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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.

A. M. L. Ekman et al.

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Response to reviewers` comments A. Ekman, C. Wang, J. Ström, and J. Wilson

We would like to express our appreciation to the two reviewers (Dr. Mary Barth and an anonymous reviewer) for their careful reading our paper, and for providing us with many constructive suggestions and comments that has led to a significantly improved manuscript.

In the following section, we give our responses to each of the two reviewers` comments or questions in a format where we first list their comments in order, and then our corresponding answer will follow.

Referee 1 (Mary Barth):

General Comments: 1. One aspect of the work that surprised me was how sensitive the

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storm characteristics (e.g., updraft speed and precipitation amount) are to the aerosol formulation. Could the authors state whether they think observed deep convection contains these same sensitivities? For example, the initial concentration of aerosols affected precipitation amount by 30-70% in some of the sensitivity simulations.

A follow-up question on this topic is whether the storm characteristics will show the same dependencies if the spatial resolution were much finer (on the order of 100s of meter).

Reply: This is a very interesting comment. Recently analyzed satellite data has shown that scattered cumulus cloud cover can be reduced from 38% in clean conditions to 0% for heavy smoke conditions (Koren et al., 2004). In the same issue of Science, Andreae et al. present results from observations of cloud properties performed in both heavy smoke from forest fires and in clean conditions. Onset of precipitation is increased from 1.5 km above cloud base in pristine clouds to more than 5 km in polluted clouds and 7 km in pyro-clouds. Elevating the onset of precipitation under smoky conditions leads to a more vigorous storm cloud (higher wind speed, larger ice hydrometeors), but the net effect on the area amount of precipitation remains unknown. It is suggested that the overall hydrological cycle should be slowed down and regional precipitation should decrease under smoky conditions. A comment on this matter has been included in the introduction and also in Section 4.7 and in the Conclusions. It is worthwhile noting that the purpose of this paper is to explore the direction rather than rate of the model response. The selected ranges for many control parameters are quite large and hence this often results in a large range of monitored results.

On the follow-up question: we do not believe that the model resolution would impact qualitatively on the model response to the changes in control parameters, though quantitative difference is expected. The latter is mainly caused by the resolution-dependency of grid supersaturation ratio, which is a critical parameter in our explicit aerosol and cloud microphysics that determines conversion rates of several microphysical processes. However, the model would conserve the total available water so that

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the directions of model response would unlikely be different in runs with different spatial resolutions.

2. Are the results at t=3h similar to what occurred throughout the simulation?

Reply: The results at t=3h are representative for what is happening throughout the simulation. In order to visualize the rain, ice and graupel development, a new figure (Figure 12) is now included in the manuscript (together with a discussion in Section 4.7).

3. One set of sensitivity simulations explored the dependence of precipitation scavenging of aerosols on aerosol, cloud, and transport properties. An interesting result from these tests showed a 40-50% increase (decrease) in precipitation amount at the surface due to an increase (decrease) in the collision efficiency of aerosols onto precipitation (rain, snow, or graupel/hail). How does precipitation depend on impact scavenging of aerosols?

Reply: Precipitation depends on the number of available CCN (as the precipitation amount depends on the development of the cloud and on the size of the cloud/rain drops). In case of higher collection efficiency, fewer aerosols will be present to continually support the condensation initiated microphysical conversions leading to rain. A comment on this has been added in the manuscript (Section 4.4).

4. What is the importance of the boundary conditions to these results? One of the major conclusions of the paper is that the supply of CCN must be continuous to maintain vigorous convection. The authors point to the importance of aerosol condensation and coagulation to sustain the CCN supply, but couldn`t the transport of CCN from outside the domain also maintain the CCN supply? The authors impose no flux of aerosols or chemical species into the model domain, yet in reality these tracers do flow into the domain from other regions.

Reply: This is an interesting suggestion. We used the radiative type of lateral boundary

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condition in our simulations (note the horizontally inhomogeneous profiles). For the outflow side of the lateral boundary, no flux of matters from outside of the domain is allowed. However, there is actually an equivalent constant (note not an additional) flux towards the domain on the inflow boundary (according to the radiation type of lateral boundary condition, the gradient of normal flux, not the flux itself, is zero at inflow side) so that the value of a given variable at this boundary point remains unchanged due to a normal flux gradient. Certainly, it would be an interesting issue to address that how important an additional supply of aerosol precursors and aerosols would be to the modeled results.

