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## Notes on state-of-the-art investigations of aerosol effects on precipitation: a critical review

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### Abstract

There is no agreement between the results of different studies as regards quantitative and even qualitative evaluation of aerosol effects on precipitation. While some observational and numerical studies report a decrease in precipitation in polluted areas, in some other observations and numerical studies aerosol-induced precipitation enhancement was reported.

This study analyses possible reasons for the discrepancy between the results. The analysis of aerosol effects on precipitation is performed using the mass and heat budgets. The analysis is concentrated on clouds and cloud systems arising in the environment with relatively high freezing level. It is shown that for such clouds aerosols increase both the generation and the loss of the condensate mass. The net effect of aerosols on the precipitation depends on the environment conditions (air humidity, buoyancy, and wind shear) as well as on the cloud type determining whether the increase in the condensate generation or in the condensate loss will dominate with increase in the aerosol concentration. In the case when the loss increases more than the generation, a decrease in precipitation will take place. If the increase in the condensate generation dominates, an increase in precipitation will take place. A classification scheme of aerosol effects on precipitation is proposed and its relation to the observational and numerical results available is analysed. Possible reasons for the uncertainties and discrepancies of the numerical results, as well as between measurements, are analysed. A discussion of unsolved problems is presented in the conclusion.

**Keywords:** aerosol effects on cloud microphysics and precipitation, precipitation efficiency, deep convection, numerical modelling

### 1. Introduction

Observations and numerical studies indicate that atmospheric aerosols influence the microphysical structure and precipitation formation in stratocumulus and cumulus clouds. An increase in the concentration of submicron aerosol particles (AP) serving as cloud condensation nuclei (CCN) increases the concentration and decreases the size of droplets (e.g., Twomey 1974, Albrecht 1989, Rosenfeld and Lensky 1998, Ramanathan *et al* 2001, Andreae *et al* 2004). Numerical models with an accurate spectral bin microphysics approach made it possible to reproduce the observed drop–aerosol

concentration dependences (e.g., Segal and Khain 2006, Kuba and Fujiyoshi 2006, Pinsky *et al* 2008b).

At the same time, the effect of aerosols on precipitation still remains a challenging problem (see the detailed overview by Levin and Cotton 2007). There is no agreement between the results of different studies as regards quantitative and even qualitative evaluation of aerosol effects on precipitation. While some observational (e.g., Albrecht 1989, Rosenfeld 1999, 2000, Borys *et al* 2000, Hudson and Yum 2001, McFarquhar and Heymsfield 2001, Yum and Hudson 2002, Givati and Rosenfeld 2004, Jirak and Cotton 2006, Hudson and Mishra 2007, Göke *et al* 2007, Xue and Feingold 2006) and numerical

(e.g., Khain and Pokrovsky 2004, Feingold et al 2005, Teller and Levin 2006, Iguchi et al 2008, Altaratz et al 2008) studies report a decrease in precipitation in polluted clouds, in some other observations (Lin et al 2006) and numerical studies (Wang 2005, Van den Heever et al 2006, Lynn et al 2005a, 2005b, Fan et al 2007a, 2007b, Lee et al 2005, 2008, Khain et al 2008b, Martins et al 2008, Li et al 2008b, 2008c) aerosolinduced precipitation enhancement was reported. In several studies (e.g., Khain et al 2003, 2004, 2005, 2008a, Kaufman et al 2005, Lynn et al 2005a, 2005b, 2007, Lynn and Khain 2007, Matsui et al 2006, Seifert and Beheng 2006, Tao et al 2007, Lee et al 2005, 2008) it was shown that aerosol effects on the precipitation amount depend on the environment conditions and on the cloud type (e.g. maritime stratocumulus clouds, single isolated cumulus clouds, severe storms, squall lines, supercell storm versus multi-cell storms, etc). Because of many uncertainties in the observations (see appendix A) and numerical modelling (see appendix B), the current stage of investigations in the topic, especially as regards quantitative evaluations, can be defined as knowledge at the accumulation stage.

In spite of the importance of the problem and a lot of publications, there have only been a few attempts to reveal the physical reasons for the different response of clouds to aerosols and to classify the observational and numerical results (Khain *et al* 2008a). This paper aims at partially closing the gap. We would like to stress that the effect of aerosols on precipitation is a very complicated multi-scale problem, so that any attempts at classification will inevitably be too simplified *a priori*. In this study we analyse the effects of small submicron aerosol particles (AP) playing the role of CCN. An important goal of the paper is to attract the attention of investigators (especially modellers) to the methodological aspects of the problem.

We will mostly concentrate on clouds and cloud systems developing in the environment with high (about 4 km) freezing level. In many cases these clouds have a warm cloud base. In spite of the fact that the role of giant CCN and ice nuclei (IN) on precipitation has been stressed in several studies (e.g., Rosenfeld *et al* 2002, Teller and Levin 2005, Van den Heever *et al* 2006), their effects on precipitation will not be analysed in the study because of many uncertainties as regards their concentration under different environmental conditions and the lack in understanding the mechanisms of their effect on precipitation. This problem is discussed in more detail in section 5. Preliminary results as regards the aerosol effects on clouds with low freezing level and relatively cold cloud base are presented in section 5 as well.

#### 2. Budget considerations

### 2.1. Governing equations

Aerosols represent only one of many factors affecting precipitation. Moreover, in many cases the aerosol factor it is not the strongest one. For instance, daily changes in atmospheric instability can determine the type and size of clouds and change precipitation by orders of magnitude. An increase in relative humidity (RH) by only 10% can increase precipitation two-three times (Fan *et al* 2007b). An increase in the boundary layer temperature by 1 °C can increase precipitation and the kinetic energy of hail at the surface two times (Khain *et al* 2008d). At the same time, the effect of aerosols on precipitation from deep convective clouds is usually evaluated as a few tens of per cent. The interest in aerosol effects is related to the continuous production of anthropogenic AP which may lead to long lasting trends in precipitation in different regions. There are no clouds (or cloud systems) in nature which differ only by the aerosol environments. This makes the observation of aerosol effects on precipitation from isolated clouds a difficult task. Experimental estimation of aerosol effects over long time periods and large spatial scales also represents a hard mathematical problem (see appendix A for more detail).

The numerical modelling of convective clouds and cloud systems is an efficient tool allowing independent assessment of the role of aerosols in precipitation suppression/enhancement. At the same time, the models introduce their own set of technical and methodological problems (see appendix B). One of the sources of errors in the evaluation of aerosol effects on precipitation is related to ignoring the budget considerations.

The equation for the condensate mass conservation (liquid + solid) M within a vertical column of the atmosphere can be written as:

$$\frac{\partial M}{\partial t} = \delta G - \delta L - \delta P + \delta F_{\rm lb},\tag{1}$$

where  $\delta L$ ,  $\delta P$ , and  $\delta F_{lb}$  are the rates of change of M by drop condensation and ice deposition (the term will be referred to as 'generation', G), evaporation and sublimation (the loss term, L), precipitation to the surface and fluxes via lateral boundaries, respectively.

The integration of (1) with respect to time from t = 0 to t gives

$$M - M_0 = G - L - P + F_{\rm lb}$$
(2)

where  $M_0$  is the mass of condensate in the atmosphere at t = 0, and G, L, P, and  $F_{lb}$  are the generation, the loss, precipitation, and the condensate flux through the lateral boundaries, respectively, accumulated over the time period t. From (2), the precipitation at the surface accumulated during the time period can be written as

$$P = G - L - M + M_0 + F_{\rm lb}.$$
 (3)

The terms on the right-hand side of equation (3) can be of the same order of magnitude, so precipitation at the surface is often just a small difference of large values. This represents one of the difficulties as regards quantitative evaluation of precipitation both in measurements and numerical simulations.

Note that the condensate generation G leads to drying and heating of the atmosphere, while the loss L leads to the moistening and cooling. If precipitation falls to the surface, this means that the atmosphere becomes warmer (net heating) and dryer.

