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Aerosol effects on deep convective clouds: impact of changes in aerosol size distribution and aerosol activation parameterization

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Abstract

A cloud-resolving model including explicit aerosol physics and chemistry is used to study the impact of aerosols on deep convective strength. More specifically, by conducting six sensitivity series we examine how the complexity of the aerosol model, the size of the aerosols and the aerosol activation parameterization influence the aerosol-induced deep convective cloud sensitivity. Only aerosol effects on liquid droplet formation are considered. We find that an increased aerosol concentration generally results in stronger convection, which for the simulated case is in agreement with the conceptual model presented by Rosenfeld et al. (2008). However, there are two sensitivity series that do not display a monotonic increase in updraft velocity with increasing aerosol concentration. These exceptions illustrate the need to: 1) account for changes in evaporation processes and subsequent cooling when assessing aerosol effects on deep convective strength, 2) better understand graupel impaction scavenging of aerosols which may limit the number of CCN at a critical stage of cloud development and thereby dampen the convection, 3) increase our knowledge of aerosol recycling due to evaporation of cloud droplets. Furthermore, we find a significant difference in the aerosol-induced deep convective cloud sensitivity when using different complexities of the aerosol model and different aerosol activation parameterizations. For the simulated case, a 100% increase in aerosol concentration results in a difference in average updraft between the various sensitivity series which is as large as the average updraft increase itself. The model simulations also show that the change in graupel and rain formation is not necessarily directly proportional to the change in updraft velocity. For example, several of the sensitivity series display a decrease of the rain amount at the lowest model level with increasing updraft velocity. Finally, an increased number of aerosols in the Aitken mode (here defined by 23 nm$\leq d \leq$ 100.0 nm) may result in a larger impact on the convective strength compared to an increased number of aerosols in the accumulation mode (here defined by 100 nm$\leq d \leq$ 900.0 nm). When accumulation mode aerosols are activated and grow at the beginning of the cloud cycle, the

supersaturation near the cloud base is lowered which to some extent limits further aerosol activation.

1 Introduction

Atmospheric aerosols act as cloud condensation nuclei (CCN) and are therefore important in cloud formation processes. Anthropogenic CCN generally give rise to an increase of the cloud droplet number concentration (CDNC), which for a constant amount of liquid water reduces the average cloud droplet size and increases cloud albedo (first indirect effect, Twomey, 1977). As a consequence, precipitation processes and cloud life time may be altered (second indirect effect, Albrecht, 1989). For mixed-phase clouds, such as deep convective clouds, the effect of an increased aerosol concentration is more elaborate. For these clouds, aerosols may affect not only the droplet formation but also the ice formation processes within the cloud, directly through heterogeneous freezing and indirectly through changing the droplet size and thereby the freezing temperature. Some cloud-resolving model studies of single (isolated) deep convective clouds show a decrease of precipitation with increasing aerosol concentration, whereas others show a precipitation increase with increasing aerosol concentration (Ekman et al., 2007; Rosenfeld et al., 2008, and references therein). Rosenfeld et al. (2008) suggested that if the warm phase precipitation of a cloud is reduced, the amount of cloud water reaching the freezing level increases. This may in turn enhance the latent heat release and give higher updraft velocities leading to more vigorous deep convection. On the other hand, smaller cloud droplets also imply a shift of the homogeneous freezing level to colder temperatures, which may decrease buoyancy and updraft velocity as more water mass has to be transported to higher levels. Based on this line of reasoning, an increase of the aerosol concentration should invigorate convection until an optimal aerosol loading is reached. Thereafter convection should be suppressed.

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Fan et al. (2009) also noted the contradicting results between different studies of aerosol effects on deep convection and suggested that vertical wind shear and relative humidity play an important role in moderating the aerosol impact on deep convection. They argued that increased aerosol concentrations always suppresses convection under conditions with strong vertical wind shear as the increased evaporation of cloud droplets leads to a more stable stratification dominating over the invigoration due to increased latent heat release. On the other hand, for weak vertical wind shear, the increase in latent heat release is at first larger than the increased evaporative cooling so that convection is enhanced until an optimal aerosol loading is reached.

Several of the model studies referred to above utilize a simplified description of the aerosol population chemistry and dynamics where an initial aerosol size distribution/concentration is assumed and the initial aerosol composition is prescribed. In some of the models, the aerosol population is advected and scavenged throughout the simulation whereas other models utilize a time-invariant aerosol concentration. Different methods of activating aerosols into cloud droplets are also used, e.g. empirical formulations or Köhler theory.

In the present study, we examine the sensitivity of a single deep convective cloud to varying aerosol concentrations and how this aerosol-induced sensitivity may depend on the complexity of the aerosol model as well as the aerosol activation parameterization. We also investigate if the size of the aerosols may affect the aerosol-induced sensitivity. In order to simplify the analysis, only the aerosol effect on liquid droplet formation is considered, i.e. the ice nuclei concentration is kept constant. Furthermore, we constrain our study and do not include the effect of droplet size on homogeneous freezing. Thereby, if the theoretical model proposed by Rosenfeld et al. (2008) is valid, the simulations should display a monotonic increase of convective strength with increasing aerosol concentration.

The paper is organized as follows. In Sect. 2 we will present the model, the simulated case and the different sensitivity simulation series. Thereafter follows a presentation of the results in Sect. 3. In Sect. 4 the results are summarized and discussed.