5. One of the many results shown was the average ice particle radius near cloud top for each of the simulations. Although the average value between simulations did not differ greatly, I found it interesting that out of the 35 cases (11 cases shown in plot) only 3 simulations (D1, E3, H3) revealed a relatively large variability during the 3h integration. Could the authors speculate as to what causes the ice particle radius to vary so much for these 3 cases, but not for the other cases? Is this a result of the storm development (intensity and structure) on cloud physics because of variations in aerosol representation?

Reply: Just for clarity, the figure is unfortunately somewhat misleading, as the x-axis is very narrow. There is actually a variance of a micrometer or so in the R, A1 and A2 simulations. Mary is right though, that the variability is much larger in the D1, E3 and H3 simulations. These cases are all simulations with very intense convection and more extensive ice formation compared to the other cases. Note that we applied to all simulations the same initial concentrations for aerosols that are capable of serving as ice nuclei. The most important source of ice crystals in the tower is the condensation-freezing mechanism, which is quite sensitive to the convective strength. Our guess is that as the upwind is much stronger, the graupel and ice particles can grow more efficiently while on the other hand more ice particles can form in the upper part of the cloud than in the other cases, and as a result you obtain a large variability in the ice

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particle radius. A comment on this matter has been made in the manuscript (Section 4.7).

6. Andreae et al. (2004), Science, 303, 1337-1342, recently published an article showing that rain is suppressed from deep convection when smoke is abundant in the atmosphere. One reason is because cloud drops activate on many, smaller CCN (there is no tail in the cloud drop distribution going to large drop sizes at cloud base), which cannot coalesce to form rain. In any of the simulations presented in this paper, or any subsequent simulations, have the authors seen a similar result to that presented by Andreae et al.

Reply: See Reply to Comment 1. We did see that (in accordance with the observations by Andreae et al.) the precipitation and storm development becomes more vigorous when more particles are present (note that Andreae et al. did not discuss the surface precipitation in their paper). However, we did not see any suppression of rain at low levels (note again that our case is a deep convection dominated by cold-rain processes; the large ice particles would be likely to fall and melt eventually). Hence, the average precipitation amount was larger in the simulations when more particles were available though the precipitation might come late or last longer. The result could however be somewhat affected by the fact that we prescribe a fixed standard deviation in the log-normal distribution for the different aerosol modes. We are planning on doing a more thorough study on the response of modeled storm to aerosol`s chemical composition and concentrations.

Referee 2:

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 1,000 to 10,000 cm-3 in urban air. Typical concentration of

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accumulation mode particles amount to 10,000 cm-3. The choice of only small number concentrations implies the following questions:

i) Why are the number concentration chosen that small?

Reply: The reviewer is right that the chosen initial concentrations are in the lower range for urban air. However, it should be noted that for comparison with observed accumulation mode particles, not only the sulfate accumulation mode number should be considered, but also the mixed mode number (as these aerosol groups are of the same size). The total initialized number of accumulation mode particles is then 1e3 cm-3, which is not an unrealistic number. Also, the number of 10,000 cm-3 cited by the reviewer perhaps is for all types of aerosols (over the urban areas or inside biomass plumes there are numerous soot particles) while we only include in our model sulfate and mixture in this size range. As for the Aitken mode particles, the initialized number is small, but particles from the nucleation mode grow very rapidly into the Aitken mode. After 10 min of simulation, the initial condition for the Aitken mode is basically "forgotten". Therefore, the initial number concentration of Aitken aerosol is not a critical parameter compared to the initial mole fractions of aerosol precursors. We have changed the formulation for the justification of the chosen initial conditions in Section 3 in the manuscript. See also reply to comment 1iii.

ii) The chemistry module is initialized with SO4 taken from the EMEP data base. Since this SO4 has to be interpreted as particulate matter, it should be identified with the initial mass of the SO4 modes. How is the SO4 mass distributed on the Aitken and accumulation modes? Is the SO4 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 discussed in more detail.