Aerosols affect all items of the moisture budget (3). Let  $\Delta()$  be the changes of terms (3) due to aerosol effects. From (3) it follows that

$$\Delta P = \Delta G - \Delta L + \Delta M + \Delta F_{\rm lb}.\tag{4}$$



**Figure 1.** The fields of rain water content calculated for thermodynamic conditions typical of the Eastern Mediterranean for high AP concentration (left) and low AP concentration (middle). The difference of the fields is presented in the right panel. The dashed line denotes the location of the coast line. The sea area is to the left from this line, the land is to the right. One can see that an increase in the AP concentration leads to a shift of rain from the sea (where negative deviation is seen) to the land.

Note that equations (3) and (4) are of general character and do not depend on the particular mechanisms causing the generation, loss, or transport of condensate (both liquid and solid).

To our knowledge, there are no observational studies in which the budget consideration would be applied for evaluation of aerosol effects on precipitation. The number of numerical studies in which a condensate budget is explicitly applied for such analysis is also quite limited. At the same time, neglecting the budget considerations can lead to errors in the evaluation of aerosol effects. As an example of such possible errors we present results of precipitation calculations in the Eastern Mediterranean during the cold (rain) season. The characteristic climatic feature of this region during the cold season is the existence of a sea-land temperature difference of  $\sim$ 6–7 °C, which induces the local land breezelike circulation. Interaction of this local circulation with the westerly background wind creates convergence in the boundary layer over the sea 10-20 km from the coastline. Clouds formed within this zone are transported eastwards by the background westerly wind. The simulations have been performed using the two-dimensional (2D) Hebrew University cloud model (HUCM) with spectral (bin) microphysics allowing calculation of size distribution functions of drops and ice particles of different types (Khain et al 2004, 2008a). The computational area was of 180 km  $\times$  17 km and located in the east-west direction (perpendicularly to the coastal line). The sea surface and the land surface temperatures were assumed equal to 20 °C and 13 °C, respectively. These temperatures are typical of December in this region. The sounding data have been chosen as typical of this time. The initial air temperature difference over the sea and over the land decreased with height and tended to zero at the top of the computational area (17 km). The westerly background wind increased linearly from 4 m s<sup>-1</sup> near the surface to 20 m s<sup>-1</sup> at z = 10 km, and was assumed to be constant above this level. The evolution of clouds was simulated during a period of 4 h. The simulations have been performed under two concentrations

of cloud condensational nuclei (CCN) (at supersaturation of 1%): 100 and 2000  $\text{cm}^{-3}$ . The initial aerosol concentrations were horizontally homogeneous and constant within the lower 2 km layer. Above this level the CCN concentration decreased exponentially. The sea-land temperature difference triggers cloud formation over the sea  $\sim 10$  km from the coastal line. Figure 1 shows the fields of rain water content obtained in the simulations with high (left panel) and low (middle panel) CCN concentrations. The difference between these fields is shown in the right panel. The delay in the raindrop formation and additional formation of ice particles with low settling velocity caused by the increase in the AP concentration leads to the spatial shift of precipitation from the sea to the land. As a result, the precipitation over the sea decreases at the expense of precipitation increase over the land, so the accumulated precipitation over the whole area changes only slightly. In the case when the computational area is located over the sea, the results could be wrongly interpreted as a significant aerosolinduced decrease in precipitation. The conclusion would be the result of neglecting the aerosol-induced change of the condensate flux through the right lateral boundary (the term  $\Delta F_{\rm lb}$  in equation (4)). In spite of the triviality of the example, such errors in the conclusions are regularly made both in measurements and numerical simulations (see the appendices). Similarly wrong conclusions can be made in the simulations performed for time periods shorter than the cloud life time when a significant mass of the condensate remains suspended in air to the end of simulation. In this case the incorrect evaluation of aerosol effects on precipitation is caused by neglecting the term  $\Delta M$  in (4).

Hence, we will analyse mainly the results of only those numerical simulations which have been performed using comparatively large computational areas (with the horizontal size larger than ~100 km) and during a comparatively long time period (longer than ~2 h). In these simulations clouds either do not reach the lateral boundaries during their lifetime or/and a quasi-stationary state is reached, so one can expect that  $\Delta P \gg \Delta M$  and  $\Delta P \gg \Delta F_{\rm lb}$  in the simulations. In this

case the budget equation simplifies to

$$P \approx G - L;$$
 and  $\Delta P \approx \Delta G - \Delta L.$  (5)

Applying conditions (5) we define the precipitation efficiency PE as the following ratio (Smith 2003, Jiang and Smith 2003, Smith *et al* 2005, Khain *et al* 2005, 2008a, Levin and Cotton 2007):

$$PE = \frac{P}{G}.$$
 (6)

In the case when a cloud (or a cloud system) disappears towards the end of the simulation, the PE calculated as the ratio of the accumulated surface precipitation to the accumulated condensate can be used to characterize the integral response of the system to aerosols. The PE can also be regarded as a function of time, PE(t), where P(t) and G(t) are the time accumulated (from t = 0 to t) precipitation and generation, respectively.

The changes of PE induced by aerosols can be written as

$$\Delta PE = \frac{\Delta P}{G} - PE \frac{\Delta G}{G},\tag{7}$$

$$\frac{\Delta PE}{PE} = \frac{\Delta P}{P} - \frac{\Delta G}{G}.$$
(8)

Note that the precipitation efficiency is often defined as the ratio of the precipitation amount to the vapour flux at the cloud base (e.g. Hobbs *et al* 1980, Ferrier *et al* 1996, Wang 2005, Sui *et al* 2007). The latter definition does not follow from the mass budget. This definition came supposedly from traditional convection parameterizations assuming that water vapour ascends within clouds only, and totally condensates reaching the cloud top. In the case when the assumption is valid, this definition is similar to that given in (6).

### **3.** Effect of aerosols on the items of the heat and condensate mass budgets

To clarify the physical mechanisms by means of which aerosols affect precipitation, it is necessary first to understand how the increase in the aerosol loading affects the items of the condensate mass and heat budgets. We illustrate the effect using the results of simulation of deep convective clouds typical of maritime tropical conditions, intermediate, and extreme continental conditions. As an example of deep convective clouds typical of maritime tropical conditions we use those observed during Experiment GATE-74, 261 day (hereafter, M clouds) (Warner et al 1980, Ferrier and Houze 1989, Khain *et al* 2005). As an example of extremely continental clouds we have chosen clouds typical of summertime Texas (hereafter, T clouds) (Rosenfeld and Woodley 2000, Khain et al 2001, Khain and Pokrovsky 2004). Simulations under maritime thermodynamic conditions were also performed for high concentration of small AP (run Mc), and simulations for extremely continental dry conditions were performed for low AP concentration (run T-m). In order to describe aerosol effects on clouds arising under moderate conditions, the clouds observed during the LBA-SMOCC campaign (The Large Scale Biosphere-Atmosphere

Experiment in Amazonia-Smoke, Aerosols, Clouds, Rainfall and Climate) on 1 Oct, 19 UT (10S 62W) and 4 Oct, 19 UT (10S 67W) in clean and polluted conditions (respectively, green-ocean (GO) and smoky (S) clouds) were simulated (Andreae et al 2004). In the study we also discuss some results of simulations of a hail storm in southern Germany under different AP concentrations (Khain et al 2008d). The specific feature of the latter case is a comparatively low freezing level (2.5 km). As a result, cold precipitation plays the main role under all AP concentrations. The simulation will be referred to as HS (Hail Storm). The conditions for all simulations are presented in table 1. Since the thermodynamic conditions were assumed to be similar in the GO and S cases, as well as in the M and M-c and in the T and T-m cases, the differences between the cloud characteristics in the simulation pairs can be attributed to aerosol effects only. The simulations were performed using the HUCM with the computational area 170 km  $\times$  17 km and a resolution of 350 m in the horizontal and 125 m in the vertical directions, respectively. The model, the soundings used and the results of the simulations are described in more detail by Khain et al (2008b, 2008a). All calculations were performed for a time period of 4 h. In all cases (except the hail storm in Germany) the clouds decayed after 2–3 h, so no condensate mass remained in the air to the end of the simulations. In the simulation of the hail storm a quasi-stationary state in storm evolution has been attained, characterized by the permanent decay of old and formation of new cloud cells. The maximum updrafts varied from about 20 to 30 m s<sup>-1</sup>, with the average maximum velocity of 26 m s<sup>-1</sup>. The peak values of updrafts are related to new cloud cells. In the quasi-stationary state the maximum values (as well as the minimum and averaged values) of the vertical velocity did not change with time, i.e. new cloud cells were of nearly the same intensity as the previous ones. The accumulated precipitation increased with time linearly, indicating the nearly constant precipitation rate at the surface. In the simulations, clouds did not reach the lateral boundaries. Hence, the budget equations can be written in the form (5).