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2 Model and simulated case

The dynamics-physics module of the cloud-resolving model (CRM) consists of the non-hydrostatic momentum equations, the continuity equations for water vapor and air mass density, the thermodynamic equation, and the equation of state (Wang and Chang, 1993a). Also included are prognostic equations for the mixing ratios (Q) as well as number concentrations (N) of cloud droplets, raindrops, ice crystals and graupel particles. All unrimed ice crystals and snowflakes are considered as one group (ice crystals) whereas the graupel category considers rimed ice particles as well as frozen raindrops. Every hydrometeor category has a unique spectrum described by the prognostic values of Q and N at each time step. The microphysical transformations are formulated based on a two-moment scheme incorporating the size spectra of particles (Wang and Chang, 1993a; Wang et al., 1995).

In the CRM, the number of CCN available for cloud droplet nucleation is predicted using an aerosol module (cf. Sect. 2.1). The number of ice nuclei (IN) is in the present study assumed to be constant. The number of aerosols available to form cloud droplets is determined by calculating the critical radius corresponding to the critical saturation ratio for droplet activation using the Köhler equation (cf. Ekman et al., 2006). In the model, all hygroscopic aerosols are considered to be potential CCN. The autoconversion of cloud droplet mass concentration (M_c) to rain water mass concentration (M_r) is parameterized according to Berry (1967) as described in Simpson and Wiggert (1969):

$$\frac{dM_r}{dt} = \frac{M_c^2}{60 \cdot \left(5 + \frac{0.336 \cdot N_c}{M_c \cdot D_b}\right)}. \tag{1}$$

Where N_c is the cloud droplet number concentration at cloud base and D_b is the relative dispersion of the spectrum taken to be 0.366 following Simpson and Wiggert (1969) (for maritime clouds). All rain drops formed are assumed to have a typical drop radius of 40 µm. Liu et al. (2007) pointed out that the number autoconversion rate is dependent on the liquid water content, droplet concentration and relative dispersion. In the applied

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parameterization, the autoconversion rate is a function of droplet size which is different from e.g. the commonly used parameterization by Kessler (1969) where the autoconversion is only dependent on cloud droplet mass concentration. Liu et al. (2007) also showed that the number autoconversion rate is not always linearly proportional to the mass autoconversion rate, i.e. that the typical drop radius is not necessarily constant. However, for a relatively high liquid water content ($\gtrsim 0.1\,\mathrm{g\,m^{-3}}$), such as in a deep convective cloud, this approximation appears to be reasonable.

Nucleation of ice crystals occurs both through homogeneous-freezing of liquid particles and by heterogeneous nucleation caused by aerosol particles. All cloud droplets present in a model grid with temperature below $-40\,^{\circ}\mathrm{C}$ are allowed to freeze immediately through homogeneous freezing. The shape of these ice crystals is given as a solid column (cf. Pruppacher and Klett, 1997) and the minimum stable size of them is given as 20 µm on the basis of observations (e.g., McFarquhar and Heymsfield, 1996). Thus, the number concentration of ice crystals formed through the condensation-freezing process can be derived using the condensed water content of frozen cloud droplets. Heterogeneous nucleation of ice crystals is in the model described using the parameterization by Cotton et al. (1986). Aggregated ice crystals are assumed to be converted to graupel if the dimension $D_i \geq D^* = 300\,\mu\mathrm{m}$. The impact of droplet size on ice formation processes is not included in the model.

A δ-four-stream radiation module based on Fu and Liou (1993) is incorporated in the CRM using predicted concentrations of gases (including H_2O and O_3) and hydrometeors to calculate radiative fluxes and heating rates. Six bands for the solar and twelve bands for the thermal part of the radiation spectrum are utilized.

The meteorological-chemical part of the CRM has been applied in several studies of the dynamics, microphysics, and chemistry in continental deep convection (e.g. Wang and Chang, 1993a,b; Wang and Crutzen, 1995) and oceanic deep convection over the Pacific (Wang et al., 1995; Wang and Prinn, 2000, 1998; Wang, 2005a,b). The chemistry submodule predicts atmospheric concentrations of 25 gaseous and 16 aqueous (in both cloud droplets and raindrops) chemical compounds including important aerosol precursors, such as sulfate and nitrate, undergoing more than 100 reactions as well as transport and microphysical conversions. Results of the chemical and dynamical parts of the model have been compared with available observations including aircraft, radar, and satellite data (Barth et al., 2007). The 3-D CRM coupled with the explicit aerosol module has been described and evaluated against observations in Ekman et al. (2004), Ekman et al. (2006), Engström et al. (2008) and Ekman et al. (2008). In the present version of the model, the horizontal resolution of the model is set to 2 km and the vertical grid interval to 400 m. The model domain covers $200 \times 200 \times 50\,\text{km}^3$.

