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► **To cite this version:**

S. Berthet, M. Leriche, J.P. Pinty, J. Cuesta, G. Pigeon. Scavenging of aerosol particles by rain in a cloud resolving model. Atmospheric Research, Elsevier, 2010, 96 (2-3), pp.325-336. <10.1016/j.atmosres.2009.09.015>. <hal-00519461>

HAL Id: hal-00519461

<https://hal.archives-ouvertes.fr/hal-00519461>

Submitted on 16 Nov 2010

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SCAVENGING OF AEROSOL PARTICLES BY RAIN IN A CLOUD RESOLVING MODEL

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Abstract

We describe a below-cloud scavenging module of aerosol particles by raindrops for use in a three-dimensional mesoscale cloud resolving model. The rate of particle removal is computed by integrating the scavenging efficiency over the aerosol particle and the drop size distributions. Here the numerical integration is performed accurately with a Gauss quadrature algorithm. The efficiency of the scavenging module is partially confirmed with experimental data. More interestingly, it is illustrated by two numerical experiments: the simulation of a forced convective circulation in a tropical cloudy boundary layer and a two-dimensional simulation of an African squall line. The results show a very selective wet removal of the aerosol particles which clearly depends on the mode radius, the width and the vertical profile of concentration. Furthermore, the squall line case shows the importance of resolving internal

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circulations to redistribute layers of aerosol particles in order to improve estimates of particle removal by below-cloud scavenging.

Key words: Below Cloud Scavenging, Collision efficiency, Gauss quadratures, aerosol transport simulations

1. Introduction

Wet deposition is the most important sink of aerosol particles (AP) in the troposphere (Pruppacher and Klett, 1997). It involves complex micro-physical interactions between AP and hydrometeors which are categorized as nucleation scavenging and impaction scavenging processes. The nucleation scavenging leads to the formation of cloud droplets and ice crystals on cloud condensation nuclei and on ice forming nuclei by heterogeneous nucleation, respectively. The impaction scavenging applies to all the dynamical and physical processes leading to the capture of AP by cloud droplets, ice crystals and mostly by falling hydrometeors. It includes Brownian diffusion, interception, inertial impaction, thermophoresis, diffusiophoresis, airflow turbulence and electrostatic attraction (Pruppacher and Klett, 1997; Andronache, 2003). The nucleation scavenging, by essence an in-cloud mechanism, dominates the impaction scavenging which is efficient once big drops are precipitating at cloud base. So the impaction scavenging is often referred to as a below-cloud scavenging (BCS) process but this does not rule out the in-cloud capture of interstitial AP by raindrops.

This study focuses on the role of BCS of AP by the raindrops thus ignoring similar effects by the snowflakes and other ice particles (see the discussion in section 4.2). We refer to the recent work of Feng (2009) for the scavenging

21 of AP by snow particles of different shapes.

22 This work is part of a larger project, which aims at describing all the pro-
23 cesses leading to the wet deposition of polydisperse AP in a three-dimensional
24 (3D) cloud resolving model.

25 According to Pruppacher and Klett (1997), high values of BCS are found
26 both for ultrafine particles (diameters less than $0.01 \mu\text{m}$) and for coarse par-
27 ticles (diameters larger than $2 \mu\text{m}$). Interestingly, for the intermediate range
28 of particle size referred as the "Greenfield gap", the BCS efficiency decreases
29 by several orders of magnitude, thus making the particles less depleted and
30 so available for in-cloud nucleation scavenging in the upper levels.

31 The explicit computation of BCS rates fits well with the present capa-
32 bilities of cloud resolving/mesoscale models for two reasons at least. First,
33 these models are able to produce 3D rain fields in a consistent way (mixing
34 ratios, and even drop concentrations depending on the microphysical scheme)
35 and second, they resolve in detail the air flow patterns which determine the
36 AP exposure duration of air parcels to BCS removal during rainfall events.
37 This contrasts with estimates of BCS in global or chemistry transport mod-
38 els where crude approximations (see a review by Sportisse (2007)) are made
39 about the size distribution of the raindrops or the rainfall rate estimates
40 (Seinfeld and Pandis, 1998; Jacobson, 2003; Loosmore and Cederwall, 2004;
41 Tost et al., 2006; Henzing et al., 2006; Feng, 2007) to cite a few. In return,
42 considering polydisperse AP and raindrops at each gridpoint of 3D domains
43 of simulation implies double integrations of the BCS rates over the two size
44 distributions (Mircea et al., 2000; Andronache, 2003; Jung et al., 2003). For
45 this reason and because of the vanishing part of the distributions, a special

46 attention needs to be drawn upon computation efficiency and accuracy, as
47 stressed by Sportisse (2007).

48 This study presents a flexible BCS module and illustrates some appli-
49 cations in the 3D mesoscale/cloud resolving model MesoNH (Lafore et al.,
50 1998). First, the BCS module has been developed based on the widely used
51 collision efficiency parameterization of Slinn (1983). The original module
52 employs a quadrature method to integrate over the raindrop and the AP size
53 distributions. Then zero-dimensional tests of the module have been done on
54 a set of rain rates and AP concentration data from the COPS experiment
55 (Wulfmeyer and coll., 2008), which took place in the Vosges-Black Forest
56 area in July 2007 (see <http://www.cops2007.de/>). Finally, the module has
57 been implemented and coupled to the microphysical schemes of MesoNH.
58 Two simulations have been performed to illustrate the BCS module and its
59 potential interactions with the transport. We studied an idealized case of
60 a shallow tropical rainband in a 2D kinematic framework from the HaRP
61 campaign (Cohard and Pinty, 2000) and a squall line case from the COPT81
62 experiment in West Africa (Lafore and Moncrieff, 1989). For both simula-
63 tions, sensitivity tests are performed on the characteristics of the initial size
64 distributions of AP and on the initial vertical profile of AP spectra.

65 **2. DEVELOPMENT OF THE BCS MODULE**

66 *2.1. Total number BCS rate*

67 A BCS module has been developed to describe the wet removal of poly-
68 disperse AP by rain precipitation inside the cloud and especially below the
69 cloud base in the three-dimensional model MesoNH.