Figure 2 shows the vertical profiles of horizontally averaged (per grid of the computational area) moistening and drying of the atmosphere for the 4 h period in the simulations of the M, T, GO, and S clouds. One can see that in the clouds developing in polluted air the condensate generation G (condensation + ice deposition), as well as the condensate loss L (drop evaporation + ice sublimation) exceed those in the clouds developing in clean air. The net aerosol effect on the heat and moisture budgets can be seen in figure 3, where the vertical profiles of horizontally averaged (per grid of the computational area) moistening/drying (upper panels) and heating/cooling (lower panels) and over the 4 h for the M and T cloud (left panels) and the GO and S cloud (right panels) cases. The differences between moistening and drying, as well as between heating and cooling, represent the net effects, which are shown as well (solid lines). The areas marked in black show the zones where greater moistening and cooling takes place in the polluted clouds; the areas marked in grey denote zones where greater moistening and cooling takes place in clean air. A larger area of black

**Table 1.** The list of simulations and parameters characterizing aerosol distributions; the initial CCN distribution was calculated using the formula  $N_{ccn} = N_o S^k$ , where  $N_{ccn}$  is the concentration of activated AP (nucleated droplets) at the supersaturation *S* (in %) with respect to water,  $N_o$  and *k* are the measured constants.

Type of cloud	Short title	$N_0 ({\rm cm}^{-3})$	k	References
Green-ocean clouds	GO cloud	(a) 100 (b) 400	0.92	Andreae <i>et al</i> (2004), Roberts <i>et al</i> (2002) and Rissler <i>et al</i> (2004)
'Smoky' clouds	S	6880	0.718	The same
Texas continental clouds with high AP concentration	T cloud	2500	0.308	Rosenfeld and Woodley (2000), Khain <i>et al</i> (2001, 2004, 2005), Khain and Pokrovsky (2004), Khain <i>et al</i> (2008a)
Texas sounding, low AP concentration	T-m cloud	100	0.921	Sensitivity study
GATE-74 deep maritime cloud	M cloud	100	0.921	Khain <i>et al</i> (2001, 2004, 2005)
Maritime sounding, but high CCN concentration	M-c cloud	2500 100	0.308 0.921	Sensitivity study. CCN distribution was represented as a sum of continental and maritime distributions
Maritime cloud, but with lower <sup>a</sup> relative humidity	M-80	100	0.921	Sensitivity experiment
Maritime cloud, but with lower relative humidity and high AP concentration	M-c-80	2500 100	0.308 0.92	Sensitivity study. CCN distribution was represented as a sum of continental and maritime distributions
Orographic clouds, low AP concentration	MAR, Mar-RH90	250	0.462	Lynn <i>et al</i> (2007)
Orographic clouds, high AP concentration	CON, Con-RH90	1250	0.308	Lynn <i>et al</i> (2007)
Mid-latitude hail storm	HS	100-6000	0.5	Khain et al (2008d)

<sup>a</sup> In M-80 relative humidity (RH) was by 10% lower over the whole atmosphere as compared to

GATE-74 conditions, so the RH at the surface was 80% instead of 90%.

zones indicates higher net atmospheric moistening and cooling, i.e. a decrease in precipitation in polluted clouds. Figure 4 shows the profiles similar to those shown in figure 3, but for the simulations of clouds with maritime sounding and low (M) and high (M-c) aerosol concentration. The Mc cloud heats and dries the atmosphere to a higher degree than the M cloud, i.e. the precipitation from the tropical maritime deep clouds increases in dirty air. Contrary to this, the precipitation from clouds developing in the comparatively dry continental atmosphere decreases with the increase in the aerosol loading. Note that while the differences between the M and M-c simulations (as well as between GO and S simulations) can be directly attributed to differences in aerosol concentrations, the differences between M and T in figure 3 cannot be attributed to aerosol concentrations alone because of different environmental conditions. We present these panels to demonstrate the difference in the budgets between deep maritime and continental clouds.

The following important conclusion can be derived from the analysis of figures 2-4: an increase in aerosol concentration leads to the increase in both the generation (*G*-term) and

the loss (L-term) of the hydrometeor mass, i.e.,  $\Delta G > 0$ and  $\Delta L > 0$ . This conclusion seems to be quite general for clouds and clouds systems with high freezing level and was found in all our simulations independent of the cloud type and environmental conditions (e.g., Khain et al 2005, 2008a). The increase in the condensate mass with the aerosol loading (which has been reported in most recent studies) can be attributed to the fact that, when the AP concentration is high, droplets are small, and continue ascending to higher levels, and grow by condensation, intensifying the formation of ice growing by deposition. The increase in the condensate generation with aerosol loading indicates the increase in convective heating by latent heat release. A higher condensate loss by sublimation and evaporation in the case of larger aerosol loading can be attributed to the fact that cloud particles in polluted clouds are as a rule smaller and fall from higher levels than those in clouds forming in clean air. Moreover, in the presence of the vertical shear of the background flow, the cloud particles tend to fall outside from the cloud updrafts, through a comparatively dry air, which increases the sublimation and evaporation. At the same time, raindrops in



**Figure 2.** Vertical profiles of horizontally averaged (per grid of the computational area) moistening and drying of the atmosphere for a period of 4 h in simulations of M, T, GO, and S clouds (adopted from Khain *et al* 2008a). One can see that in clouds developing in polluted air, condensation and deposition, as well as evaporation and sublimation, are larger than in clouds developing in clean air.

maritime clouds fall from lower levels within cloudy humid air, so the precipitation loss is relatively low. Thus, with other conditions being similar, polluted clouds must have higher vertical updrafts and downdrafts than clouds developing in clean air. This convective invigoration that is sometimes accompanied by the increase in the cloud top height has been reported in many recent observational and numerical studies (Koren *et al* 2005, Khain *et al* 2003, 2005, 2008b, 2008a, Lynn *et al* 2005a, 2005b, Van den Heever *et al* 2006, Fan *et al* 2007a, 2007b, Lee *et al* 2005, etc)<sup>1</sup>. The net effect of aerosols on precipitation is determined by the relationship between the condensate production  $\Delta G$  and the condensate loss  $\Delta L$ . The condition  $\Delta L > \Delta G$  means a decrease in precipitation, while the conditions are schematically plotted in figure 5. Let us consider the initial condition corresponding to a certain aerosol loading (point A in figure 5). The increase in aerosol loading leads to an increase of both L and G. The diagonal line in figure 5 separates the quadrant into two zones. The upper zone corresponds to the condition  $\Delta L > \Delta G$ , i.e., to the precipitation decrease (scenario 1); in the zone below the line  $\Delta L < \Delta G$ , i.e. the precipitation increases (scenario 2). The realization of the first or the second scenario depends on the environmental conditions and on the cloud type. Below we are going to attribute different observational or numerical results to one of these scenarios.

### 4. Factors determining the aerosol effects on precipitation

Below we will discuss three main factors which affect the relationship between  $\Delta L$  and  $\Delta G$ : the cloud type (stratocumulus and small cumulus, single deep convective

<sup>&</sup>lt;sup>1</sup> Wood (2007) and Stevens and Seifert (2008) investigated the dynamical feedback of aerosols on the depth of stratocumulus and small cumulus clouds. Wood reported thickening of stratocumulus clouds if their cloud base is lower than 400 m and thinning if their cloud base is higher than 400 m.