2.1 Aerosol module

The evolution in time and space of the aerosol population is described using a multi-modal aerosol model originally developed by Wilson et al. (2001) and modified as described in Engström et al. (2008) and references therein. In the present study, four different modes are used to represent the aerosol population. These four modes are: nucleation mode aerosols (here defined by $d \leq 23$ nm), Aitken mode aerosols (here defined by 23 nm$\leq d \leq 100.0$ nm), accumulation mode aerosols (here defined by 100 nm$\leq d \leq 900.0$ nm) and coarse mode aerosols (here defined by $d \geq 900.0$ nm). The size distribution within each aerosol mode is assumed to be log-normal and is described by three parameters: number, mass, and standard deviation. To reduce the computational burden, the standard deviation is prescribed (1.59 for all modes). Both number concentrations and mass mixing ratios of the four aerosol modes, i.e. all together 8 variables, are incorporated in the cloud-resolving model as prognostic variables undergoing transport, mixing, dry deposition, and nucleation as well as impaction scavenging besides aerosol microphysical processes. The advection scheme used to calculate the transport of these aerosol variables is a revised Bott scheme as described in Wang and Chang (1993a). The nucleation aerosol mode has a continuous source through the formation of new aerosols from H_2SO_4 (supplied by SO_2 oxidation calculated in the chemistry module of the model) and H_2O (Vehkamäki, 2002). The condensation coefficient as well as the intra- and inter-modal coagulation coefficients for each aerosol mode is determined from the theory of Fuchs (1964), using the geometric mean radius of each mode.

In addition to the nucleation scavenging of aerosols through the formation of cloud droplets (cf. Sect. 2) aerosols can also be scavenged through collision with falling raindrops, graupel or ice crystals, i.e. precipitation (impaction) scavenging. In the present version of the model, graupel and ice scavenging are only considered for nucleation and accumulation mode aerosols whereas impaction scavenging by rain is considered for all aerosols. In the model, the collision efficiency of aerosols with raindrops E varies with size and is prescribed for the different aerosol bins (cf. Ekman et al., 2004). The removal by rain drops is efficient for either small or large particles whereas the collision efficiency for particles in the 0.1 to 1.0 µm size range is relatively low. Recycling of aerosols through evaporation of cloud particles is not treated by default in the CRM, i.e. the aerosols are assumed to be scavenged when they are in droplets or ice crystals. However, in Engström et al. (2008), recycling of aerosols was found to be an important process for representing the aerosol size distribution in the air surrounding a deep convective cloud. Thus, we also examine the aerosol-induced sensitivity of deep convective strength to this process in a separate sensitivity series.

2.2 Simulated case and sensitivity simulation series

The simulated case is a cumulonimbus cloud with extended anvil over the Indian Ocean observed during the INDOEX campaign (Ramanathan et al., 2001) and described further in Engström et al. (2008). Following the definition by Fan et al. (2009), this is a case with relatively weak vertical wind shear and high relative humidity. Initial aerosol concentrations are based on observations and shown in Fig. 1. Sensitivity simulations are conducted for high (as in Fig. 1), medium (50% of the aerosol concentration shown in Fig. 1) and low (25% of the aerosol concentration shown in Fig. 1) pollution levels. The aerosol composition is assumed to be 100% $(NH_4)_2SO_4$. All gas concentrations are initialized as in Engström et al. (2008). The total integration time is 5 h.

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To test how the aerosol-induced sensitivity of deep convection may depend on the complexity of the aerosol model as well as the aerosol activation parameterization, four versions of the CRM are used:

1. *Emp-Const*: Aerosol concentration is given as number of CCN. All aerosols in the accumulation and Aitken mode are assumed available as CCN. The aerosols are not advected or scavenged. All newly formed cloud droplets are assumed to have the same size (radius=1 µm). The number of activated CCN (N_{CCN}) is calculated using the empirical formula: $N_{CCN} = \mathrm{CCN} \cdot S^{k}_{s,w}$, where S is the supersaturation and $k_{s,w}$ is a constant equal to 0.7.

2. *Emp-Adv*: As in simulation *Emp-Const* but the aerosols are advected and scavenged by precipitation as in Wang (2005a).

3. *Aero-Koehler*: Fully interactive aerosol-cloud model as described in Sect. 2.1 and in Ekman et al. (2007).

4. *Aero-Koehler-Eva*: As in *Aero-Koehler* but the recycling of aerosols through evaporation/sublimation of hydrometeors is considered as in Engström et al. (2008).

We also conduct two additional sets of sensitivity simulations to investigate if the size of the aerosols may affect the aerosol-induced sensitivity:

5. *Aero-Koehler-Acc*: Same setup as in *Aero-Koehler* but all changes in aerosol concentration are assumed to take place within the accumulation mode.

6. *Aero-Koehler-Ait*: Same setup as in *Aero-Koehler* but all changes in aerosol concentration are assumed to take place within the Aitken mode.

Note that the total change in aerosol *number* concentration is the same in *Aero-Koehler-Acc* and *Aero-Koehler-Ait*, it is only the size that is different.

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3 Results

All variables are evaluated after 3 h of simulation and the average over the whole time period is used. All changes discussed are significant at a 95% confidence level (using a student t-test) if nothing else is stated. If any significant discrepancy/change can be seen for other evaluation time periods than 3 h, this will be discussed. Figure 2 illustrates that the cloud development is slightly different in the different sensitivity series, but that the main part of the convective event is encompassed when 3 h is used as evaluation time.

3.1 Cloud droplet number concentrations

Comparing the different sets of sensitivity simulations, the one with constant aerosol concentration (*Emp-Const*) clearly activates the largest number of aerosols (Fig. 3). This is not a surprising result as no aerosols are removed by precipitation in this simulation series. The activation of more aerosols in *Emp-Const* leads to a higher liquid water content compared to the other sensitivity series (cf. Sect. 3.3), in particular at levels above 4–5 km, which in turn is reflected in a higher average updraft velocity at higher altitudes as displayed in Fig. 2. The set of simulations including an explicit aerosol cycle (*Koehler-Aero*) activates a larger number of aerosols than the *Emp-Adv* simulations. The empirical formulation requires 1% supersaturation to activate all available CCN (cf. Sect. 2.1). Using the Koehler equation for calculating the effective radius at which aerosols are activated results in that all accumulation mode aerosols and a large part of the Aitken mode aerosols are activated at a lower supersaturation (approx. 0.7%) in *Koehler-Aero*.

