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Simulation of the Dynamics, Microstructure and Cloud Chemistry of a Precipitating and a Non-Precipitating Cloud by Means of a Detailed 2-D Cloud Model

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Abstract

The predictions of our 2-D cloud model with detailed microphysics for June 3, 1992 were compared with observations made on the same day during the field experiment CLEOPATRA on June 3, 1992. Using the observed vertical temperature and humidity distribution and the observed aerosol characteristics as model input, excellent agreement was obtained between the observations and the theoretical predicitions for: (a) the cloud base and the cloud top, (b) the liquid water content and drop size distribution and (c) the number concentration of CCN. The chemical composition of cloud- and rain water was in good agreement with the observations. In addition it was for the first time possible to verify by observation our predictions (Flossmann et al.; 1985, Flossmann and Pruppacher, 1988; Ahr et al., 1990; Flossmann, 1991) that the condensation process produces a sharp cut-off in the aerosol particle size distribution, leading to the formation of a cloud interstitial aerosol consisting of particles smaller than the cut-off radius.

Zusammenfassung

Simulation der Dynamik, Mikrostruktur und Chemie einer regnenden und einer nicht regnenden Wolke mittels eines zweidimensionalen Wolkenmodelles

Die Aussagen unseres 2-D Wolkenmodelles mit detaillierter Mikrophysik für den 3. Juni 1992 wurden verglichen mit den Beobachtungen, die an einer regnenden und einer nicht regnenden Wolke während des Feldexperimentes CLEOPATRA gemacht wurden. Als Modelleingabe diente die vertikale Verteilung der Temperatur und Feuchte am 3. und 4. Juni 1992 sowie die chemischen und physikalischen Charakteristika des an diesem Tage vorherrschenden Aerosols. Gute Übereinstimmung zwischen Theorie und Beobachtung wurde gefunden für: (a) die Wolkenbasis und Wolkenobergrenze, (b) den Flüssigwassergehalt der Wolke und das Tropfenspektrum, und (c) die Anzahlkonzentration der CCN. Die chemische Zusammensetzung des Wolken- und des Regenwassers fügte sich zufriedenstellend in die beobachteten Werte ein. Zusätzlich war es zum ersten Male gelungen, durch ein Feldexperiment unsere theoretische Voraussage zu verifizieren, welche besagt, daß der Kondensationsprozess eine relativ scharfe Abbruchkante in der Größenverteilung des Aerosolpartikelspektrums produziert und daß somit ein wolkeninterstitielles Aerosol vorhanden sein muß.

1 Introduction

The mechanisms by which atmospheric air pollutants are removed by clouds and precipitation play an important role in determining the distribution and concentrations of pollutants in the atmosphere as well as their deposition on the ground. In addition, wet removal is the cause for the acidity found in rainwater. The damaging effects of acid rain on the biosphere are well known. Numerous attempts have been published in literature which describe the up-take of particulate and gaseous pollutants by clouds and rain using cloud models. Past modelling studies have used a variety of dynamic frameworks with various degrees of success. Also the treatment of the microphysical processes in these models varied greatly, ranging from very detailed approaches to highly parameterized formulations. For a

review of some recent modelling approaches see Flossmann and Pruppacher (1988). This review shows that most models lack realism from numerous points of view or have not been applied to realistic scenarios documented by field experiments. An exception to this are the computations of Flossmann and Pruppacher (1988) who applied their 2-D cloud model with detailed microphysics and in- and below cloud scavenging to a specific case in Hawaii, and of Flossmann (1991, 1994) who applied the same model to Day 261 (18.9.74) of the GATE campaign. Although the micro- and macrostructure of the model cloud agreed well with those observed no comparison could be made regarding the predicted cloud chemistry since in both studies no data for the chemical content of the cloud and rainwater and for the chemical composition of the particulate and gaseous constituents were observed on the particular model day.