70 BCS of AP is related to the AP size distribution but also to the raindrop
71 size distribution through the BCS coefficient. Here a difficulty arises due to
72 the complex mathematical expression of the BCS coefficient which must be
73 integrated over wide distributions, up to infinity for analytical, log-normal
74 or gamma laws. In order to simplify crudely the computation of a BCS rate,
75 Tost et al. (2006) and Loosmore and Cederwall (2004) assume a monodisperse
76 distribution of the raindrops (or equivalently an empirical "representative"
77 raindrop diameter as shown in Fig. 4 of Sportisse (2007)) to determine the
78 BCS coefficient $\gamma(d_p)$ of AP of size d_p and the falling raindrops. A less empiri-
79 cal method used by Andronache (2003) and by Feng (2007) takes explicitly
80 into account the spectral nature of the raindrop distribution by performing
81 a numerical integration up to a large raindrop diameter. In order to keep
82 an accurate estimate of BCS effects in MesoNH, we choose to compute BCS
83 coefficients by means of a Gauss quadrature method (Press et al., 1992). In-
84 finite integrals for BCS coefficients are computed as a weighted sum for a
85 small number (here ~ 20) of optimized abscissas (each abscissa is deduced
86 from a particle or drop diameter by a change of variable). Then, the final
87 BCS rate of AP is obtained by integrating the BCS coefficient $\gamma(d_p)$ over the
88 AP size distribution using another Gauss quadrature, adapted to the AP size
89 distribution.

90 The AP are represented by the sum of several log-normal distributions
91 $n_{pi}(d_p)dd_p$ depending on the number of modes:

$$dN_p = \sum_{i=1}^l n_{pi}(d_p)dd_p = \sum_{i=1}^l \frac{N_{pi}}{\sqrt{2\pi}d_p \ln\sigma_i} e^{-\left(\frac{\ln(d_p/\overline{d}_{pi})}{\sqrt{2\ln\sigma_i}}\right)^2} dd_p$$

92 where l is the number of modes with index i , d_p is the AP diameter, N_{pi} ,

93 the number concentration, σ_i , the geometric standard deviation of the log-
 94 normal distribution, and $\overline{d_{pi}}$ is the median diameter. For the time being,
 95 we use a single-moment scheme for the AP in MesoNH (the number con-
 96 centration N_p of each mode is a prognostic variable), so σ_i and $\overline{d_{pi}}$ are kept
 97 constant, depending on the AP types¹. Traditionally three modes ($l = 3$)
 98 are superimposed: the Aitken nuclei mode, the accumulation mode and the
 99 coarse mode.

100 The raindrop size distribution $n_R(D_d)$ in MesoNH is modeled by a gen-
 101 eralized gamma distribution $n_R(D_d) = (\alpha/\Gamma(\nu)) \lambda_R^{\alpha\nu} D_d^{\alpha\nu-1} \exp(-\lambda_R D_d)$. In
 102 this study, $n_R(D_d)$ is reduced to the classical Marshall-Palmer law by taking
 103 $\alpha = \nu = 1$ for the two shape parameters:

$$n_R(D_d)dD_d = N_0 e^{-\lambda_R D_d} dD_d \quad (1)$$

104 where $N_0 = 8 \times 10^{-3} m^{-3} mm^{-1}$, λ_R is the slope parameter and D_d the drop
 105 diameter.

106 The method used to calculate the BCS coefficient follows the concept
 107 of the collision efficiency E between an AP and a raindrop (Slinn, 1983;
 108 Pruppacher and Klett, 1997; Seinfeld and Pandis, 1998). E expresses the
 109 number of AP collected by the falling raindrop in the volume of air swept
 110 out by the raindrop. A value of $E = 1$ implies that the raindrop collects
 111 all the particles encountered along the path of the fall. Most of the recent

¹The residency time of falling raindrops ($H/V_{fall} \sim 2 - 5 \times 10^2$ s where $H \sim 1000$ m is the height of the cloud base and $V_{fall} \sim 2 - 5$ m/s, the fallspeed of the raindrops) is short compared to the time scale of the BCS ($\Lambda^{-1} > 10^4$ s according to Sportisse (2007)) so, a possible modification of the median diameter $\overline{d_{pi}}$ of the AP size distribution is clearly a second-order effect compared to the variability of the rainfall rates.

112 studies about BCS are based on this concept (Andronache, 2003; Loosmore
 113 and Cederwall, 2004; Henzing et al., 2006; Tost et al., 2006; Feng, 2007).

114 There is no available theoretical solution of the Navier-Stokes equation for
 115 the prediction of E . Therefore Slinn (1983) has formulated a semi-empirical
 116 expression of the collision efficiency E , taking into account three of the most
 117 understood collection processes: the Brownian diffusion, the interception and
 118 the inertial impaction. Some studies (Slinn and Hales, 1971; Wang et al.,
 119 1978; Tinsley et al., 2005) describe the relative impact of other processes
 120 such as thermo-, diffusiophoresis and electric forces upon E . The inclusion
 121 of these processes means to estimate the electric charge of each particle and to
 122 determine the local relative humidity along particle trajectories, complicating
 123 heavily the computation of E . In general these processes are ignored, as in
 124 this study. After non-dimensionalizing the equation of motion for the air, the
 125 AP and the raindrops, Slinn (1983) found an expression of E that depends
 126 on five dimensionless parameters :

$$127 \quad Re = \frac{D_d U_t(D_d) \rho_a}{2 \mu_a}, \quad Sc = \frac{\mu_a}{\rho_a \mathcal{D}}, \quad St = \frac{2 U_t(D_d) \tau_a}{D_d}$$

$$\phi = \frac{d_p}{D_d}, \quad \omega = \frac{\mu_w}{\mu_a}, \quad \text{and} \quad St^* = \frac{1.2 + (1/12 \ln(1 + Re))}{1 + \ln(1 + Re)}$$

128 with symbols defined in Table 1. Seinfeld and Pandis (1998) or Sportisse
 129 (2007) provide expressions for \mathcal{D} and τ_a which depend on d_p . In MesoNH,
 130 the terminal velocity of raindrops of diameter D_d is approximated by a sim-
 131 ple power law dependence in diameter that includes the air density effect
 132 expressed by Foote and Du Toit (1969):

$$U_t(D_d) = a D_d^b \left(\frac{\rho_{a0}}{\rho_a}\right)^{0.4}$$

133 Finally the analytical Slinn's expression of E that fits experimental data
 134 is expanded as:

$$\begin{aligned}
 E(D_d, d_p) = \frac{4}{Re Sc} & \left[1 + 0.4Re^{1/2}Sc^{1/3} + 0.16Re^{1/2}Sc^{1/2} \right] \\
 & + 4\phi \left[\omega^{-1} + (1 + 2Re^{1/2})\phi \right] \\
 & + \left[\frac{St - St^*}{St - St^* + 0.667} \right]^{3/2} \left(\frac{\rho_p}{\rho_w} \right)^{1/2}
 \end{aligned} \tag{2}$$