Increasing the number of aerosols from low to medium and high concentrations results in an average increase of cloud droplets in all simulations (Table 1). Interestingly, when adding aerosols into the Aitken mode (*Koehler-Aero-Ait*), a higher number of activated aerosols is generated after 3 h of simulation compared to if the additional aerosols are assumed to be of accumulation mode size (*Koehler-Aero-Acc*). This is also true when comparing *Koehler-Aero-Ait* with the series where recycling of aerosols from hydrometeor evaporation/sublimation is considered (*Koehler-Aero-Eva*). For a given updraft velocity, a larger aerosol will consume more water vapor as it grows, both before and after activation. Hence, the supersaturation right below and above cloud base is lower in *Koehler-Aero-Acc* than in *Koehler-Aero-Ait* when using medium and high aerosol concentrations. In addition, the aerosol module does not include scavenging of Aitken mode aerosols by graupel whereas accumulation mode aerosols are efficiently scavenged (cf. Sect. 2.1). As a result, the CDNC concentration increases at all levels where liquid water is present in the *Koehler-Aero-Ait* simulations whereas the increase of CDNC in *Koehler-Aero-Acc* (as well as the other sensitivity simulations) is mainly restricted to the lowest 2 km of the model.

The larger increase of aerosols activated in *Koehler-Aero-Ait* in combination with the higher supersaturation generates more latent heat release which results in higher updrafts and even more activated aerosols. Thereby more cloud water can reach the freezing level in the *Koehler-Aero-Ait* simulations using medium and high aerosol concentrations which in turn generates an additional increase of latent heat release and higher updrafts (cf. next subsection).

3.2 Updraft

The average updraft velocity ($\geq$1 m s^{-1}) generally increases with increasing aerosol concentration in all sensitivity simulations (Fig. 4 and Table 2). This is agreement with the theory presented by both Rosenfeld et al. (2008) and Fan et al. (2009) (for a case with relatively low vertical wind shear and high humidity). However, there are two exceptions that do not follow the conceptual model presented by Rosenfeld et al. (2008):

Firstly, for the *Koehler-Aero-Acc* series, the increase in wind speed between low and medium aerosol concentration is not significant. In addition, the number of grid-points with updraft velocities ($\geq$1 m s^{-1}) is lower when using medium compared to low aerosol concentrations. After 2 h of integration there is a small, but significant, increase in updraft velocity for the *Koelher-Aero-Acc* simulations using medium and high aerosol concentrations but after this time period the efficient depletion of accumulation mode aerosols, mainly due to impaction scavenging by graupel, inhibits the formation of new cloud droplets and thereby the release of latent heat. The results are similar restricting the analysis to updraft velocities larger than 5 ms^{-1}. Examining updraft velocities larger than 0.1 ms^{-1}, there is a signficant decrease in updraft velocity for *Koehler-Aero-Acc* when using medium compared to low concentrations of aerosols.

Secondly, for the simulation *Koehler-Aero-Eva* using high aerosol concentrations, the average updraft is actually slightly lower than the *Koehler-Aero-Eva* using medium aerosol concentrations (the difference is still significant at a 95% confidence level). The number of grid-points with updraft velocities ($\geq$1 ms^{-1}) is also lower. The difference in updraft velocity for the *Koehler-Aero-Eva* simulations using medium and high aerosol concentrations is most pronounced at the beginning of the simulation. Both simulations display higher updraft velocities than the simulation with low aerosol concentration, as more latent heat is released due to more condensation of cloud water. However, the simulation with high aerosol concentrations has more cloud water present than the simulation with medium aerosol concentration (due to the higher CDNC), and this reduces the buoyancy.

Figure 5 shows that for medium aerosol concentrations, *Koehler-Aero*, *Koehler-Aero-Acc Koehler-Aero-Ait* all display an increase in latent heat release between 1 and 6 km altitude after 20 min of simulation. However, after 40 min of simulation, the evaporative cooling is larger than the increase in latent heat release in the *Koehler-Aero-Acc* simulation which suppresses the convection.

Interestingly, the increase in average updraft between the three types of activation formulation varies between 5 and 7% for medium aerosol concentrations and between 11 and 14% for high aerosol concentrations. These differences are all significant, except the difference between the *Emp-Adv* (medium aerosol concentration) and *Koehler-Aero* (medium aerosol concentration) simulations. The simulation where recycling of aerosols is considered (*Koehler-Aero-Eva*) generates a significantly higher (approx. 5%) average updraft velocity for the case with low aerosol concentrations compared to the other *Koehler-Aero* simulations. On the other hand, the percentage increase in updraft velocity with increasing aerosol concentration is lower in *Koehler-Aero-Eva* than in *Koehler-Aero*. After approximately 1 h of simulation, evaporation is larger in the *Koehler-Aero-Eva* series than in e.g. *Koehler-Aero*, which stabilizes the stratification below 5 km and limits the increase in updraft velocity (Fig. 6).