During the recent field study CLEOPATRA, carried out from May 11 to July 31, 1992 in the pre Alpine region west of Munich (Germany) (Meischner, 1993), detailed data were collected by air plane and on the ground. These characterized the dynamics, the microphysics, as well as the chemical constituents in the formed clouds, the precipitation and in the air. In order to verify our 2-D model with regard to its prediction of the chemical evolution of a cloud, we applied the model to the cloud development of June 3, 1992 during this field experiment. A brief summary of some pertinent results of this modelling endeavour shall be given below.

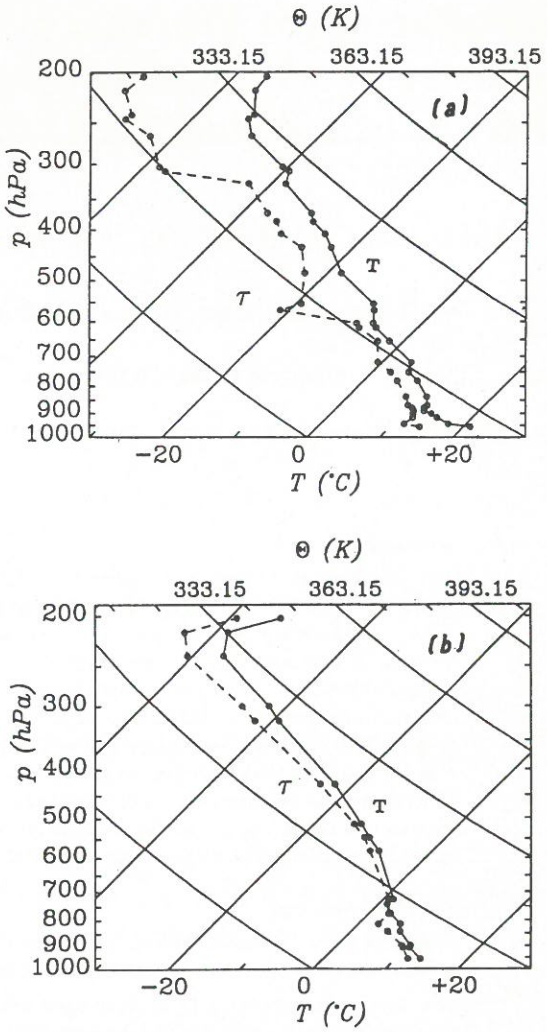


Figure 1 Vertical distribution of the temperature and the dewpoint on June 3, 1992 at 12 GMT (a) and on June 4, 1992 at 0:00 GMT (b) in Munich.

2 The Model Used

The basic dynamic framework employed in the present study is that employed and described in detail by Flossmann and Pruppacher (1988). The model consists of the two dimensional, slab symmetric version of the 3-D model described by Clark (1977, 1979), Clark and Gall (1982), Clark and Farley (1984) and Hall (1980). For the sake of brevity we shall refrain here from reiterating the dynamic and microphysical details of this models but refer the reader to Flossmann and Pruppacher (1988) for the dynamic framework and to Flossmann et al. (1985) for the microphysical details.

3 Initial Conditions

The weather situation of June 3, 1992 at the site of the field experiment was characterized by local cumulus in pre- and post-frontal air. The convective clouds in the morning and early afternoon of June 3 did not precipitate. Precipitation was observed in the late afternoon of June 3. For comparison with observation, we chose to model a non-precipitating cumulus in the early afternoon of June 3 (henceforth called *cloud-1*) and a precipitating cumulus in the late afternoon of the same day (henceforth called *cloud-2*). To cloud-1 the temperature and humidity distribution of June 3, 12:00 GMT (Figure 1a) was applied. Since no sounding was available for the late afternoon of June 3 the sounding applied to cloud-2 was interpolated between June 3, 12:00 GMT and June 4, 0:00 GMT (Figure 1b).