135 The analytical expression of the collision efficiency E contains three terms
 136 corresponding to distinct physical contributions depending on the AP diam-
 137 eter d_p :

- 138 1. The first term is the contribution of the *Brownian diffusion*. It de-
 139 notes the random motion of small particles under the influence of the
 140 surrounding fluid molecules. It dominates for AP with $d_p < 0.01\mu m$.
- 141 2. The second term is the collection by *interception*. The particles follow
 142 the turbulent air motion in the wake of the drops. They are collected
 143 when their trajectory is close enough to the drop. This process affects
 144 AP with diameter between 0.01 and 2 μm including AP in the "Green-
 145 field gap".
- 146 3. The third term refers to the *inertial impaction*. It occurs when the
 147 particles do not follow the mean air flow because of their inertia that
 148 takes them off the streamlines. This term is efficient for large particles
 149 ($d_p > 2\mu m$) with a restriction to cases where $St > St^*$ in Eq. 2 (Feng,
 150 2007).

151 Wet deposition by BCS is a 1st order decay process so that,

$$\frac{\partial \psi(d_p, t)}{\partial t} = -\gamma(d_p)\psi(d_p, t) \tag{3}$$

152 where $\psi(d_p, t)$ is the AP size distribution in number or mass. $\gamma(d_p)$ is the BCS
 153 coefficient for particles of diameter d_p . It results from the integration over
 154 the raindrop size distribution of the collision efficiency E times the volume
 155 swept by a falling raindrop D_d per unit of time,

$$\gamma(d_p) = \int_0^\infty \frac{\pi}{4} D_d^2 U_t(D_d) E(D_d, d_p) n_R(D_d) dD_d \quad (4)$$

156 A Gauss-Laguerre algorithm (Press et al., 1992), a variant of the Gauss
 157 quadrature technique, is used for the first time to integrate the above expres-
 158 sion. The method is based on a n -orthogonal polynomial set associated to
 159 the weighting function, here the size distribution of the raindrops (Eq. 1).
 160 The method is a high-order optimal one which is exact when the function to
 161 integrate is smoother than a polynomial of degree $(2n - 1)$. The χ_i symbols
 162 denote the weights and x_i , the abscissas, then :

$$\int_0^\infty x^\beta \exp(-x) f(x) dx \simeq \sum_{i=1}^n \chi_i f(x_i).$$

163 Finally, the BCS coefficient (Eq. 4) is integrated over the AP diameters
 164 d_p to obtain the BCS rate with respect to the total number of AP using
 165 Eq. 3:

$$\left. \frac{\partial N_p}{\partial t} \right|_{BCS} = \int_0^\infty \left. \frac{\partial n_p(d_p)}{\partial t} \right|_{BCS} dd_p = - \int_0^\infty \gamma(d_p) n_p(d_p) dd_p. \quad (5)$$

166 This time, the integration is performed using a Gauss-Hermite quadrature
 167 formula with appropriate weights χ'_i and abscissas x'_i :

$$\int_0^\infty \exp(-x'^2) f(x') dx' \simeq \sum_{i=1}^n \chi'_i f(x'_i).$$

168 As confirmed by a recent study (Andronache, 2003), Fig. 1 shows the
 169 BCS coefficient $\gamma(d_p)$ for increasing rainfall rates R from 0.1 to 100 mm.hr⁻¹.

170 The rainfall rate $R = \int_0^\infty (\pi/6)(\rho_w/\rho_a)D_d^3U_t(D_d)n_R(D_d)d(D_d)$ is a function
 171 of the slope parameter λ_R which defines the Marshall-Palmer distribution
 172 of the raindrops (see Eq.1). Three distinct regimes of the BCS coefficient,
 173 corresponding to those of the collision efficiency E , i.e. *Brownian motion*,
 174 *interception* and *inertial impaction*, can be seen in Fig. 1 with a marked
 175 "Greenfield gap" for $0.1 \mu\text{m} < d_p < 1 \mu\text{m}$. The coarse AP are the most
 176 efficiently scavenged, the ultrafine particles are moderately scavenged and
 177 the scavenging is low for the intermediate mode. Moreover one can notice
 178 in Fig. 1 that the logarithm of the BCS coefficient is proportional to the
 179 logarithm of R , with a good approximation. These results are in agreement
 180 with those of previous studies (Andronache, 2003; Loosmore and Cederwall,
 181 2004; Tost et al., 2006; Feng, 2007).

182 2.2. Mass BCS rate

183 The computation of the AP scavenging rate allows us to study the evo-
 184 lution of the number concentration for each AP mode ($N_{free} = N_p$) and
 185 also to estimate the total mass of AP (m_{scav}) captured by the drops ; m_{scav}
 186 constitutes the depleted part of the AP in mass.

187 The budget equation of m_{scav} is composed of three terms: a transport
 188 tendency (marked "*transp*"), a mass BCS removal term ("*BCS*") and an
 189 additional sedimentation term ("*sed*") because m_{scav} moves at the terminal
 190 fall speed of the drops.

$$\frac{\partial m_{scav}}{\partial t} = \left. \frac{\partial m_{scav}}{\partial t} \right|_{transp} + \left. \frac{\partial m_{scav}}{\partial t} \right|_{BCS} + \left. \frac{\partial m_{scav}}{\partial t} \right|_{sed}$$

191 The mass of the free AP which are scavenged by rain, is also computed

192 with the Gauss-Hermite quadrature formula, so according to Eq. 3, one gets:

$$\frac{\partial m_{scav}}{\partial t} \Big|_{BCS} = - \frac{\partial m_{free}}{\partial t} \Big|_{BCS} = \frac{\pi}{6} \rho_p \int_0^\infty d_p^3 \gamma(d_p) n_p(d_p) dd_p$$

193 where both AP sphericity and constant density for each particle mode are
194 assumed.

195 The mass sedimentation rate of the scavenged particles is taken propor-
196 tional to the sedimentation rate of the raindrop mixing ratio q_R :

$$\frac{1}{m_{scav}} \frac{\partial m_{scav}}{\partial t} \Big|_{sed} = \frac{1}{q_R} \frac{\partial q_R}{\partial t} \Big|_{sed},$$

197 suggesting that statistically, the mass of scavenged AP contained in an indi-
198 vidual drop of size D_r is proportional to the mass of the drop.