To more carefully examine the impact of graupel scavenging on the accumulation mode aerosols, we conducted an additional set of sensitivity simulations omitting all impaction scavenging of accumulation mode aerosols (simulation series *Koehler-Aero-Acc-Noimpact*. For this set of simulations, the updraft does increase monotonically with increasing aerosol concentration (5.1% and 6.3% for medium and high aerosol load, respectively). It is worthwhile noticing that the increase is still significantly different compared to the *Koehler-Aero-Ait* series. Figure 6 shows that as graupel scavenging of accumulation mode aerosols is removed, enough aerosols are present for new cloud droplets to form (and freeze) and thereby the latent heat release is larger than the evaporative cooling.

3.3 Cloud water

Cloud water content increases with increasing aerosol concentration for all sets of simulations, except in *Koehler-Aero-Eva* where a slight decrease in cloud water content is present after three hours of simulation when comparing high versus medium aerosol concentrations (Fig. 7 and Table 3). However, after only one hour of integration, there is a monotonic increase of cloud water content with increasing aerosol concentration for this simulation series as well. For *Koehler-Aero-Acc*, the increase in cloud water when comparing medium and low aerosol concentrations is not significant. If impaction scavenging of accumulation mode aerosols is removed (series *Koehler-Aero-Acc-Noimpact*), the formation of graupel increases significantly with increasing aerosol concentrations (24 and 50% for medium and high aerosol concentrations, respectively).

In general, the increase is substantially larger (43–76%, comparing high and low

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aerosol concentrations) for the simulations including a full aerosol cycle (*Koehler-Aero*, *Koehler-Aero-Eva*, *Koehler-Aero-Acc* and *Koehler-Aero-Ait*) than for the simulations using a simplified description of the aerosol population (13–23% increase). The simulations *Emp-Const* and *Emp-Adv* contain more cloud water in the simulation with low aerosol concentration than *Koehler-Aero*. This can be explained by the fact that for the empirical simulations, all cloud droplets have the same equilibrium size (cf. Sect. 2.1). *Koehler-Aero-Ait* displays more cloud water in the reference (low aerosol concentration) simulation than *Koehler-Aero* and *Koehler-Aero-Acc*, which is reasonable considering the larger number of aerosols activated (cf. Fig. 3).

3.4 Graupel

All simulation series, except *Koehler-Aero-Acc* and *Koehler-Aero-Eva*, display an increase of graupel with increasing aerosol concentrations (Fig. 8 and Table 4). The increase is small but significant when using high compared to medium aerosol concentrations for *Emp-Const* and *Koehler-Aero*. For the *Koehler-Aero-Eva* series, a slight decrease can be noted when comparing high versus medium aerosol concentrations which is consistent with the slight decrease in updraft velocity and cloud water formation.

For *Koehler-Aero-Acc* there is a significant decrease in graupel amount when using medium aerosol concentrations. Comparing *Koehler-Aero-Acc* and *Koehler-Aero*, the development of graupel is relatively similar up to approximately 1–1.5 h of integration. After this time period, the convection in *Koehler-Aero-Acc* is suppressed (cf. Sect. 3.2) and thereby also the graupel formation. If impaction scavenging of accumulation mode aerosols is removed, then the graupel content increases with increasing aerosol concentration (by 13 and 24% for medium and high aerosol concentrations, respectively).

3.5 Rain

For *Emp-Const* and *Emp-Adv*, the amount of rain at the lowest model level decreases with increasing aerosol concentration, despite the increase in vertical wind speed and graupel formation (Fig. 9 and Table 5). The decrease in rain rate occurs due to more efficient evaporation of rain drops below the freezing level and, to some extent, also due to the less efficient warm rain formation. Above 5 km, the rain drop mass actually increases with increasing aerosol concentration (not shown).

The rain formation at the lowest model level increases with increasing aerosol concentration in *Koehler-Aero* and *Koehler-Aero-Ait*, but the increase peaks at medium aerosol concentrations. The difference in rain formation between medium and high aerosol concentration is not significant for *Koehler-Aero-Ait* and only significant at a 90% confidence level for *Koehler-Aero*. For high aerosol concentrations, the rain formation above 5 km still increases in both these simulation, but the evaporation of rain drops below the freezing level increases. A higher aerosol load is required for a significant increase in rain formation to occur in *Koehler-Aero-Acc* compared to *Koehler-Aero* and *Koehler-Aero-Ait*. For medium aerosol concentrations, the amount of rain at the lowest model level actually decreases compared to the simulation with low aerosol concentrations in *Koehler-Aero-Acc*, which is consistent with the lower rate of graupel formation. If impaction scavenging of accumulation mode aerosols is removed, then the rain rate will increase with increasing aerosol concentrations (by 9 and 25% for medium and high aerosol concentrations, respectively).

4 Summary and conclusions

In the present study, we utilize a cloud-resolving model to examine aerosol-induced sensitivity of deep convective strength for a single cloud case with relatively weak vertical wind shear and high relative humidity. To simplify the analysis, we only consider aerosol effects on liquid droplet formation, i.e. the impact of aerosols (and cloud droplet size) on freezing processes is not included. We examine how the aerosol-induced sensitivity may depend on the complexity of the aerosol model as well as the aerosol activation parameterization. We also investigate if the size of the aerosols may affect the aerosol-induced change in deep convective cloud properties. In total, we conduct six sets of sensitivity simulations using low, medium and high aerosol concentrations.