The model domain chosen had a vertical extent of 10 km and a horizontal extent of 20 km. The grid spacings were 200 m in the vertical and 400 m in the

horizontal. Our 2-D model domain was oriented according to the main wind direction in the south-east–northwest direction. The local wind was south-easterly. The particle size spectrum measured on that day was fitted by superimposing three lognormal distributions following Jaenicke (1988) according to:

$$\frac{dN_{APa}}{d\ln r_N} = f_{APa}(\ln r_N) = \sum_{i=1}^{3} f_{APa,i}(\ln r_N) \qquad (1)$$

$$= \sum_{i=1}^{3} \frac{n_i}{(2\pi)^{1/2} \log \sigma_i \ln 10} \cdot \exp\left(-\frac{[\log (r_N/R_i)]^2}{2(\log \sigma_i)^2}\right)$$

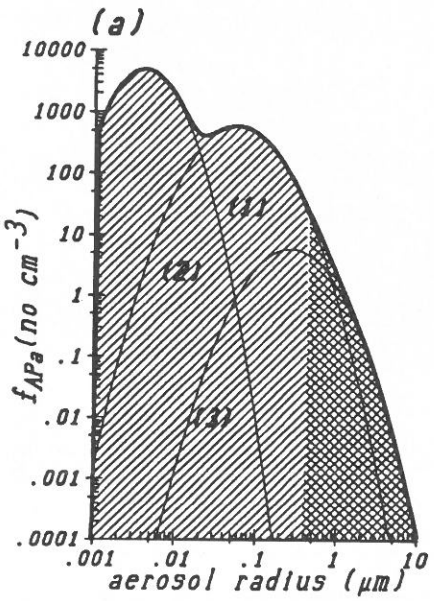
where r_N is the radius of the dry aerosol particle, R_i is the mean geometric radius, n_i is the concentration of the aerosol particles, σ_i is the standard deviation of the lognormal distributed aerosol particles in category i, N_{APa} is the total particle concentration in air, and f_{APa} is the number density distribution function. Observations showed that it was reasonable to consider that all particles of $r_N \leq 0.4\ \mu m$ contained a water soluble fraction of about 50 %, consisting of $(NH_4)_2SO_4$, and that particles with $r_N > 0.4\ \mu m$ contained a water-soluble fraction of about 10 %, of which 90 % consisted of NH_4NO_3, 4 % of Na_2SO_4 and 6 % of NH_4Cl. The total number concentration of particles was 8816 cm^{-3} corresponding to a total mass concentration of 59 $\mu g\ cm^{-3}$. The aerosol particle concentration was assumed to decrease with height exponentially according to

$$f_{APa,i}(z) = f_{APa,i}(z_0) \cdot e^{-z/H_i}. \qquad (2)$$

For particles with $r_N \leq 0.4\ \mu m$ the scale height H_i was assumed to be 3 km, for particles with $r_N > 0.4\ \mu m$ $H_i = 2$ km. The overall number distribution and volume distribution of the aerosol particle size spectrum is given in Figures 2a, b.

Table 1 Parameters characterizing the aerosol particle spectrum on June 3, 1992; n_i = number of aerosol particles cm^{-3}, R_i = mean geometric aerosol particle radius (μm), σ_i = standard deviation of particles in mode i.

mode i	n_i	R_i	$\log \sigma_i$
1	8000	0.00403	0.270
2	1250	0.05505	0.343
3	12	0.2540	0.343