199 3. QUALITATIVE APPLICATION TO THE COPS EXPERIMENT

200 COPS (Convective and Orographically-induced Precipitation Study) was
201 a 3 month international field campaign with part of a program aiming at
202 improving precipitation forecasts (Wulfmeyer and coll., 2008). COPS took
203 place in Summer 2007 in south-western Germany and north-eastern France.
204 One of the goals of COPS was to collect time series of high resolution surface
205 data including as many aerosol, cloud, and precipitation variables as possible,
206 to be used as lower boundary conditions for mesoscale models.

207 Looking at the BCS process, we are interested by two types of COPS
208 data that are both in situ measurements at ~ 3 m above ground level :

- 209 • Mean rainfall rate calculated each 10 min from precipitation measure-
210 ments with an Optical Rain Gauge (ORG);

211 • Time evolution of AP concentrations sampled by a Grimm optical par-
212 ticles counter with a time step of 1 min. The AP concentration is
213 provided for 15 classes of particles: size cut radius (μm) are [0.15; 0.2;
214 0.25; 0.325; 0.4; 0.5; 0.8; 1; 1.5; 2; 2.5; 3.75; 5; 7.5; 10; 20].

215 After comparing the two types of COPS data (precipitation intensity
216 and AP concentrations), some sequences have been selected for which the
217 aerosol depletion could be attributed to the wet removal by precipitation:
218 the assumption is made that other processes could be neglected because of
219 the short time duration of these sequences. Fig. 2 shows the COPS AP
220 concentration data for Julian day 184.38 to 184.8 as well as the precipitation
221 rate attesting the importance of scavenging for large AP.

222 For the moment, only the first precipitation event (between 184.38 and
223 184.5) is examined. During this rainfall event, the AP concentration of classes
224 1 to 8 ($r_p < 1.5 \mu m$), is not perturbed. In contrast, the AP concentrations
225 of large diameters (classes 9 to 11) vary and are depleted according to the
226 rainfall event. This depletion is partly explained by BCS because, as shown
227 in Fig. 1, the BCS rate is the highest for large diameters (classes 9 to 11)
228 while small ones (1 to 8) correspond to the "Greenfield gap". Thus, the
229 data suggest the potential effect of BCS upon the evolution of AP in the
230 troposphere. Therefore, the implementation of this process in the aerosol
231 continuity equation is necessary in cloud resolving models such as MesoNH.

232 In order to simulate the depletion of the AP by BCS using the first pre-
233 cipitation event from the COPS data, a mean particle diameter $\overline{d_m}$ has been
234 defined for each class and the corresponding BCS coefficient $\gamma(\overline{d_m})$ calcu-
235 lated. The method consists in initializing the AP concentrations with mea-

236 sured values. Then the mean rainfall intensity data are incorporated in the
237 BCS module for the selected rainy episode. The temporal evolution of the
238 AP concentrations of each class of size N_i is only driven by the calculated
239 BCS rates.

240 For each class, the percentage of AP concentration depleted during the
241 rainfall event, is calculated by comparing the AP concentration at the begin-
242 ning and at the end of the event. This is done for measured and simulated
243 data. As a result, the computed percentage does not reflect the entire de-
244 pletion of each class of the COPS measurements. Nevertheless, the influence
245 of the BCS effect is reproduced selectively as a function of the particle class
246 diameter. For class number 3 (accumulation mode) and class number 10
247 (coarse mode) for instance (see the legend above Fig. 2), the simulated de-
248 pletion percentages are 0.1% and 87% whereas the measured ones are 10.8%
249 and 54%, respectively. The discrepancy between the measured and simulated
250 trends can be attributed to other processes such as turbulent transport, mi-
251 crophysics and to the assumption of homogeneous vertical profiles in the
252 precipitating zone.

253 4. 2D SIMULATIONS WITH MesoNH

254 The numerical experiments are performed to test and to evaluate the
255 impact of BCS on a multimodal AP population. In all cases, particles are
256 transported by a moist flow defined by warm or mixed-phase microphysical
257 cycles producing rainfalls of varying characteristics along the course of the
258 simulation. The rain mixing ratio ($q_R = \int_0^\infty (\pi/6)(\rho_w/\rho_a)D_d^3 n_R(D_d) d(D_d)$)
259 is used to determine the raindrop size distribution $n_R(D_d)$ and to integrate

260 the BCS term (Eqs. 4 and 5) in MesoNH.

261 *4.1. Warm shallow convection: the "HaRP" test case*

262 The idealized "HaRP" test case comes from "The Hawaiian Rainband
263 Project" that took place in 1990 (Szumowski et al., 1998). It aims at simu-
264 lating a precipitating cell forced by an idealized time-varying non-divergent
265 circulation. The duration of the simulation is 50 min. The numerical ex-
266 periments are performed with the MesoNH model using a standard Kessler
267 scheme for the microphysics and a highly accurate PPM scheme for the trans-
268 port of the scalar fields. The computational domain extends over 180×60
269 gridpoints with a spacing of 50 m in the x and z directions. The time step
270 is 5 sec. The concentrations of the particle modes are continuously replen-
271 ished through the open lateral boundaries in the lowest 250 m where inflow
272 conditions occur.

273 A first run is illustrated in Figs 3a-d showing the particle concentrations
274 (grey scale), the rain mixing ratios (contours of 0.1, 0.2, 0.5 and 1.0 g/kg)
275 and the flow circulation with arrows. The simulation is initialized for a
276 single log-normal distribution of AP with a median diameter of $2 \mu\text{m}$. The
277 geometric standard deviation $\sigma = 2$ and the initial concentration $N = 3$
278 cm^{-3} are representative of marine conditions (see mode "b" in Table 1 of
279 Andronache (2003)). At the beginning of the simulation, the moist flow
280 converges horizontally in the low levels initiating the formation of a cloud
281 cell. Quickly after reaching the autoconversion threshold, the cloud droplets
282 convert partially into raindrops which start to precipitate and to evaporate
283 below cloud base. At the same time, the AP are entrained in the ascending
284 motion of the flow (see the AP concentration in Fig. 3a taken after 20 min).

285 After 25 min (Fig. 3b), the AP continue to rise through the rainshaft where
286 they are partially depleted by BCS. Then the rain reaches its maximum
287 intensity at 35 min therefore leading to an efficient depletion of AP by BCS
288 in Fig. 3c. At the end of the simulation (so after 50 min in Fig. 3d), the AP
289 flux diverges at the top of the cloud showing a nearly 50 % reduction of the
290 AP concentration by BCS.