**Figure 3.** The vertical profiles of horizontally averaged (per grid of the computational area) moistening/drying (upper panels) and heating/cooling (lower panels) over a 4 h period in the simulations of M and T clouds (left panels) and GO and S clouds (right panels) (adopted from Khain *et al* 2008a). The differences between moistening and drying, as well as between heating and cooling represent the net effects, which are shown as well (solid lines). Areas marked in black show zones where greater moistening and cooling takes place in the polluted clouds; the areas marked in grey denote zones where greater moistening and cooling takes place in clean air. A larger area of black zones indicates higher net atmospheric moistening, cooling, and decrease in precipitation in polluted clouds.

clouds, orographic clouds, cloud systems), the air humidity, and the wind shear.

### 4.1. Stratocumulus and warm rain cumulus clouds

From the budget point of view the obvious suppression of precipitation from small cumulus and stratocumulus clouds in dirty air reported in many studies (e.g., Albrecht 1989, Rosenfeld 1999, 2000, Feingold *et al* 2005, Cheng *et al* 2007, Altaratz *et al* 2008) can be explained as follows. An increase in the AP concentration leads to the formation of a large amount of small drops. Collisions of these droplets in stratocumulus clouds become so inefficient that drizzle does not form during droplet residence time within clouds. According to the simulations with a new trajectory ensemble model of stratocumulus cloud (Pinsky *et al* 2008b, Magaritz *et al* 2007, 2008) the increase in the AP concentration from  $\sim$ 200 to  $\sim$ 600 cm<sup>-3</sup> can totally prevent drizzle formation

in stratocumulus clouds (figure 6). The increase in AP concentration significantly decreases the drop size in small cumulus clouds as well. In both cases cloud droplets or small raindrops efficiently evaporate during their slow settling. Small isolated cumulus clouds experience intense mixing with the environment, which additionally increases  $\Delta L$  by evaporation. This conclusion fully agrees with the results of Xue and Feingold (2006), who wrote 'the complex responses of clouds to aerosols are determined by competing effects of precipitation generation and droplet evaporation associated with entrainment. As aerosol concentration increases, precipitation suppression tends to maintain the clouds and lead to higher cloud LWP (liquid water pass), whereas cloud droplets become smaller and evaporate more readily, which tends to dissipate the clouds and leads to lower cloud fraction, cloud size, and depth.' A decrease in relative humidity dramatically intensifies the evaporation. In these clouds an increase in the AP concentration corresponds to



**Figure 4.** The same as in figure 3, but for the simulations of clouds with maritime sounding and low (M) and high (M-c) aerosol concentration. The M-c cloud heats and dries the atmosphere to a larger degree than the M cloud, i.e. the precipitation from the tropical maritime deep clouds increases in dirty air.



**Figure 5.** Scheme illustrating two possible scenarios of aerosol effects on precipitation with increase in concentration of small AP. If the increase in generation is lower than the increase in the precipitation loss, precipitation suppression will be observed (scenario 1). If the increase in generation is larger than the increase in the precipitation loss, precipitation enhancement will take place (scenario 2).

the case  $\Delta L > \Delta G$ , i.e. stratocumulus and small convective clouds are located above the diagonal in figure 5 (scenario 1).

### 4.2. Isolated deep convective clouds

The analysis of the condensate mass and the heat budgets for isolated GO and S clouds (figures 2 and 3) indicates that an increase in the AP concentration increases the condensate loss to a higher degree than the production, i.e.  $\Delta L > \Delta G$ . As a result, with other conditions being similar the precipitation from a single S cloud is lower than that from a corresponding GO cloud (figure 7). Similarly, precipitation decreases in the case of summertime Texas clouds (see curves T and T-m in figure 8). As it was shown by Rosenfeld and Woodley (2000), Khain et al (2001), Khain and Pokrovsky (2004) and Khain et al (2008a), the strong atmospheric instability and high AP concentration typical of these clouds lead to the formation of a significant amount of ice crystals by homogeneous freezing. These crystals spread over large areas and sublimate. Very low relative humidity (about 35%) leads to efficient ice sublimation and drop evaporation. Thus, an increase in AP concentration leads to a significant increase in  $\Delta L$ , which exceeds the increase in the condensate generation. Thus, isolated clouds simulated in Brazil and Texas correspond to scenario 1 ( $\Delta L >$  $\Delta G$ ). Their location in figure 5 is above the diagonal.

The opposite effect of AP on precipitation was found in simulations of deep maritime clouds. As one can see in figure 4, the increase in AP concentration leads to a higher increase in condensate generation than that of the condensate loss ( $\Delta L < \Delta G$ ). Correspondingly, precipitation from maritime clouds increases with increase in the AP concentration (see curves M and M-c in figure 8). We attribute this effect to the fact that the relative humidity of the maritime atmosphere is high. Under wet environmental conditions the condensate generation increases dramatically (figure 8), but the evaporation loss of the condensate increases to a lesser extent. This relationship determines the high precipitation efficiency of maritime clouds (see right panel of figure 7). To justify the effect of humidity on cloud-aerosol interaction, supplemental simulations of maritime clouds were performed with the relative humidity 10% lower than in the M and M-c runs. These



Vertical profiles of rain flux (drizzle)

**Figure 6.** Vertical profiles of drizzle flux at different time instances in simulations of stratocumulus clouds observed in the DYCOMS-II field experiment (research flight RF07). Vertical profiles of the fluxes are plotted by different colours with time increment of 5 min. Simulations were performed using a novel trajectory ensemble model described in Pinsky *et al* (2008b). The increase in the AP concentration from  $\sim$ 200 to  $\sim$ 600 cm<sup>-3</sup> fully inhibits drizzle formation in maritime stratocumulus clouds under thermodynamic conditions of the DYCOMS-II field experiment. Adopted from Magaritz *et al* (2007).



**Figure 7.** Time dependence of the accumulated surface precipitation amount (left) and precipitation efficiency (right) in all simulations including two GO simulations with No = 400 and 100 cm<sup>-3</sup>, the maritime GATE-74 and summertime continental Texas clouds. Precipitation and efficiency for pyro-clouds is adopted from Khain *et al* (2008a). In the case of biomass burning precipitation falls from the secondary clouds. Precipitation from pyro-clouds continues during the whole simulation period because of continuous surface heating (hot spot) in the fire zone (see Khain *et al* 2008a for detail).

simulations correspond to the 80% relative humidity (RH) near the surface and referred to as M-80 (low AP concentration) and M-c-80 (high AP concentration). Figure 8 shows that a decrease in RH by only 10% led to a dramatic decrease in precipitation in M-80 and to the inversion of the precipitation response to aerosols; while the precipitation in the M-c case was larger than in the M clouds, the precipitation in the M-c-80 case is smaller than that in the M-80 clouds. This means

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**Figure 8.** Time dependence of the accumulated surface precipitation in the simulations M, M-c, M-80 and M-c-80, and T and T-m. One can see that increase in the AP concentration under maritime conditions leads to an increase in precipitation. In the case of Texas summertime clouds, the decrease in the AP concentration leads to the precipitation enhancement. A decrease in RH by only 10% led to a dramatic decrease in precipitation in the M-80 clouds and to the inversion of precipitation response to aerosols; while the precipitation in the M-c case was larger than that in the M clouds, the precipitation in the M-c-80 case is smaller than that in the M clouds.

that relative humidity is one of the major factors determining the positive response of precipitation in deep tropical maritime clouds to the aerosol loading and the negative response of stratocumulus, warm rain cumulus, and extreme continental clouds.

#### 4.3. Convective systems

Under certain thermodynamic conditions (e.g., high convective available potential energy (CAPE), significant wind shear, etc) downdrafts created by primary clouds lead to the formation of secondary clouds. Khain et al (2003), Khain et al (2004, 2005), Lynn et al (2005a) found that aerosols foster the formation and intensification of secondary clouds and squall lines. Van den Heever and Cotton (2007) also demonstrated the significant role of aerosols in storm splitting and secondary storm development, as well as in the associated surface precipitation. There are at least two reasons for this intensification. The first mechanism is the same as in case of isolated clouds: aerosols lead to extra latent heat release. The second mechanism is related to the increase in the air convergence in the boundary layer caused by stronger downdrafts in the former polluted clouds. These aerosol effects will be referred to as dynamic aerosol effects. To clarify the mechanisms favourable for the formation of secondary clouds, let us analyse the time dependences of precipitation efficiency of polluted and nonpolluted clouds presented in figure 7 (right panel). According to the results, the PE of smoky clouds is 2-3 times lower than that of the clouds arising in clean air. The PE of tropical M clouds is 4-5 times higher than that of T clouds. These results agree well with the evaluations of PE presented by Braham

(1952, 1981), Marwitz (1972), Heymsfield and Schotz (1985) and Li *et al* (2002a). The fact that the PE is higher the M clouds can be attributed to a relatively small precipitation loss under high relative humidity. The PE of pyro-clouds (Khain *et al* 2008a) remains small in spite of continuous generation of new clouds by surface heating.

