page 767, line 17:
'substantially', Figure 6 shows that the UT Aitken MNC decreases by less than 10%. One should better say 'significantly' (for instance) instead.

12. page 767, line 20:
'... decreases approximately 40%'. According to Figure 6 the decrease is only about

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20%.

13. page 767, line 28ff.:
It should be explained in more detail whether the nucleation rate is increased by only fasten the nucleation process or by increasing the amount of nucleating gas.

14. page 768, section 4.3.:
It should be explained in more detail why the precipitation rate is increased although the convection is less effective. Is this caused by a reduced number of CCN growing larger to precipitate more efficiently?

15. page 770, line 7:
Why are the particles larger or fewer in D5 where the total mass is divided by 2?

16. page 770, lines 10-11:
Why is the coagulation and condensation inefficient when many particles and consequently a large surface area concentration is availble? Do you mean that the growth of the particles due to coagulation and condensation is inefficient because there are to many competing particles?

17. page 770, lines 22-23:
What is the time-scale of SO_2 oxidation in the simulations? Is the production of gas phase sulphate realy that effective that many large particles can be formed? This should be discussed because the SO_2 Oxidation is eventually a limiting process in the whole system.

18. page 771, lines 3-4:
Why does the aerosol nucleation rate dependent on the initial H_2SO_4 concentration? Do you mean that the amount of pre-existing particles determines whether nucleation or condensation occurs preferently or that initial H_2SO_4 controles the amount of nucleating material? The EMEP SO_4 should be mainly or even purely particulte SO_4. Are the EMEP SO_4 concentrations splitted in particulate and gas phase SO_4 to initialize

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the model? This must be explained in more detail because initial H_2SO_4 seems to a driving parameter.

19. page 772, lines 16-18:
This mechanism should be explained in more detail. Why can the ice particles grow more efficiently when less Aitken particles are available? How is the ice formed an what is the role of aerosols?

20. page 772, lines 27-29:
It should be mentioned that the aerosol number concentrations are realistic in case D7 (see, II.1).

21. page 774, lines 21ff.:
This dicussion should include the points mentioned under II.6.

22. page 775, line 9:
change Sect. 2.2 to Sect. 2.1

23. Figures 6-10:
The title 'droplet/particle NC' of plots b and c is misleading. Use ice/droplet particle NC as in the figure caption.

24. Figure 11 (a):
The title 'Cloud radius at 2km' is misleading. Use 'cloud particle radius' or 'droplet radius' or similar.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 753, 2004.

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