291 In summary, the 2D simulation reproduces an interesting situation where
292 AP are transported and accumulated by a forced convective flow in a region
293 where raindrops are formed, precipitate and then lead to BCS. A key-aspect
294 of our BCS treatment is that at high resolution, the BCS efficiency in a
295 2D/3D precipitating system is thoroughly linked to the flow pattern and to
296 the microphysical scheme. This aspect was underevaluated in most previ-
297 ously cited studies where observed or simulated rainfall rates are used to
298 estimate the BCS rate of AP.

299 A generalization of this "HaRP" case study has been undertaken in order
300 to explore the sensitivity of several AP size distributions to the BCS. In this
301 series of experiment, the median diameter $\overline{d_p}$ varies logarithmically in the
302 interval $10^{-3} - 10^1 \mu\text{m}$ while the geometric standard deviation σ is taken in the
303 [1-6] range (see the compiled size distribution cases of Andronache (2003)).
304 The results obtained for a Marshall-Palmer raindrop size distribution are
305 plotted in Fig. 4. The figure represents the percentage of removed AP after
306 50 min of simulation. These scavenged AP concentrations are time and space
307 averaged in the regions where the rain mixing ratio is larger than 0.01 g/kg.

308 Inspection of Fig. 4, shows a continuous enhancement of the BCS with σ
309 for $\overline{d_p} < 1 \mu\text{m}$. This feature results from the fact that increasing the width

310 of the AP log-normal distribution and holding $\overline{d_p}$ fixed, increases the AP
 311 size interval over which the scavenging efficiency is integrated (see Fig. 1).
 312 Consequently and because the weighting effect of the scavenging coefficient
 313 of the smallest particles dominates, the BCS increases with σ . The situation
 314 appears more complicated in the "Greenfield gap" interval $0.1 - 2 \mu\text{m}$ of $\overline{d_p}$
 315 where the BCS efficiency of AP is reduced. In this region however, the large
 316 sensitivity of the AP size distributions is fully explained by the head and the
 317 tail of the AP size distribution which involve higher and higher scavenging
 318 efficiencies as σ increases. Moving to $\overline{d_p} > 2. \mu\text{m}$, i.e. to coarse AP mode,
 319 one can notice that the BCS is no more dependent on the value of σ . $\overline{d_p}$ is
 320 the leading parameter for the BCS in this AP size range. As a result, the
 321 three distinct regimes which show up in Fig. 4 are clearly correlated to the
 322 variations of the scavenging coefficients shown in Fig. 1.

323 A similar series of simulation performed with a Gamma ($\alpha = 1, \nu = 2$)
 324 raindrop size distribution (Eq. 1) leads to a further slight enhancement of
 325 BCS (not shown here). This effect is explained simply by considering the
 326 higher concentration of big drops in the "Gamma" case compared to the
 327 Marshall-Palmer case.

328 The "HaRP" results show the difficulties to predict the potential scav-
 329 enging of multiple AP modes without an accurate BCS module coupled to a
 330 microphysical scheme and taking into account the transport of the particles.
 331 The numerous combinations of AP log-normal distribution parameters and
 332 rain intensity clearly leads to a large variety of scavenging efficiencies.

333 *4.2. Tropical squall line: the "COPT" test case*

334 The "COPT" test case (Caniaux et al., 1994) is typically a 12 hour simu-
335 lation of a tropical squall line with kilometer scale-resolved internal circula-
336 tions, a 2D turbulence scheme and mixed-phase microphysics. The model is
337 initialized with 3 successive layers of aerosol of 2 km depth starting from the
338 ground level. For each layer, the same multimodal population of particles
339 corresponding to the "Dust Layer" case reported by Andronache (2003), is
340 chosen for the simulation of an African squall line. The domain contains
341 320×44 gridpoints unevenly spaced in the vertical ($\Delta z = 70$ m at ground
342 level and $\Delta z = 700$ m above 12 km). The horizontal resolution is 1.25 km.
343 The model is integrated with a time step of 10 sec. A gravity wave damping
344 layer is inserted between 17 km and the model top at 22.5 km. A constant
345 speed transformation is used to compensate for the motion of the squall line.
346 No fluxes are considered in the surface layer. Convection is initiated by form-
347 ing a -0.01 K.s^{-1} artificial cold pool in the low levels of a small domain during
348 10 min.

349 Although liquid drops and frozen particles can coexist in a squall line, the
350 study focuses on the BCS of AP by rain. In mixed-phase clouds, impaction
351 scavenging by ice is enhanced by phoretic effects (Martin et al., 1980) and
352 probably, by electric forces (Miller, 1990; Andronache, 2004) as these clouds
353 become electrified by non-inductive charging mechanisms involving ice-ice
354 shocks (Mansell, 2000; Barthe et al., 2005). This last effect clearly makes the
355 in-cloud impaction scavenging of interstitial AP by ice difficult to describe in
356 detail. However as the ice nucleation scavenging should dominate according
357 to Alheit et al. (1990), the impaction scavenging of AP by ice particles is

358 probably of secondary importance. As a result and because we concentrate on
359 (tropical) warm precipitation events, in- and below-cloud scavenging effects
360 of AP by ice and snow are not considered in this study.

361 The aim of this case experiment is to explore the BCS efficiency of AP
362 layers in a long-lived mixed-phase precipitating system. In this case, we show
363 that the residency time of air parcels (see the discussion about the merits of
364 this aging tracer in Lucas-Picher et al. (2008)) is a useful tool to interpret
365 the temporal aspects of the AP removal in rainy areas. Therefore, a series of
366 simulations, without (SCAV₀) and with (SCAV₁) application of BCS to the
367 dust particles, are performed. In each case, the dust particles are transported
368 by the two-dimensional circulation that develops in the organized convective
369 system. Figs 5a-c show the difference of dust concentrations between the
370 twin simulations SCAV₀ and SCAV₁ taken after 9 hours of simulation. The
371 plots of AP field differences allows to picture the net BCS component of the
372 dust particle budget in a complex precipitating system.