The main point related to the problem of the secondary cloud formation is the following: since  $PE = \frac{P}{G} = \frac{G-L}{G}$ , the increase of PE and G with the increase in the air humidity means that the difference G-L should also increase, which corresponds to the increase in the temperature gradients between the zones of condensation (updrafts) and the zones of sublimation and evaporation (downdrafts).

Further more detailed investigations of dynamical aerosol effects on clouds and precipitation were performed by Lynn *et al* (2005b), Tao *et al* (2007), Fan *et al* (2007a, 2007b), Lee *et al* (2005, 2008) and Khain *et al* (2008d). In all the studies mentioned above the formation of secondary clouds (including new clouds within squall line) in polluted air led to an increase in precipitation in moist air.

Fan et al (2007b) and Tao et al (2007) focused on the role of air humidity in the formation of secondary clouds and squall lines. In these studies simulations were performed using the 2D Goddard Flight Center Cloud Model (GCM) with spectral microphysics described by Khain et al (2004).It was shown that an increase in the aerosol concentration under high humidity intensifies convection and leads to stronger evaporative cooling as compared to the clean air conditions. Figure 9 (adopted from Tao et al 2007) shows the vertical profiles of the evaporative rate in the simulations of squall lines observed in TOGA CORE (Central Pacific) and in PRESTORM (Oklahoma region, USA) field experiments. The simulations were performed for low and high AP concentrations. One can see that (a) the evaporative rates (precipitation loss) are higher in the dryer atmosphere (PRESTORM case); and (b) the difference in the evaporation rates in polluted and clean cases is larger in moist maritime air (the TOGA CORE case). The results agree well with the main statement of the present study that an increase in aerosol loading increases both the generation and the loss of condensate (i.e. cooling) in systems with high freezing level. Increased cooling intensified downdrafts and led to an intensification of maritime squall lines and increased generation of the condensate. As a result, an increase in aerosol loading increases the condensate generation to a higher extent than the condensate loss, which leads to rain enhancement in the Pacific squall line. The simulations indicate some aerosolinduced decrease in precipitation in the Oklahoma (drier) case, and no significant effect in the Florida squall line.

Lee *et al* (2005, 2008) simulated the evolution of cloud ensembles using the Weather Research Forecast (WRF) model (NCAR) with two-moment bulk parameterization under different wind shears. They found that an increase in aerosol loading leads to an increase in precipitation in the case of high humidity, strong wind shear, and instability. The strong shear and aerosols lead to self-organization of deep convection, intensification of deep clouds, and weak clouds decaying. The effect of aerosols on the formation of secondary clouds in the



**Figure 9.** Vertical profiles of averaged evaporation rates in maritime (TOGA CORE) (left) and continental (PRESTORM) (right) squall lines (adopted from Tao *et al* 2007). One can see that in the maritime case the evaporative cooling is much stronger in dirty air. This cooling fosters intensification of the squall lines and leads to precipitation enhancement.

presence of a significant wind shear can be attributed to the following. In the case of a strong wind shear, ice particles and small drops are detrained from the zone of updrafts and transported downwind from cloud updrafts, making the particles fall and evaporate in relatively dry air, which increases the downdrafts and the boundary layer convergence (as was shown in these studies). The latter fosters the formation of secondary clouds, producing a new condensate. Thus, the combined effect of moisture and wind shear leads to the condition ( $\Delta L < \Delta G$ ) which corresponds to scenario 2 (increase in precipitation) in figure 5.

Using the WRF model with SBM, Khain and Lynn (2008) simulated evolution of a supercell storm in clean and polluted air. They reported increase in precipitation with the aerosol loading in wet atmosphere and a decrease in precipitation with the increase in the aerosol loading under lower (by 10%) air humidity.

In most numerical studies dedicated to aerosol effects on precipitation, clouds with warm cloud base and high (about 4 km) freezing level are simulated. In these cases an increase in the AP concentration decreases warm rain and intensifies the formation of ice precipitation. The number of simulations of aerosol effects on microphysics and precipitation in mixed phase clouds with comparatively cold cloud base (low freezing level) is quite limited. Seifert and Beheng (2006) used a 3D model with two-moment bulk parameterization of microphysics to simulate supercell and multi-cell storms. They found that while an increase in the aerosol loading decreases precipitation from ordinary single cells and supercell storms, it leads to precipitation enhancement in multi-cell cloud systems. Humidity (buoyancy) and the wind shear were found to be the most important parameters determining the difference in the dynamics of the storms. Their conclusion agrees well with the results of the studies referred to above, namely that the formation of multi-cell structures due to the combined effect of the wind shear, the buoyancy, and aerosols lead to precipitation enhancement. The simulations of cloud systems with low freezing level are discussed in more detail in section 5.

#### 4.4. Orographic clouds

Givati and Rosenfeld (2004) and Jirak and Cotton (2006) reported a decease of precipitation over the mountain slopes located downwind from urban areas. Lynn et al (2007) simulated the development of clouds and precipitation in the Sierra Nevada Mountains using the 2D version of the WRF model (NCAR) with the spectral (bin) microphysics described by Khain et al (2004). The simulations were produced using either maritime ('clean-air') or continental ('dirty-air') aerosols for a 3 h time period. Figure 10 shows the spatial distribution of accumulated surface precipitation towards the end of the simulations in cases of low (MAR) and high (CON) AP concentrations. The left panel shows the results obtained under the relative humidity typical of the region. The right panel shows precipitation distribution obtained in the simulations with increased air humidity (the simulations are referred to as Mar-RH90 and Con-RH90; see table 1). In these simulations the initial relative humidity was set equal to 90% from the surface to 2 km and 50% from 2 to 5 km. Figure 10 (left panel) shows that an increase in the aerosol loading leads to a decrease in precipitation and to a spatial shift downwind. The accumulated surface precipitation decreased by about 30% from 0.44 mm in clean air to 0.32 mm in polluted air. This result agrees with that reported by Givati and Rosenfeld (2004). The agreement indicates that the increase in the concentration of anthropogenic aerosols is a plausible mechanism of the precipitation decrease over the mountain Analysis of the results shows that the increase in slopes. aerosol loading leads to warm rain suppression and to the generation of a significant amount of snow. Since snow has low settling velocity, snow is advected downwind (along the slope), which increases precipitation over the mountain peak. These cloud ice and snow particles evaporate on the downwind side of the highest mountain peak because of the very low humidity (desert) to the east of the peak. In addition, the relative humidity dramatically decreases in downdrafts over the downwind slope. Because of the sublimation of cloud ice and



**Figure 10.** The spatial distribution of accumulated surface precipitation from orographic clouds to the end of simulations in cases of low (MAR) and high (CON) AP concentrations. The left panel shows the results obtained under relative humidity typical of the region. The right panel shows the precipitation distribution obtained in simulations with increased air humidity (the simulations are referred to as Mar-RH90 and Con-RH90). The grey curve denotes the topography (adopted from Lynn *et al* 2007).

snow, the simulation with continental aerosols produced less precipitation over the whole mountain slope.

Higher humidity decreased the cloud base level and triggered the cloud formation further upwind on the mountain slope where the vertical velocity was smaller than further downwind on the slope. As a result, the droplet concentration turned out to be relatively small, and droplet spectra distributions were able to develop to produce raindrops. The efficient warm rain formation occurred even under a high AP concentration (but with some time delay and spatial shift in the downwind direction). Besides, the high relative humidity reduces the precipitation loss caused by drop and ice evaporation. Thus, the increase in air relative humidity decreased the difference in precipitation amounts between the clean- and dirty-air simulations, and even changed the sign of this difference: the increase in humidity leads to an increase in precipitation in clean and polluted air to 3.62 and 3.78 mm, respectively.