For the analysis of the model simulations, the average updraft is taken as a measure of the deep convective strength. We find that, in general, the model study supports the conceptual model that increased aerosol concentrations result in stronger convection as outlined by Rosenfeld et al. (2008) (and if only aerosol effects of liquid droplet formation is considered). However, there are a two main exceptions where the updraft does not increase monotonically with increasing aerosol concentrations. Both these exceptions illustrate the need to account for changes in evaporation processes when considering aerosol effects on convective strength and precipitation. In addition, one sensitivity series shows that graupel scavenging may efficiently deplete the number of aerosols suitable as CCN, which limits the formation of cloud droplets and thereby the release of latent heat. It is also worthwhile noting that a decrease in buoyancy, resulting from an increase in aerosol concentration and thereby cloud water, may take place already during the initial stages of cloud development before any freezing is initiated.

The sensitivity simulations show that the complexity of the aerosol model and the choice of aerosol activation parameterization significantly impacts on the aerosol-induced convective cloud sensitivity. When using different aerosol activation parameterizations, the increase in vertical wind speed (comparing high and low aerosol concentrations) varies between 11 and 14% if wind velocities $\geq 1\,\mathrm{m\,s^{-1}}$ are considered and between 10 and 19% if wind velocities $\geq 5\,\mathrm{m\,s^{-1}}$ are considered. Regarding the complexity of the aerosol model, graupel scavenging of aerosols appears to be a crucial process as it efficiently removes aerosols at a critical time point of the cloud evolution. However, this process is currently poorly understood and often described in a simplified manner in models. Recycling of aerosols through cloud droplet and ice crystal evaporation/sublimation is also found to be an important process for the aerosol-induced convective cloud sensitivity. For example, when aerosol recycling is considered, the convective strength reaches a maximum at medium instead of high aerosol concentrations and the sensitivity of the deep convective strength is significantly smaller.

An increased number of aerosols in the Aitken mode (here defined by 23 nm$\leq d \leq$ 100.0 nm) may result in a larger impact on the convective strength compared to an increased number of aerosols in the accumulation mode (here defined by 100 nm$\leq d \leq$ 900.0 nm). As accumulation mode aerosols are activated and grow in the beginning of the cloud cycle, the supersaturation near the cloud base is lowered. Thereby, more aerosols in the Aitken mode are activated compared to in the accumulation mode during the active phase of the convective cloud development, which in turn results in a larger increase of the convective updraft.

The sensitivity simulations show that the change in graupel and rain formation is not always directly proportional to the change in updraft velocity. Several of the sensitivity simulations display a decrease of the rain amount at the lowest model level with increasing updraft velocity. This decrease was generally caused by an increased evaporation of rain drops. As the response of the convection (both in terms of strength and in terms of precipitation) was found to be significantly different when using different complexity of the aerosol model and aerosol activation parameterization, it underlines the importance of better understanding the two-way interaction between aerosols and clouds. The results may also to some extent explain why different cloud-resolving model studies have shown different results regarding aerosol-induced deep convective cloud sensitivity.

The relatively small differences in convective strength obtained for all sensitivity simulations comparing medium and low aerosol concentrations suggest that single deep convective clouds developing in an environment with weak vertical wind shear and high relative humidity are relatively insensitive to changes in aerosol concentration. This finding corroborates the results by Fan et al. (2009) and suggests that aerosol effects on isolated deep convective clouds are more likely to be distinguished in dry environments with large vertical wind shear.

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Table 1. Percent change in cloud droplet number concentration (compared to simulation with low aerosol concentration) for the different sensitivity simulations.

	Medium [%]	High [%]
Emp-Const	71	191
Emp-Adv	61	168
Koehler-Aero	71	191
Koehler-Aero-Eva	66	212
Kohler-Aero-Acc	61	168
Koehler-Aero-Ait	300	567

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Table 2. Percent change in updraft (compared to simulation with low aerosol concentration) for the different sensitivity simulations.

	Medium [%]	High [%]
Emp-Const	4.9	11.2
Emp-Adv	5.9	14.0
Koehler-Aero	7.2	13.0
Koehler-Aero-Eva	2.7	2.4
Koehler-Aero-Acc	1.3	10.1
Koehler-Aero-Ait	7.6	10.8

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Table 3. Percent change in cloud water (compared to simulation with low aerosol concentration) for the different sensitivity simulations.

	Medium [%]	High [%]
Emp-Const	9.6	13.1
Emp-Adv	10.6	23.0
Koehler-Aero	43.5	54.6
Koehler-Aero-Eva	48.7	43.0
Kohler-Aero-Acc	1.9	68.1
Koehler-Aero-Ait	58.3	75.9

Table 4. Percent change in graupel (compared to simulation with low aerosol concentration) for the different sensitivity simulations.

	Medium [%]	High [%]
Emp-Const	30.7	31.3
Emp-Adv	15.9	27.3
Koehler-Aero	30.1	30.5
Koehler-Aero-Eva	13.1	7.6
Kohler-Aero-Acc	–5.4	34.4
Koehler-Aero-Ait	30.1	48.4

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Table 5. Percent change in rain water (compared to simulation with low aerosol concentration) at model level 1 for the different sensitivity simulations.