373 The squall line develops in a series of narrow convective cells on the left
374 followed by a large stratiform area with much less precipitation on the right
375 of the figures. Here, the plotted field difference (SCAV₀-SCAV₁) focuses on
376 the coarse mode concentration of the dust particle distribution. The plotted
377 fraction of the dust particles is always positive because of the reduced fraction
378 of particle concentration when the BCS process is considered. Figs 5a-c differ
379 only by their respective initialization: the initial dust layer was between [0-2]
380 km for Fig. 5a, [2-4] km for Fig. 5b and [4-6] km for Fig. 5c. To summarize,
381 the figures are showing at the same scale, the net scavenging efficiency of
382 each initial layer of coarse dust particle mode ($d_{p1} = 0.55 \mu\text{m}$, $\sigma = 2.5$).

383 The upper glaciated part of the cloud system and the raining cells are also
384 depicted in the figures. The initial concentration of the coarse mode is 20
385 cm^{-3} in all the simulations. A rapid inspection of Figs 5a-c indicates that
386 all the dust layers are affected by the scavenging process which is efficient
387 enough to eliminate much more than 1 cm^{-3} (scale maxima) of coarse mode
388 particles in the heavy precipitating convective region of the squall line.

389 As expected the lowest dust layer (Fig. 5a) is the most affected by the
390 scavenging removal because of stronger convergence and circulation flow in-
391 tensity in the lower levels, and a longer time exposure to the precipitating
392 area. Another interesting feature displayed in Figs 5a-c is the fate of the net
393 scavenged particles in the upper glaciated region of the squall line, that is
394 well above the ice freezing level at 4.5 km height. In Fig. 5a the relatively
395 large difference of concentration well above the ice melting layer reflects the
396 indirect scavenging impact in the upper glaciated region : the non-scavenged
397 particles in simulation SCAV_0 are transported upwards while they are de-
398 pleted in simulation SCAV_1 due to efficient BCS in the low levels. Therefore
399 the particle concentration in the squall line upper region is clearly sensitive
400 to BCS of low levels AP sources with potential feedback on the ice nucleation
401 process by selectively modifying the ice nuclei spectra (for instance contact
402 freezing nuclei which efficiency depends on particle size). The same analysis
403 holds for Fig. 5b with a lower scavenging efficiency and less upward transport
404 of the "mid-level" particle layer in the convectively rainy region (leftmost pre-
405 cipitating cells) because of a shorter residency time. In contrast, scavenging
406 is enhanced in the trailing stratiform part of the squall line (right part of
407 the [0-2] km layer in Figs 5a-b). This feature is explained by the presence of

408 bigger raindrops (raindrops partially evaporated during their fall) which are
409 more efficient to scavenge AP in the [2-4] km layer.

410 Fig. 5c shows that the BCS process still perturbs the field of AP when
411 the dust particles are concentrated in the [4-6] km layer. Surprisingly the
412 low-level rainy area of the squall line where convective cells develop, reveals a
413 lot of scavenged particles even if they were not present initially in this region.
414 The explanation is that the dust particles are recycled by the density current
415 of the squall line (Lafore and Moncrieff, 1989). Clearly speaking, it is the
416 internal dynamics of the squall line (convective cells, mesoscale subsidence
417 and rear-to-front flow below the stratiform region) that transports the air and
418 the dust of the midlevels in a downward branch, and which is responsible for
419 the BCS depletion in the rainy area ahead. This effect is illustrated in Fig. 6
420 where the time evolution of a selected vertical profile (taken at $x = 225$
421 km) of the [4-6] km AP layer, is plotted with the rain mixing ratio. Three
422 stages are identified in the figure. During the first 3 hours of simulation,
423 the initial vertical distribution of AP is relatively undisturbed. Then, AP
424 are appearing in the lower(upper) levels due to down(up)ward transport by
425 the first convective cells of the squall line. Finally after 6 hours, the AP are
426 continuously filling the atmosphere up to 12 km height. During this period
427 of time, rainy cells are precipitating in the lowest 4.5 km allowing raindrops
428 to scavenge the transported AP.

429 As expected, BCS is less affecting the upper glaciated region of Fig. 5c
430 in comparison to that of Fig. 5a. The reason is that rain comes from the
431 melting of graupel particles at 4.5 km altitude leading to a very shallow layer
432 where BCS can operate locally during the ascent of the dust particles.

433 In order to illustrate the importance of transport, here combined to BCS
434 in the squall line (see also Lafore and Moncrieff (1989)), the residency time of
435 air parcels having been in contact with rain, is shown in Fig. 5d after 9 hours
436 of simulation with half-hourly contours starting after 2 hours of simulation.
437 The residency time (RT) is a 3D tracer which is easy to integrate along
438 the course of the simulation. It includes a transport term followed by a
439 conditional time incrementation step, here characterized by a "rain" mask.
440 At each time step, the rainy areas are selected by considering the gridpoints
441 passing a mask where the rain mixing ratio is larger than 0.01 g/kg and so
442 where RT is augmented by the value of the timestep.

443 Without "rain" mask (not shown here), the bulk structure of RT repro-
444 duces the mean wind shear with inflow (outflow) conditions below (above)
445 7.5 km height at the left boundary. More interestingly with the "rain" mask,
446 there is a close resemblance between the fine scale structures of RT in Fig. 5d
447 and the scavenged AP fields in Fig. 5a-c. For instance the wedge pattern of
448 the scavenged AP in the low levels is reflected by a continuous increase of
449 RT at the same place. Similarly, holes of scavenged AP at the melting level
450 (4.5 km) correspond approximately to a reduced RT, meaning that fresher
451 air parcels are less depleted by BCS. Finally, Fig. 5d reveals that a great
452 amount of air in the glaciated part of the squall line has been exposed to the
453 rain during 30 min at least. This demonstrates that the amount of scavenged
454 AP depends on the rainfall rate but that it is also strongly correlated to the
455 residency time of AP under the rainshaft.

456 The last point to examine is the mass of scavenged AP by wet deposition
457 within the underlying surface of the COPT squall-line. Fig. 7a presents the

458 surface cumulated mass of scavenged AP, assuming a density $\rho_p = 2600$
459 kg.m^{-3} , for the 3 layers of AP after 9 hours of simulation. The cumulated
460 rainfall, peaking slightly downwind at 82 mm.hr^{-1} , is plotted in Fig. 7b for
461 comparison. Fig. 7a shows that BCS is most pronounced for the mid layer
462 ([2-4] km) case where it reaches 0.32 kg.m^{-2} . Surprisingly, more AP are
463 scavenged from the upper layer ([4-6] km) than from the lower layer ([0-2]
464 km). The explanation comes from the observation that the big drops which
465 are the most efficient to scavenge AP, are present in the top layer while they
466 are partially evaporated when they reach the bottom layer.