Thus, again, we can see that (a) humidity crucially affects the precipitation amount, and (b) aerosol effects on precipitation (and even the sign of the precipitation response) substantially depend on relative humidity. In the case of orographic precipitation, changes in humidity also affect the spatial precipitation distribution. Simulations show that in the case of comparatively low humidity  $\Delta L > \Delta G$ , i.e. aerosols decrease the precipitation, while at high humidity  $\Delta L < \Delta G$ , so aerosols increase the precipitation.

### 5. Discussion and conclusions

Figure 11 summarizes the results of the classification of aerosol effects on the precipitation from clouds and cloud systems with a high freezing level. The analysis shows that a significant fraction of the discrepancies of the results reported concerning the precipitation response to aerosols can be attributed to

different cloud types and different atmospheric conditions used in different studies.

Hence, there is no general answer to the question: Do aerosols decrease or increase precipitation? Aerosols affect cloud microphysics and dynamics, changing the items of the mass budget determining the precipitation amount. For conditions with high freezing level an increase in the aerosol loading increases both the generation and the loss of the condensate mass. The net effect depends on whether the increase in the condensate generation is larger or smaller than the increase in the condensate loss. The effect depends both on the environment conditions and on the cloud type. Aerosols decrease precipitation (and can fully suppress it) in stratiform and stratocumulus clouds, as well as in warm rain cumulus clouds.

Aerosols seem to decrease precipitation from isolated deep clouds developing in very dry unstable atmospheres (similar to summertime Texas clouds). There are some numerical results indicating a slight decrease in precipitation from clouds developing over the ocean in zones where the relative humidity is relatively low (below 70–80% near the surface). An increase in the air humidity increases the generation of condensate more than the loss. As a result, under high humidity typical of tropical convection, aerosols increase precipitation from deep convective clouds.

An increase in aerosol loading leads to an increase in the evaporation of the precipitation mass and to the acceleration of downdrafts, which fosters the formation of secondary clouds (or the formation/intensification of squall lines). Such an aerosol-induced dynamic effect leads to an increase of the condensate generation to a higher extent than the loss, and, consequently, to a precipitation increase in the zone of convection. The formation of secondary clouds depends on the wind shear and the instability of the atmosphere (including the instability of the boundary layer), and on the air humidity. We suppose that there exist some thresholds in these



Figure 11. The classification scheme of aerosol effects on precipitation. The magnitudes of air humidity, buoyancy, and wind shear specify the major environmental factors determining the precipitation response to aerosols.

atmospheric parameters (as well as in the convection triggering mechanisms) whose exceeding leads to the quantitative change in the convection type from singe clouds to the organized structures of dynamically related clouds or squall lines.

Simulations (e.g. Lynn *et al* 2005b, Lee *et al* 2005, 2008) show that aerosols tend to intensify deep convective clouds and suppress small clouds. This tendency is especially pronounced in the moist tropical air masses. This tendency can lead to a spatial redistribution of precipitation and intensification of squall lines and storms. We attribute the differential sensitivity of different clouds to aerosols to two mechanisms: (a) suppression of precipitation from small clouds and the invigoration of deep convective clouds as is discussed above, and (b) suppression of small clouds in the vicinity of deep convective clouds by compensating downdrafts in the surroundings of deep clouds.

We can see, therefore, that, even in the case of net precipitation enhancement, there will be the areas where precipitation decreases. This means that the effects of aerosols on precipitation should be evaluated depending on the metric used for the analysis (Lee *et al* 2008).

In all cases an increase in air humidity increases the precipitation efficiency, which is especially important in polluted air cases. The scheme shown in figure 11 is not exhaustive in several aspects. In the study we did not consider, for instance, the effect of radiative aerosol properties on the atmospheric stability/instability.

Besides, the scheme is only qualitative because of both physical and numerical reasons. *To physical reasons* we attribute significant gaps in the knowledge of warm and especially ice cloud microphysics as well as some dynamical effects. Among them we shall mention the following.

(a) The concentration and spatial distribution of giant CCN (GCCN) (particles with dry radii exceeding 5–10  $\mu$ m) under different conditions are actually unknown. In spite of significant efforts performed to measure GCCN during past several decades, the most frequently referenced data as regards GCCN date back to 1953 (Woodcock 1953). It is clear, however, that the concentration of GCCN is by several orders of magnitude lower than that of small aerosols serving as CCN. According to some studies, the role of GCCN is very significant (e.g. Rosenfeld *et al* 2002, Yin *et al* 2000). At the same time, according to other studies (e.g., Khain *et al* 2006), the influence of GCCN on precipitation from deep convective clouds turned out to be comparatively weak (even under GCCN concentrations as

high as  $0.1-1 \text{ cm}^{-3}$ ). Note first that wet radii of aerosols near the cloud base are 3-5 times larger than those of dry particles, so aerosols with dry radii of 1  $\mu$ m and 2  $\mu$ m have wet radii of 5  $\mu$ m and 8  $\mu$ m, respectively (e.g. Segal *et al* 2007). These sizes are smaller than those of GCCN by factors of only 1.5-2. Besides, smaller droplets grow by diffusion faster than larger ones (Rogers and Yau 1989). In the presence of high concentration of droplets forming on small CCN, the supersaturation in polluted clouds is low and the GCCN (by the way, GCCN remain non-activated aerosols, since their critical radius cannot be attained in the course of diffusion growth) grow by diffusion very slowly. As follows from simulations by Khain et al (2000), the concentration of  $\sim 20 \ \mu m$  droplets (which are able to trigger efficient collisions) formed on small aerosols by diffusion growth and autoconversion is significantly larger that of those formed on GCCN. If the CCN concentration is low the precipitation forms rapidly no matter whether GCCN exist or not. The effect of GCCN on precipitation from small cumulus and stratocumulus clouds should be substantially higher. At the same time, simulations of the drizzle formation in a stratocumulus cloud using a Lagrangian trajectory ensemble model (Magaritz et al 2008) show that there exist efficient mechanisms of drizzle formation even if the CCN spectrum does not contain GCCN. Further investigations are required in this field. Anyway, the effect of GCCN should be opposite to that of small CCN, i.e. GCCN foster warm rain formation at low levels, decreasing the supercooled liquid water content (LWC) at high levels.

(b) Collision rates between droplets and, especially, between ice particles themselves, as well as between ice and water (the rate of riming) are known only by order of magnitude. Recent studies (Pinsky et al 2007, 2008a, Wang et al 2005, Xue et al 2008a, 2008b, Franklin et al 2005) showed that in deep clouds turbulence increases the collision rate between droplets several times, i.e. the collision rate between droplets is determined largely by turbulence and not by gravity. Since turbulence is highly inhomogeneous within clouds, collisions are triggered first in some specific zones of clouds with enhanced turbulence (Benmoshe et al 2008). Collision rates between ice particles as well between ice particles and water drops are not well known even in the pure gravity case. At the same time, these collisions determine the formation of large precipitating particles (hail, graupel, snow), which can reach the surface without significant evaporation. One can suspect that turbulence increases these collision rates even higher than those between droplets (Pinsky and Khain 1998). Being taken into account, turbulence must increase the precipitation efficiency, and possibly the precipitation amount from deep clouds. Benmoshe et al (2008) showed that convection invigoration caused by aerosols also increases the turbulence intensity and the collision rate. The latter indicates that aerosol effects are not limited only by the decrease of droplet size at the stage of diffusion growth. Further investigations are required to quantitatively evaluate the net aerosol effects on precipitation, taking into account aerosol effects on the turbulent intensity in clouds.