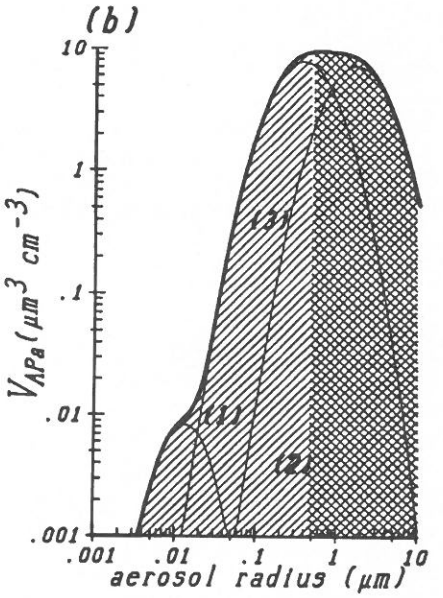


Figure 2 Number distribution function f_{APa} (a) and volume distribution function V_{APa} (b) of the aerosol particles observed on June 3, 1992 and fitted by Equation (1) with the numbers in Table 1. Total number concentration $N_{APa} = 8816\ cm^{-3}$, total particle mass $w_{APa} = 59\ \mu g\ m^{-3}$. The hatched section represents the particles of $r_N \leq 0.4\ \mu m$ containing a water soluble fraction of 50 %, consisting of $(NH_4)_2SO_4$. The cross hatched section represents the particles with $r_N > 0.4\ \mu m$ containing a water-soluble fraction of 10 %, of which 90 % consists of NH_4NO_3, 4 % of Na_2SO_4 and 6 % NH_4Cl.

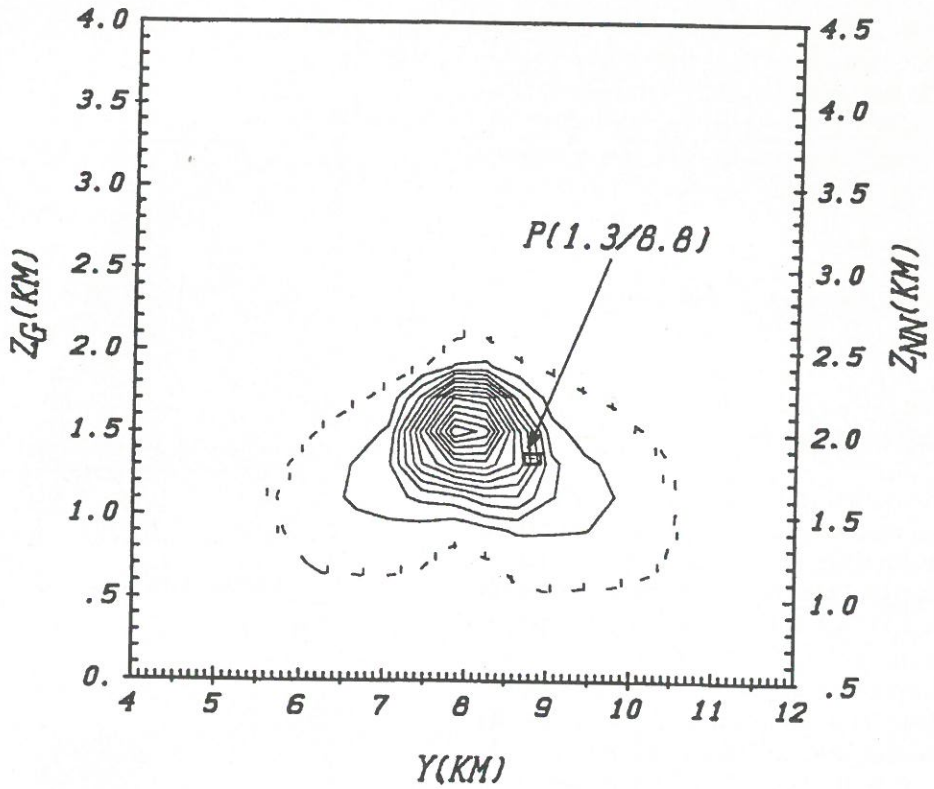


Figure 3 Model cloud-1 after 10 minutes cloud time (45 minutes model time). The isolines represent lines of constant liquid water content w_L. The spacing between the lines is 0.06 g kg^{-1}. The outermost dashed line represents the cloud outline. $w_{L,\,max}$ = 1.2 g kg^{-1}. Z_G represents the height above ground level and Z_{NN} the height above sea level.

4 Results and Conclusions

The temperature and humidity profile of June 3, 1992, 12:00 GMT allowed non-precipitating cumuli to develope. After 10 minutes cloud life time (45 minutes modeltime) model cloud-1 showed in Figure 3 had developed. The isolines represent the lines of constant liquid water content. The outermost dashed line is taken to represent the cloud outline and is given by a liquid water content of 0.063 g kg^{-1}. We noticed that the cloud base was located at a height Z = 0.9 km with respect to the ground (= 1.4 km with respect to the height NN above sealevel considered by the observing airplane). The cloud top was located at Z = 2.0 km (= 2.5 km with respect to NN). These levels corresponded well with the findings of the airplane from which it was observed that the cloud base was at 1.3 km with respect to NN and the cloud top at 3.5 km with respect to NN.