467 In summary, the COPT case offers many opportunities to investigate
468 different aspects of AP scavenging by rain in an organized convective sys-
469 tem where resolved flow dynamics and transport of tracers are well analysed
470 (Lafore and Moncrieff, 1989). Here the implementation of a performant PPM
471 transport scheme in the model is also crucial to get a deep insight into the
472 complexity of the net BCS aspect of AP at mesoscale.

473 5. CONCLUSIONS

474 A BCS module has been implemented in the mesoscale/cloud resolving
475 model MesoNH. The module is based on the collection efficiencies of Slinn
476 (1983) with an accurate and optimized numerical integration over unbounded
477 raindrop and AP size distributions. Then the study explores the sensitivity
478 of BCS to various aspects of the initial AP size distribution and for two
479 precipitating events. Here in contrast with 0D/1D studies and experiments
480 performed at larger scale, the combined effect of transport and BCS acting
481 on AP is stressed and illustrated in the framework of 2D simulations.

482 The HaRP numerical experiment underlines the size dependent deple-
483 tion of an AP population by BCS as illustrated by changing the median
484 diameter of log-normal distributions: the ultrafine particles are depleted, the
485 intermediate ones remain unaltered and the large particles are scavenged
486 dramatically. The results show also a significant sensitivity to the standard
487 geometric deviation parameter of the AP size distributions. The simulation
488 of the COPT squall line demonstrates the importance of coupling transport
489 and microphysics to track the BCS effects in organized precipitating system.
490 Here the location, with respect to the freezing level, of a layer of dust parti-
491 cles is a key-parameter to analyse the net effect of BCS. Some details of the
492 depleted AP fields can be interpreted with the help of the time residency of
493 air parcels in the rainshaft.

494 In conclusion, BCS of particles is expected to remove giant condensation
495 nuclei (sea salt particles) and large ice forming nuclei (mineral dust particles)
496 by precipitation with possible consequences on the nucleating properties of
497 drizzling marine stratus and convective clouds with an ice phase. Therefore
498 the next step of this work will address questions about the nucleation scav-
499 enging processes. The combination between BCS and the activation of cloud
500 condensation nuclei is still an intriguing process in the short range evolution
501 of the cloud cover of clean precipitating marine boundary layers (e.g. the
502 formation of open cells in marine stratocumulus (Rosenfeld et al., 2006)).

503 **Acknowledgements**

504 The authors are deeply grateful to T. Maric, now at University of Wash-
505 ington, Seattle, WA, for the implementation of the PPM transport scheme of

506 Colella and Woodward (1984) in MesoNH. We also thank C. Galy-Lacaux of
507 Laboratoire d’Aérodologie for useful discussion on the wet deposition of aerosol
508 particles and J. Duron for her precious help in preparing some of the figures.
509 Thanks are also due to anonymous referees who helped us to improve in depth
510 a first version of the manuscript. This research was financially supported by
511 University Paul Sabatier, Toulouse, France.

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$\begin{cases} a = 842 \\ b = 0.8 \end{cases}$	Parameters of $U_t(D_d)$	N_{pi}	Number concentration of AP mode i
d_p	AP diameter	$n_{pi}(d_p)$	AP size distribution of mode i
$\overline{d_{pi}}$	Median diameter of mode i	$n_R(D_d)$	Raindrop size distribution
D_d	Drop diameter	q_R	Rain mixing ratio
\mathcal{D}	AP diffusivity	R	Rainfall rate
$E(D_d, d_p)$	AP-droplets collision efficiency	Re	Raindrop Reynolds number
$f(x_i)$	Function to integrate	r_p	AP radius
i, l	Index of AP mode	Sc	AP Schmidt number
m_{free}	Mass of free AP	St	AP Stokes number
m_{scav}	Total mass of scavenged AP	St^*	Critical Stokes number
n	Order of the Gauss quadrature	$U_t(D_d)$	Terminal fall velocity of raindrops
N_0	Marshall-Palmer parameter	x_i, x'_i	Abcissa for Gauss quadrature
N_{free}	Number concentration of free AP		
α	Parameters of $n_R(D_d)$	$\Psi(d_p, t)$	Mass or number AP size distribution
β	Parameter of the Gamma function	ρ_{a0}	Reference air density at ground level
$\gamma(d_p)$	BCS coefficient	ρ_a	Reference density of dry air
Γ	Gamma function	ρ_p	AP density
λ_R	Marshall-Palmer slope parameter	σ_i	Standard deviation of mode i
μ_a	Dynamic viscosity of dry air	τ_a	Relaxation time of the collected AP
μ_w	Water viscosity	χ_i, χ'_i	Weights for Gauss quadrature
ν	Parameters of $n_R(D_d)$	ω	Viscosity ratio
ϕ	Ratio of diameters		

Table 1: Definition and unit of symbols used in this article.

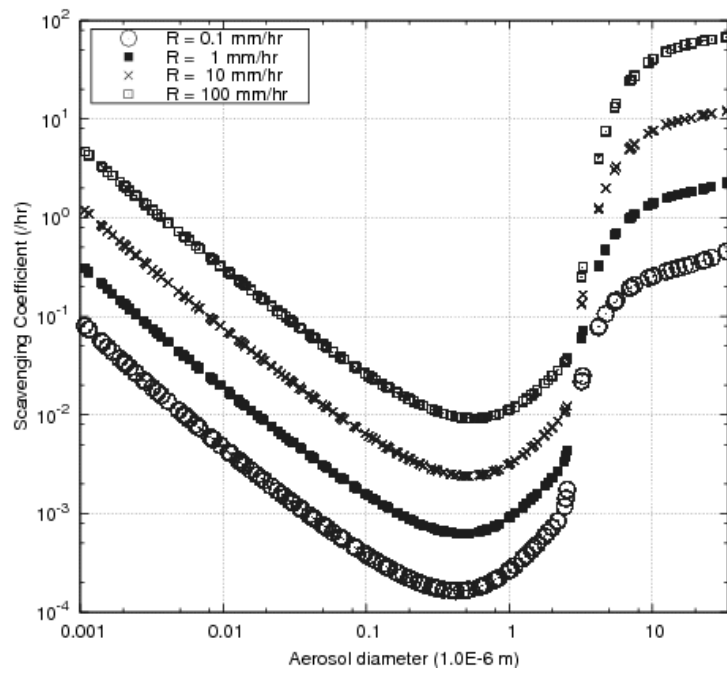


Figure 1: Scavenging coefficient as a function of particle size and rain rate (calculated with Eq.(4))