- (c) Note also that small scale turbulence also affects the mixing of clouds with the environment air (e.g., Andrejczuk *et al* 2006, Grabowski 1993, 1995, 2007, Kruger *et al* 1997, Khain *et al* 2004, Xue and Feingold 2006). At the same time, the problem of adequate description of turbulent mixing and entrainment is not yet solved (see, e.g., Stevens *et al* 2005). Models with a resolution of 50 m and better are required to investigate the effects of mixing and turbulent effects on size distributions and rain formation.
- (d) As was shown in some studies (e.g. Van den Heever et al 2006), the effects of IN on surface precipitation can be significant. However, the concentration of IN under different conditions is not known. Besides, the mechanisms of ice production and their relationship with IN are not well understood.

The scheme shown in figure 11 is not exhaustive in another important aspect: namely, it relates to clouds and cloud systems arising under high freezing level. The number of numerical studies considering aerosol effects on the microphysics of mixed phase clouds with cold cloud base and low freezing level is quite limited. At the same time, the microphysical processes in cloud systems with a high freezing level (above 4 km) and relatively low freezing levels (2-2.5 km) may be quite different. In cloud systems with high freezing level and low AP concentration, warm rain dominates. An increase in the AP concentration decreases the warm rain and intensifies ice (cold) processes, leading to convective invigoration and other effects discussed above. In the case of the low freezing level, warm rain turns out to be inefficient under any reasonable AP concentration. In this case ice processes play the major role in both clean and dirty environments, and convective invigoration is not pronounced. As was shown by Rosenfeld and Khain (2008) and Khain et al (2008d), in clouds with comparatively low freezing level, the processes of hail formation become of crucial importance. Van den Heever et al (2006) reported that an increase in the AP concentration favours the formation of large hail. The aerosol effects of hail formation were investigated in more detail recently by Khain et al (2008d, 2008e). A hail storm in southwest Germany at Villingen-Schwenningen on 26.06.2006 was simulated using the HUCM. The model has been significantly improved to allow the simulation of big hail. In particular, the calculation of conditions leading to the wet growth of hail was implemented. Besides, the process of riming snow to graupel was described by the calculation of rimed fraction within snow particles and recalculation of the bulk density of rimed particles. To investigate aerosol effects on precipitation and hail size distribution, simulations were carried out for CCN concentrations ranging from 100 to  $6000 \text{ cm}^{-3}$  (at 1% supersaturation). The increase in the AP concentration leads to an increase of supercooled LWC at higher levels. Hail embryos efficiently collect small water drops and grow rapidly. The maximum precipitation and kinetic energy of hail and graupel falling to the surface was reached at the CCN concentration of about  $3000 \text{ cm}^{-3}$ 



**Figure 12.** Time dependence of accumulated kinetic energy of large graupel and hail at the surface in the hail storm in southern Germany (26 June 2006) simulated by the HUCM under different CCN concentrations. One can see that the kinetic energy reaches its maximum at the AP concentration of about  $3000 \text{ cm}^{-3}$ .

(figures 12 and 13). Analysis of the heat and mass budgets indicates a significant difference of aerosol effects on tropical and mid-latitude storms. Under tropical conditions, an increase in the AP concentration increases both condensate generation (leading to convective invigoration) and condensate loss, decreasing the precipitation efficiency. In the mid-latitude storm simulated, an increase in the AP concentration does not increase the generation of condensate (see figure 14). Accordingly, a very weak convective invigoration was found. At the same time, aerosols foster large graupel and hail formation. Because of the high fall velocity, big hail and graupel efficiently collect cloud water and do not sublimate during their fall, which decreases precipitation loss. A somewhat 'paradoxical' result was found, namely that small aerosols increase the precipitation efficiency of deep mixed



**Figure 13.** The same as in figure 12, but for accumulated precipitation. One can see that precipitation increases with the increase in AP concentration from 100 to  $3000 \text{ cm}^{-3}$  and then decreases with further increase in the AP concentration.

phase storms (see figure 15), which contrasts with the scheme of aerosol effects on cloud systems with a high freezing level. As a result, an increase in the CCN concentration leads in the case simulated to an increase in the accumulated rain and hail mass, as well as to an increase in the hail size. The biggest hail stones simulated were of  $\sim$ 4 cm in diameter. Note that during the first hour precipitation was maximal at low AP concentration (100 cm<sup>-3</sup>). This result agrees with that of Teller and Levin (2006), who simulated the 1 h development of an isolated single cloud using an axisymmetric spectral microphysics model. In our simulations the precipitation begins with the formation of secondary cells giving rise to the hail storm.

The decrease in precipitation at AP concentrations exceeding  $3000 \text{ cm}^{-3}$  can be attributed to the formation of



**Figure 14.** Vertical profiles of horizontal averaged heating and cooling (left) and moistening and drying (right) in the hail storm in southern Germany calculated for low (M: blue curves) and high (C: red curves) aerosol concentrations during a period of 180 min. Areas marked pink/light blue denote zones where net heating and drying are larger/smaller in the high AP concentration case, respectively. Simulations were performed using an updated version of the HUCM (Khain *et al* 2008e).



**Figure 15.** Time dependence of precipitation efficiency in clean and polluted air in the hail storm that took place in southern Germany (as calculated using the HUCM). The increase in the precipitation efficiency is caused by hail formation.

a significant amount of ice crystals formed as the result of homogeneous freezing of small droplets at heights of about 10 km.

More investigations of aerosol effects on clouds/ cloud systems with relatively low freezing level are required. The main point that should be stressed here is that hail formation must be simulated with all possible accuracy, because the results will dramatically depend on the representation of hail in the models (see, e.g., Noppel *et al* 2008, Khain *et al* 2008e). As follows from results by Khain *et al* (2008b, 2008a, 2008d, 2008e, 2008c), there should exist thermodynamic situations (characterized by the cloud base temperature and cloud depth), that separate the cases when aerosols decrease the precipitation efficiency (as in figure 7) and when aerosols increase the precipitation efficiency due to the hail and large graupel formation (as in figure 15).

Note that the scheme plotted in figure 11 is not exhaustive as regards the aerosol effects on precipitation at larger scales. For instance, Rosenfeld *et al* (2007), Zhang *et al* (2007) and Khain *et al* (2008b) reported that aerosols influence the dynamics and precipitation from large mesoscale systems, such as tropical cyclones. The scheme in figure 11 also does not consider aerosol effects on precipitation on synoptic and global scales. In spite of several papers having been published, this topic remains largely unclear, because of the lack of knowledge of to how represent aerosol effects in the GCM models (taking into account the high accuracy required).

Discrepancies and uncertainties in the evaluation of aerosol effects can be caused by *numerical reasons*, as discussed in detail in appendix **B**. The potential source of errors is the treatment of the budget concerning atmospheric aerosols. Most bin microphysics schemes take into account the decrease in the aerosol concentration and change of the aerosol size distributions by nucleation. Some of the bin schemes include the release of aerosols into the atmosphere during droplet evaporation. At the same time, most bulk schemes

assume an infinitely large source of aerosols. The difference in aerosol budget treatment can result in significant differences as regards the evaluation of aerosol effects on precipitation (Seifert *et al* 2006).

Note in conclusion that in spite of many difficulties and uncertainties remaining, significant progress has been achieved during the past decade as regards the understanding of the mechanisms by means of which aerosols influence precipitation. However, significant efforts are required to make the estimations reliable from a quantitative point of view.

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### Appendix A. Uncertainties of the evaluation of aerosol effects from the measurements

In this appendix we present several examples of the uncertainties in evaluations of precipitation response to aerosol loading.

(a) Unjustified generalization of the results of measurements. The first example concerns the unjustified generalization of the results obtained for clouds of a certain type to all clouds. A decrease in the droplet size in polluted clouds found in situ measurements and from satellites (Albrecht 1989, Rosenfeld 1999, 2000) was first interpreted (the interpretation appeared in earlier studies, and has been cited in a great number of successive studies) as the evidence of the aerosol-induced suppression of precipitation in all cases. As is shown, these conclusions are valid for stratocumulus and warm rain small cumulus clouds. The net effect of the aerosols on precipitation (including cold precipitation) cannot be derived from these studies.