From Figure 3 it is also evident that the cloud had a maximum liquid water content of 1.2 g kg^{-1} located about in the center of the cloud. Since this location is hardly representative for the liquid water content observed by the airplane considering its flight track at 1500 m we rather chose for comparison the more representative point P (1.3/8.8) indicated by an arrow in Figure 3. The liquid water content at that location was 0.6 g m^{-3}. This model prediction is in good agreement with the observed liquid water content which carried at the flight level between 0.1 and 0.8 g m^{-3} along the flight track.

In Figure 4 we have plotted the drop size distribution numerically determined for the same location. Also plotted in this figure is the observed drop size distribution along the flight track at 1500 m. We notice that the agreement between model prediction and observation is satisfactory both with respect to the shape of the distribution as well with respect to the location of its maximum at which about 2500 drops per cm^3 at a radius interval between 6 and 8 µm were observed.

Measurements of the concentration of cloud condensation nuclei (CCN) at a supersaturation of

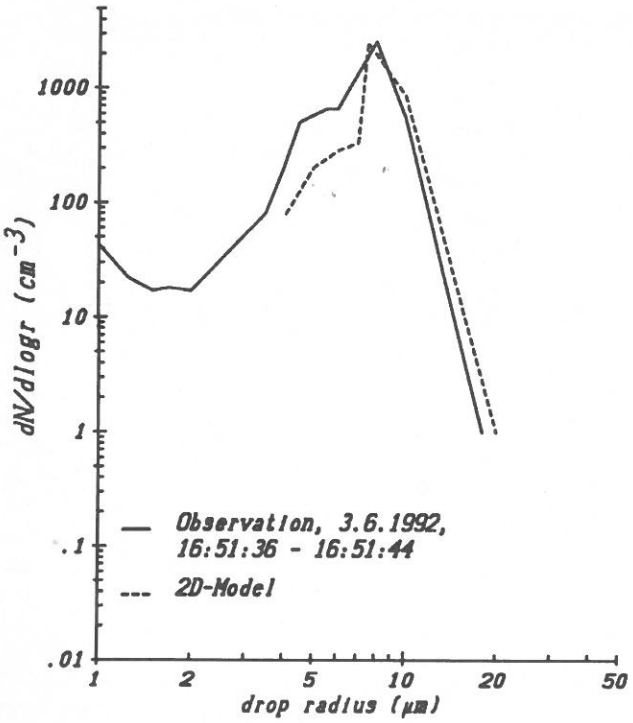


Figure 4 Comparison of the observed and computed drop size distribution at point P (1.3/8.8) in model cloud-1 after 10 minutes cloud time.