Reply: This is a misunderstanding perhaps caused by an unclear description in the manuscript. Only gases in the model are initialized with EMEP-data (not SO4 and not

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aerosols). The aerosol mass is initialized by assuming that all aerosols within each mode are spherical, has a radius of 6.29e-9 for the Aitken mode and 0.485e-6 for the accumulation mode and a density of 1.7 g cm-3. This is now clarified in the manuscript.

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 then initiate cloud nucleation. However, the small particle number concentrations chosen may promote the nucleation of new aerosol particles rather than condensation of H2SO4. 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.

Reply: In the D7 simulation, the total amount of condensate H2SO4 on particles is still smaller than the total coagulated mass to the accumulation mode and the total nucleated H2SO4 mass only differs by 2%. We also repeated the R and A series simulations (R_high and A_high, see comment below) with a substantially higher initial Aitken and accumulation mode number concentration, to check if the condensation process became more important. In the A3_high simulation (where condensation is shut off), the difference in Aitken MNC at 10.4 km is larger than for the A3 simulation (37% compared to less than 10%), but the difference is still smaller than for the A1_high simulation. Coagulation is thus still a more important growth process compared to condensation. See also reply below.

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 is 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

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simulations are considered it would be necessary to repeat also the other simulations.

Reply: The R, A and F simulations have been repeated with an initial Aitken and accumulation mode aerosol number concentration equal to 1e3 and 1e4 cm-3, respectively (R_high, A_high and F_high). The convection is more intense in the simulations with higher initial aerosol concentration (higher vertical wind speed, more graupel, more precipitation, etc.). However, even with the higher initial aerosol number concentration, coagulation is still the most important growth process. In addition, aerosol nucleation is still an important process. If the aerosol nucleation is decreased by a factor of 2 over the whole model domain (F1_high), the Aitken MNC at 10.4 km decreases by 78% (compared to less than 5% for the F1 simulation). As the condensation on particles is more efficient in the F1_high simulation compared to the F1 simulation, less H2SO4 is available for nucleation and there are not enough small particles formed to sustain the storm cloud. For the F2_high and F3_high, the sensitivity is about the same as for the original F2 and F3 simulations. In summary, the results are somewhat different for the simulations with higher initial aerosol concentration, but the conclusions remain the same.

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 the heteorogeneous 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 does not sufficiently prove the model quality. The discussion on the evaluation cold 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.

Reply: The comment of the reviewer has been well taken. We wish we could give a more detailed description/discussion in this already a long paper of the previous works

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using this model. The cloud-resolving model has been extensively used in many case studies before incorporated with the explicit aerosol module (before this paper) and these previous works has been reported in several of the cited references. Interested readers should be able to judge the model performance based on these publications. We simply want to emphasize on the new addition, i.e. the explicit aerosol module, to the cloud-resolving model in the current paper. Citations of unpublished papers have been revised based on their newest versions. Heterogeneous uptake of chemical species do not affect on the major processes giving the major conclusion of this paper. Heterogeneous chemistry will be discussed in a future paper by C. Wang, which is now submitted to Journal of Geophysical Research.

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 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.

Reply: We agree with the reviewer that this is an important aspect. A discussion is now included in Section 4.4 in the manuscript.

4. Emissions of primary particles are neglected. 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.

Reply: The simulation time of 3h is very short, and during this time period most emission sources of primary particles would be small. If a BC source of 10Gg BC/1deg x

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1deg/yr is assumed, it implies a BC source of approximately 40e-9 g m-2s-1. A sensitivity simulation including this source has been performed (assuming as in Wilson et al., 2001, that 50% of the emissions are emitted as OC and 50% as BC) and the impact on precipitation, aerosol concentration at 10.4 km, SO2 concentration and CO concentration is small (less than 10%). Even if we increase the emission source by a factor of three, the changes in all parameters are small. The largest change is seen in precipitation with an increase of about 30%. Most likely the major impact of BC aerosols on cloud takes place through radiation rather than microphysics. We have plans in near future for a more comprehensive study of BC and OC aerosols` impact on cloud formation and development, with an addition to the model of their radiative effects.

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 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.