	Medium [%]	High [%]
Emp-Const	−10.1	−19.7
Emp-Adv	−11.9	−10.5
Koehler-Aero	34.1	15.9
Koehler-Aero-Eva	42.5	35.0
Kohler-Aero-Acc	−4.6	38.6
Koehler-Aero-Ait	38.6	31.2

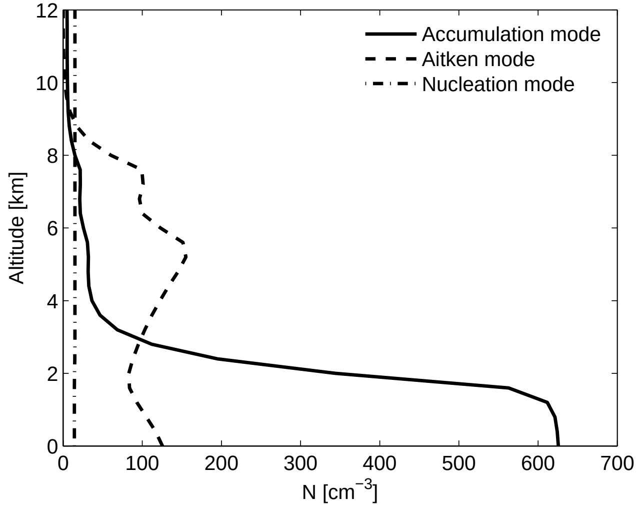

Fig. 1. Vertical profiles of initial aerosol number concentrations (high aerosol concentration case).

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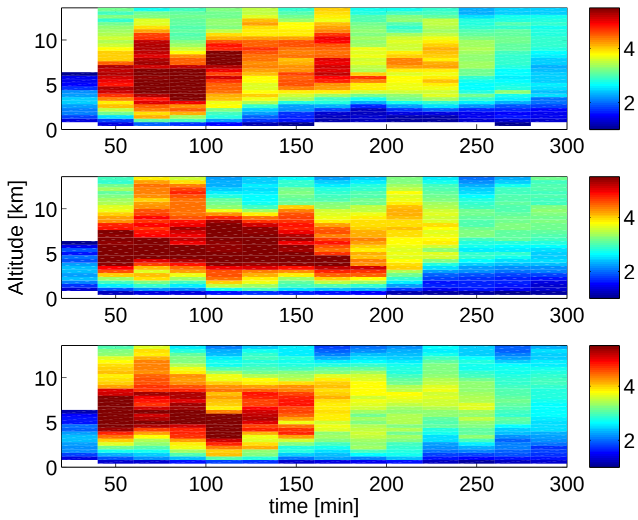

Fig. 2. Horizontally averaged vertical wind speed ($\geq 1\,\mathrm{m\,s^{-1}}$) during the 5-h simulation using high aerosol concentrations. Emp-Const (top panel), Emp-Adv (middle panel) and Koehler-Aero (bottom panel).

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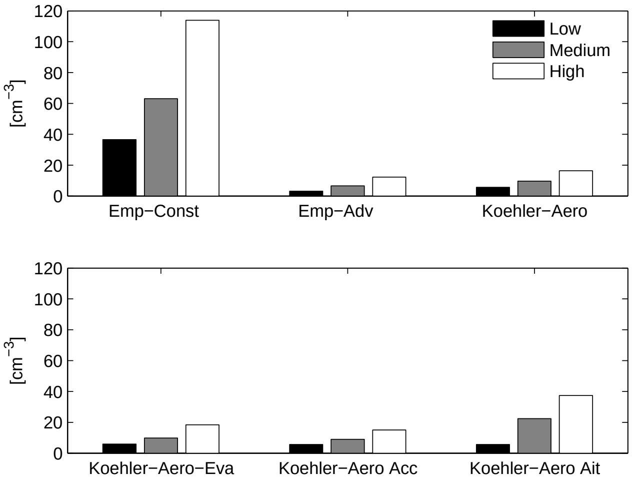

Fig. 3. Domain- and time-averaged cloud droplet number concentration after 3 h of simulation.

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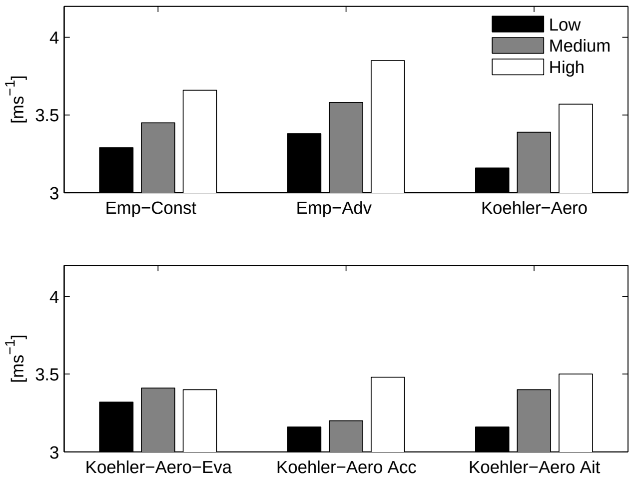

Fig. 4. Domain- and time-averaged vertical wind speed (≥1 ms^{-1}) after 3 h of simulation.