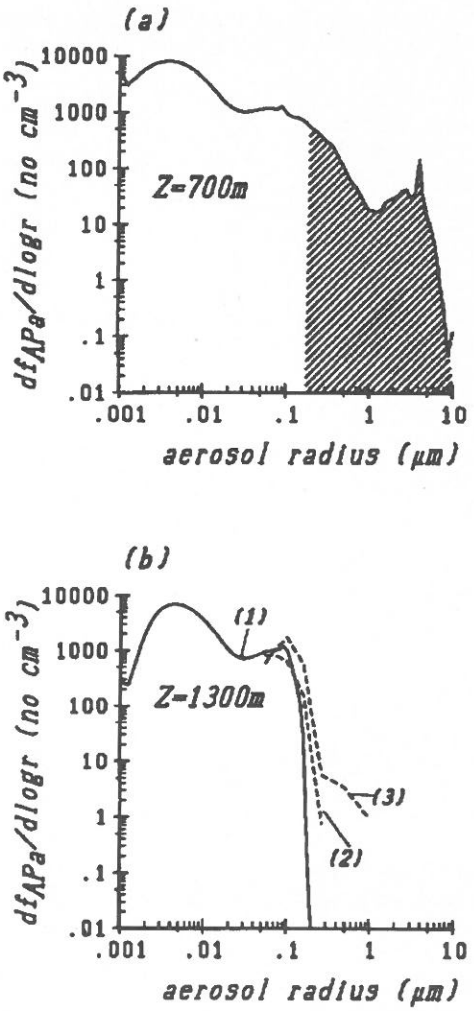


Figure 5 (a) Observed aerosol particle size distribution in air at point P (0,7/8.8) *below* the base of cloud-1. (b) Particle size distribution of the interstitial aerosol in cloud-1 at point P (1.3/8.8) *inside* the cloud. Curve (1) of Figure 5b is computed aerosol spectrum at Z = 1300 m above ground-level (= 1700 m above NN), curve (2) is observed aerosol spectrum at a level of 1500 m above NN inside the cloud, curve (3) is observed aerosol spectrum at Z = 2100 m above NN at the cloud edge.

0.5 % were carried out during various flights at a level of 1 to 3 km. On the average 300 CCN cm^{-3} were observed at the indicated supersaturation. In good agreement with observation we computed with our 2-D cloud model for this level a CCN concentration of 250 to 300 cm^{-3} at a supersaturation of 0.5 % at cloud times between 15 and 20 minutes.

A significant contribution to the understanding of nucleation scavenging could be made by comparing the observed size distribution of the aerosol particles outside (Figure 5a) and inside the cloud (Figure 5b). Comparison of these two figures shows that virtually all aerosol particles with radius larger than 0.2 μm became activated to drops and thus were removed from the aerosol in the air (hatched area in Figure 5a). Looking at Figure 5b we also notice that this "cut-off" of the aerosol particle concentration is quite sharp and excellently predicted by the results of our cloud model for point P (1.3/8.8). The existence of such a cut off has been suggested by Flossmann et al. (1985) and Ahr et al. (1990), using their entraining cloud parcel model and by Flossmann and Pruppacher (1988) and by Flossmann (1991) applying their 2-D cloud model to a case in Hawaii and to Day 261 of the GATE campaign.

In Table 2 we have listed the concentrations of some selected ions observed in the cloud water of cloud-1 at a flight track level of about 1.5 km over NN. Also listed in the table are the concentrations of the same ions predicted for a comparable level after 10 minutes cloud time. We notice that the range and mean values of these concentrations agree well with the theoretical predictions, considering that in our model computations the chemical composition of the cloudwater is only due to aerosol particle uptake. Gas uptake was not considered. The disregard of NH_3 uptake might explain that the observed

Table 2 Concentration of selected ions in the cloud water of cloud-1. Comparison between the observations of Jaeschke and the present model calculations.

Ion	range of concentrations observed (μmol/liter)	range of concentrations computed (μmol/liter)	mean value of observed concentrations (μmol/liter)	mean value of computed concentrations (μmol/liter)
$[SO_4^=]$	100–470	128–496	325	306
$[NO_3^-]$	22–880	168–616	344	306
$[CL^-]$	44–65	37–135	56	55
$[NH_4^+]$	58–955	44–1608	379	1032
$[Na^+]$	43–100	37–135	63	55