The second example concerns an unjustified generalization of the results of measurements performed in a particular place to much larger areas. In a well known study by Borys *et al* (2000), a decrease in the amount of snow in the polluted air was reported. The measurements were performed at a meteorological station located within a complex terrain area (the Rocky Mountains). No measurements of the precipitation amount downwind or upwind from this station have been performed (personal communication, 2007). At the same time, an increase in aerosol concentration leads to a delay of the surface precipitation and to the precipitation shift in the downwind direction (e.g., Givati and Rosenfeld 2004, Lynn et al 2007). Hence, a decrease in precipitation in one place may be accompanied by an increase in the precipitation downwind. The effect of such possible spatial precipitation shift on the net precipitation amount is not obvious. Respectively, the conclusion concerning the decrease in snow precipitation in polluted air is strictly

valid for the particular measurement point and cannot be applied to the whole region.

The third example concerns the problem of mathematical difficulties of statistical analysis of precipitation trends. As is known, the rate and spatial distribution of precipitation are extremely non-uniform. A comparatively small number of intense rain events can determine the annual precipitation amount. The sets of surface meteorological stations are often too rare to measure precipitation accurately and especially the possible trends related to aerosol effects (or some other factors). Annual rainfall records at a single station have large variations that are sometimes larger that the mean annual rainfall (Sumner 1988). Even over continental areas where data from several hundred stations are averaged, the variations in adjacent 30 yr periods can be as high as 25% (Hulme and New 1997). The large variability, asymmetry, and non-normal distribution of annual rainfall at a single station over several decades completely mask any long-term trend that might exist in the record (Paldor 2008). In an attempt to overcome this inherent large variability of annual rainfall at a single station, the station-to-station rainfall ratio was employed recently by Givati and Rosenfeld (2004), Alpert et al (2008), Jirak and Cotton (2006) and in some other studies to reveal precipitation trends that could be attributed to aerosol effects on precipitation. The authors of the first two studies investigated precipitation downwind from the urban zone in Israel using nearly similar data sets and reported opposite precipitation trends. Paldor (2008) showed that the opposite results can be attributed to the fact that 'the evaluation of trends in rainfall at a station based on the trend line of rainfall ratio lacks statistical validity'.

- (b) Aerosols represent only one factor (among many factors) affecting precipitation. For instance, an increase in aerosol concentration can be accompanied by changes of the atmospheric stability, the surface temperature, etc. Revealing the aerosol effect represents quite a difficult problem.
- (c) Apart from very few studies, most investigations have dealt with the effects of anthropogenic aerosols on shallow warm non-precipitating or slightly precipitating clouds. There are no direct observations supporting or denying the aerosol effects on precipitation from deep convective clouds and storms. The first attempt to evaluate aerosol effects over Brazil during the period of biomass burning has been undertaken only recently (Lin *et al* 2006). In this study, precipitation was evaluated from satellites. Quite unexpectedly the authors found a precipitation enhancement caused by the biomass burning. Earlier it was assumed that precipitation decreases in the zones of biomass burning. More studies are required to justify the rain retrievals from satellites.

The problems related to observational determination of aerosol effects are described in more detail by Levin and Cotton (2007). The difficulties of the evaluation of aerosol effects on precipitation from observations increase the role of numerical models which make it possible to reveal the precipitation changes related only to aerosols.

### Appendix B. The uncertainties of evaluation of aerosol effects using numerical models

*Numerical reasons* leading to the discrepancies and uncertainties in the evaluation of aerosol effects on precipitation can be divided into two large groups. *The first group* is related to different (and sometimes questionable) designs of numerical simulations (too small computational area, crude resolution, performance of simulations for the time period shorter than the life time of a single cloud, the utilization of models which cannot take into account wind shear, utilization of 2D models for simulation of atmospheric phenomena with pronounced 3D properties, etc).

The properties of single clouds can depend on the way of cloud triggering. For instance, a strong cool pool is often used to trigger squall lines. At the same time, the utilization of another kind of triggering (say, a warm temperature pulse) may not lead to squall line formation.

It should be pointed out that many numerical models do not obey any moisture conservation and artificially introduce a source of moisture in the model. A positive-definite advection scheme is at least a necessary criterion for moisture conservation.

In many simulations, precipitation from the secondary clouds contributes mostly to net precipitation. Hence. the evaluation of aerosol effects in simulations where the contribution of secondary clouds is taken into account may differ from those obtained in simulations where the contribution of secondary clouds is neglected. In this sense the utilization of 1D parcel models and axisymmetric models, which do not take into account wind shear and do not make possible the development of secondary clouds, can hardly produce reliable evaluations of aerosol effects. Twodimensional models allow simulation of wind shear and secondary clouds, and they can be successfully used for the simulation of isolated clouds and squall lines, orographic clouds, etc. The advantage of 2D models is the possibility to use high model resolution (up to a few tens of metres) to simulate deep convective clouds. At the same time, such phenomena as supercell and multi-cell storms should be simulated using 3D models. The 3D models are able to describe the spatial structure of precipitation much more realistically than 2D models. At the same time, the model resolution used in 3D simulations is comparatively crude (1–3 km). The utilization of such resolution does not allow simulation of clouds with characteristic sizes below  $\sim$ 4–10 km, because of crucial errors in reproduction of wavelengths of  $2\Delta x - 4\Delta x$  (where  $\Delta x$  is the distance between the neighboring grid points). As was reported in several studies, aerosols tend to invigorate deep convective clouds and storms, while decreasing the intensity and precipitation from small clouds. The physical meaning of these results was discussed in the present study. Here we note that artificial damping of small clouds by using crude resolution can also lead to this result.

*The second group of reasons* for significant dispersion of the results is related to the differences in the microphysical schemes used. As was mentioned above, precipitation at the surface represents often a small difference between two large values: the generation of hydrometeor mass by condensation and ice deposition and the loss of the hydrometeor mass by ice sublimation and drop evaporation. An accurate calculation of both the generation and loss of precipitating masses is required to calculate the precipitation amount. For instance, a 10% error in the calculation of evaporation may lead to 100% error in the precipitation amount. Revealing aerosol effects on precipitation imposes much heavier demands on the precision of the calculations of the components of the heat and moisture budget in numerical models than are needed just for the calculation of precipitation amounts.

At the same time, the relatively low accuracy of widely used bulk-parameterization schemes as regards precipitation calculation is well known (e.g., Lynn et al 2005b, Lynn and Khain 2007, Li et al 2008b). The microphysical structure of clouds in these schemes is parameterized using empirical and semi-empirical relationships. At the same time, effects of aerosols represent the finest properties of cloud microphysics, which can hardly be accurately described within the frame of one- or even two-moment bulk-parameterization schemes. Most cloud physical processes are highly nonlinear and dramatically change the particle size distributions. For instance, evaporation should decrease the concentration of small particles, keeping the largest ones. At the same time, prescribing the gamma distribution or Marshall-Palmer distribution 'keeps' a large concentration of small raindrops in surface precipitation. Another example of the same kind is related to the freezing process. It is known that large drops should freeze first, removing the tail of large drops in the drop size distributions. The assumption of a gamma distribution 'restores' the tails of the raindrop distributions, which leads to significant errors in the calculation of most processes (see, e.g., Li et al 2008b). Another example of errors that can be introduced by state-of-the-art bulk parameterizations is related to the absence of the aerosol budget. In most bulk-parameterization schemes aerosol size distributions and aerosol concentrations are assumed unchangeable during nucleation process. The chemical composition is also changed as a result of nucleation, because aerosols of different chemical compositions have different nucleation rates. Neglecting the changes in aerosol concentration and size distribution during the 'nucleation scavenging' can dramatically affect the results of simulations (see Seifert et al 2006). Ignoring the changes of the aerosol size distributions during nucleation is especially questionable in studies where periodic lateral boundary conditions are used.

In this sense the spectral bin microphysical schemes have a significant advantage as compared to the bulk schemes, because they calculate the changes in the size distributions caused by aerosols (Lynn *et al* 2005b, Li *et al* 2008b), and take into account the change in the aerosol concentration and size distribution during nucleation. At the same time, the spectral bin microphysical schemes are much more time consuming than the bulk schemes. Besides, the spectral bin microphysical schemes have their own technical problems, as described in detail by Khain *et al* (2000).

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