Reply: We might have misunderstood the reviewer`s question. For the Aitken MNC, CO concentration and SO2 concentration, the major issue of interest in this study is to examine the redistributions of these species by convection, in particularly their abundance in the middle and upper troposphere (the free troposphere), as impacted by changes in control parameters. These results are mainly dependent on the convective strength. For the droplet/crystal size at 10.4 and 4 km, respectively, there is a slight shift in the height of the anvil and base between different simulations, their dependency on the convective strength, however, is the same to that of redistribution of chemical

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species. On the other hand, the shape of cloud is also a direct result of convective strength; the impact on the cloud shape of changes in control parameters is thus similar to that on the redistribution of chemical species or microphysical properties we chosen in the analyses. If we make the comparison at one higher, or one lower level, the conclusions remain the same. Note that the output step (5-10 minutes) is much longer than the model time step (5 second). Therefore, using statistical rather than snapshot data in analyses is necessary.

5. 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 formed at the cloud base. It should be discussed in more detail that the modal concept of the aerosol module can cause problems in cloud simulations if size dependent activation is considered.

Reply: The modal models, mainly because of the use of a size distribution prescribed by a given analytic function, might under- or overestimate the activated number of aerosols. The argument is based on the assumption that the prescribed function might not well describe the actual distribution of particle in a high-resolution sense. In particular, when the activate size is very close to the ending size of accumulation mode, it really corresponds to the tail of the size spectrum of accumulation mode, the representation is thus reasonably questioned due to the very low density of particles larger than this size (note the size spectrum is indeed a probability distribution function). Theoretically speaking, the same error also exists in the beginning size range of accumulation mode (the size limit separating Aitken and accumulation mode as expressed in the reviewer`s comment). However, this is least likely to happen in practice because of the mass conservation and number conservation restricted to each of these aerosol modes

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(realizing that when the activate size is between Aitken and accumulation, the activation is actually to convert the whole accumulation mode to cloud droplet). Therefore, the reviewer might have accidentally addressed a correct point with a wrong example here. In our opinion, this potential error of modal model cannot be easily solved by another commonly used model, i.e., the section model, which uses a limited number of size sections (cf. e.g. Zhang et al., 1999, Aerosol Sci. Technol., 31, 487-514). Actually, H series is designed to explore the influence of this error in modeled results.

Other comments:

* Reviewer2, III1. Title.

Reply: We have adopted the reviewer`s suggestion to change the title to "Explicit simulations of aerosol physics in a cloud-resolving model: A sensitivity study based on an observed convective cloud case"

* Reviewer2, III6.

Reply: Because the sedimentation velocities of aerosols and cloud droplets (non-precipitating particles) are very small and thus they both basically travel with air. Kinetic collisions between these two types of particles (i.e., the impact scavenging of aerosols) are hence negligible compared to the collision between aerosols and much larger precipitating particles.

* Reviewer2, III8.

Reply: No, the composition compounds in the mixed modes are not diagnosed separately. The ratio between BC/OC/SO4 is assumed to be constant. This is now commented in the manuscript.

* Reviewer2, III17.

Reply: We are not sure if we have understood the comment of the reviewer correctly, which states, "time-scale of SO2 oxidation in the simulations". We think that this ques-

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tion could be partially answered by using the results of sensitivity simulation series A. Condensation of H2SO4 is not as important as coagulation as a growth process for the particles, but if condensation is removed, considerably fewer large particles are formed. The H2SO4 that is initially present in the model is very rapidly wet scavenged. Hence, the H2SO4 that is available for condensation comes from oxidation of SO2. Nucleation of particles takes place throughout the simulation. During 3h of simulation a total of approximately 1e28 new particles are formed. The clear-sky oxidation of SO2 by OH is relatively slow (SO2 life-time of approximately 2weeks), so in-cloud oxidation of SO2 by H2O2 and O3 should be the major production path for H2SO4. Hence, not only SO2 is the limiting factor but also the oxidants H2O2 and O3.

* Reviewer2, III18.

Reply: The nucleation rate is dependent on the H2SO4 concentration as H2SO4 controls the amount of material available for nucleation. Cf. also reply to comment II1.

* Reviewer2, III20.

Reply: We consider the original aerosol concentrations applied as realistic too (cf. reply to comment II1). However a comment that the D7 number concentration is not unrealistic has been added.

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