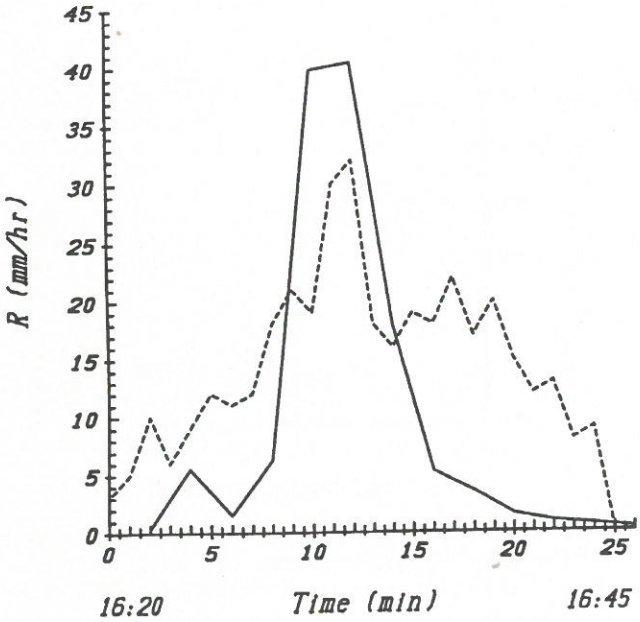


Figure 6 Comparison of the observed (dashed line) and computed (solid line) rain rate on the groundat the point Y = 10 km (P(0./10.)) in the model domain of cloud-2.

concentrations of NH_4^+ are lower than the computed concentrations.

The sounding obtained by interpolation between June 3, 12:00 GMT and June 4, 0:00 GMT caused precipitating clouds to develope. The rate of precipitation produced by model cloud-2 on the location P (0./10.) is illustrated in Figure 6 where comparison is made with the precipitation rate observed between 16:20 and 16:45 GMT. We note that the time period during which the precipitation rate was predicted to have a maximum agrees well with the observation. Unfortunately a quantitative comparison between model prediction and observation could not be made sine the rain event studied was embedded in a large field of precipitating convective clouds whose precipitation overlapped each other. This is the reason for the observed precipitation before and after the main event modelled.

Table 3 Comparison of the concentration of selected ions in the rainwater from cloud-1. Comparison between the observation of Bächmann and the present model calculations.

Ion	range of concentrations observed in rainwater μmol/liter	range of concentrations computed in rainwater μmol/liter	mean value of computed deposition rates in rainwater mg/(m²hr)
$[SO_4^=]$	2–44	1 –45.5	8.8
$[NO_3^-]$	6–58	1.45–46.2	4.8
$[CL^-]$	2–22	0.14–4.6	0.3
$[NH_4^+]$	17–195	3.7–132.6	4.5
$[Na^+]$	1–18	0.1–2.3	0.15

In Table 3 the predicted concentrations of the salt ions in the rainwater on the ground are listed and compared with the concentrations observed. We note that the predicted ranges of concentrations fall well within the concentration ranges observed.

In Figure 7 we have plotted the predicted (at 48 minutes model time) and observed (at 18:00 GMT) concentration of sulfate in rainwater as a function of raindrop size at P (0./10.). At that time the predicted rainrate was 6.7 mm/hr and in fair agreement with the observation. We notice that for drops with radius larger than 275 μm the predicted concentration agreed surprisingly well with the observation.

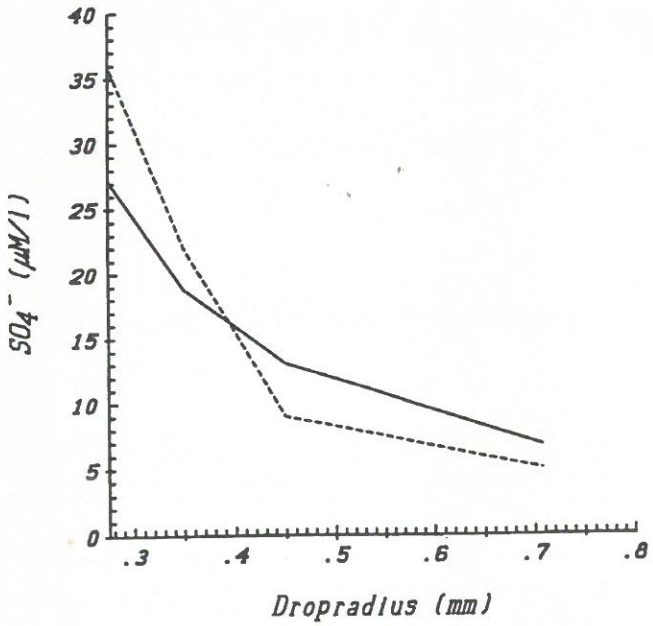


Figure 7 Comparison of the observed (solid line) and computed (dashed line) sulfate concentration as function of drop size in the rain on the ground falling from cloud-2.

Acknowledgement

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