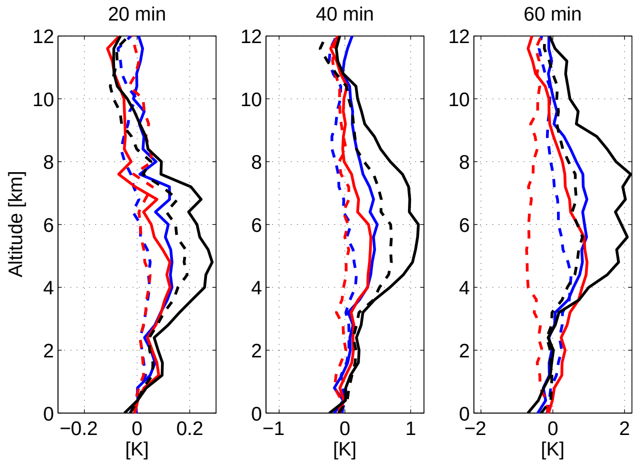

Fig. 5. Domain averaged difference in temperature increase/decrease after 20, 40 and 60 min integration time for the simulations using medium (dashed lines) and high (full lines) aerosol concentration (compared to the case with low aerosol concentrations). *Koehler-Aero* (blue line), *Koehler-Aero-Acc* (red line) and *Koehler-Aero-Ait* (black line). Note the different scales on the x-axis.

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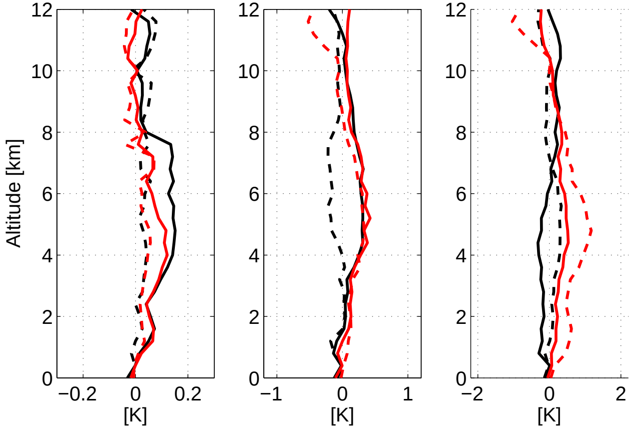

Fig. 6. Domain averaged difference in temperature increase/decrease after 20, 40 and 60 min integration time for the simulations using medium (dashed lines) and high (full lines) aerosol concentration (compared to the case with low aerosol concentrations). *Koehler-Aero-Eva* (black line) and *Koehler-Aero-Acc-Noimpact* (red line). Note the different scales on the x-axis.

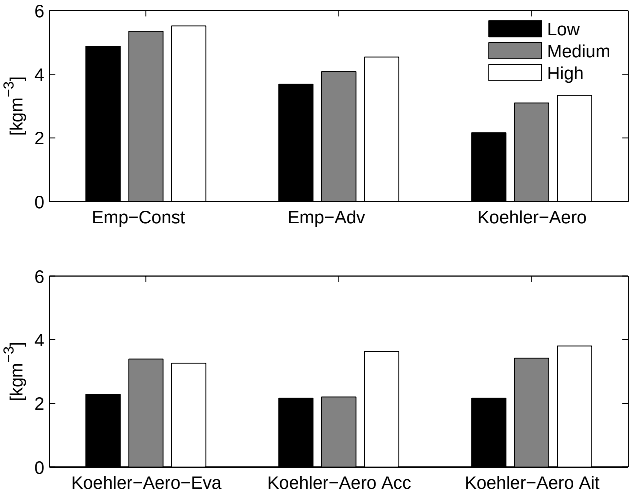

Fig. 7. Domain- and time-integrated sum of cloud water after 3 h of simulation.

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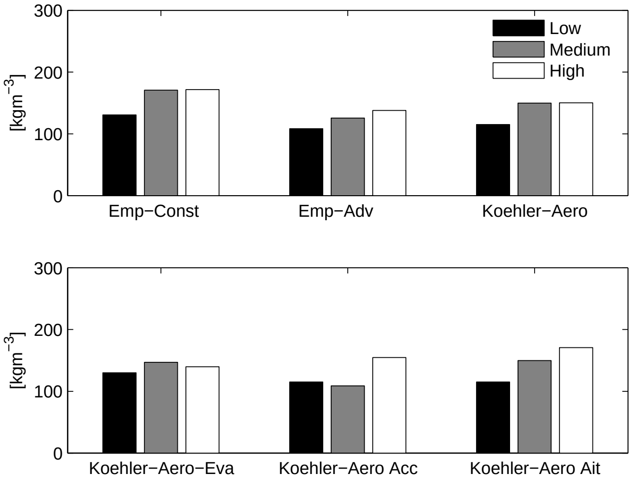

Fig. 8. Domain- and time-integrated sum of graupel after 3 h of simulation.

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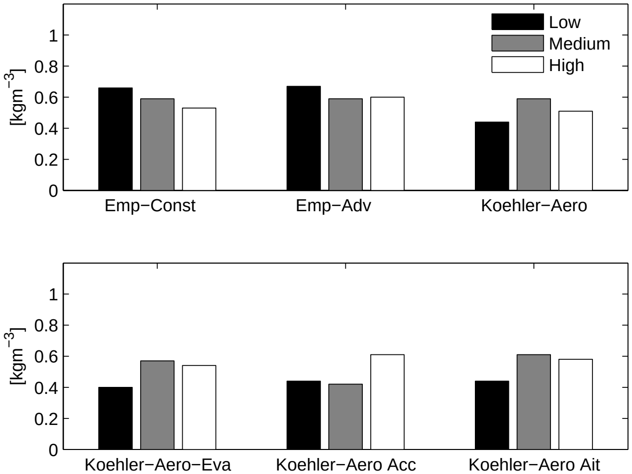

Fig. 9. Domain- and time-averaged sum of rain water at model level 1 after 3 h of simulation.

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