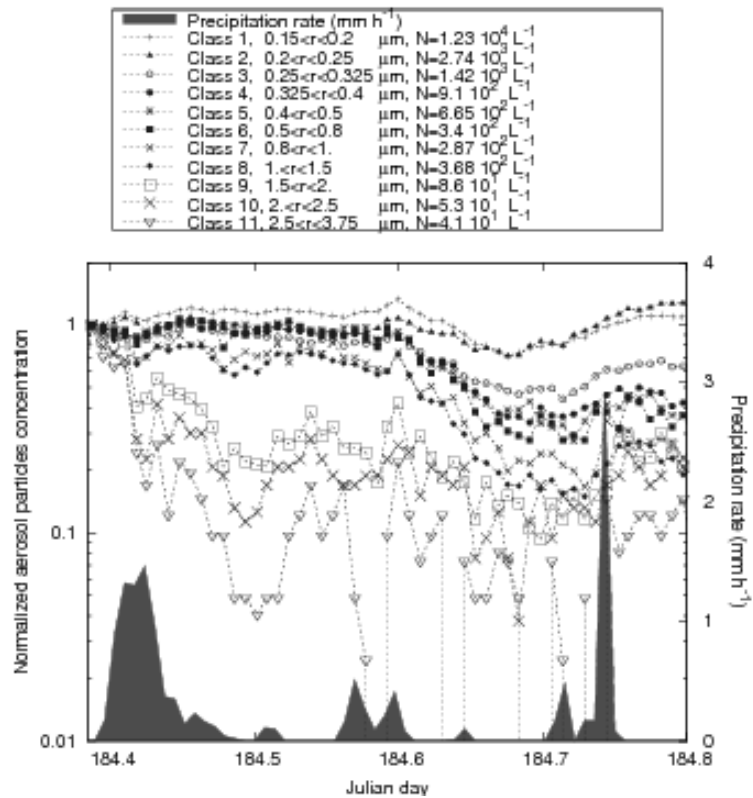


Figure 2: A short sequence of aerosol data and rainfall rate observed during COPS experiment

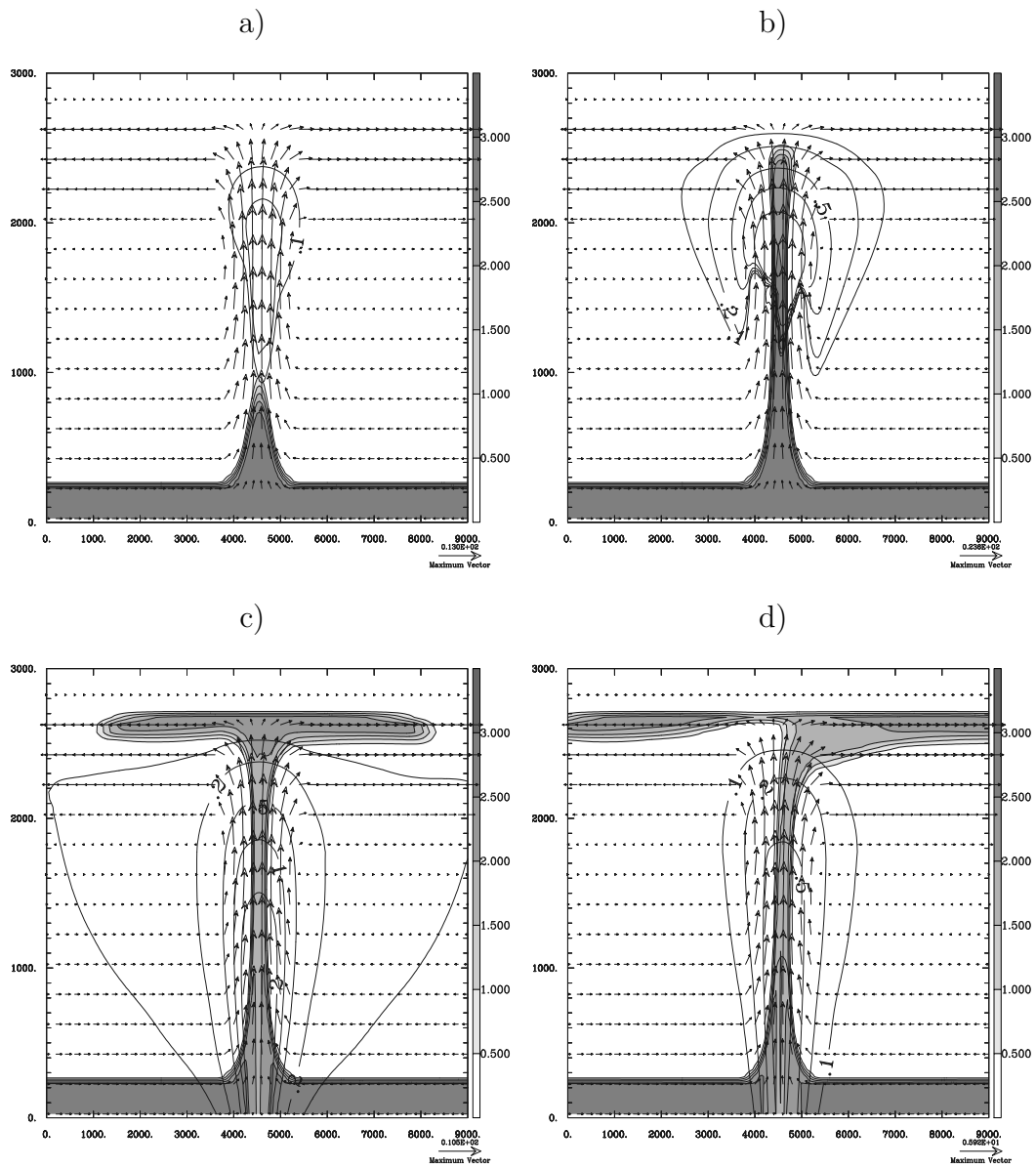


Figure 3: Concentration [cm^{-3}] of a log-normal particle mode with $N = 3 \text{ cm}^{-3}$, $\bar{d}_p = 2 \text{ }\mu\text{m}$ and $\sigma = 2$ after a) 20 min, b) 25 min, c) 35 min and d) 50 min of simulation of the "HaRP" case. Rain mixing ratio contours (for 0.1, 0.2, 0.5 and 1.0 g/kg) and flow structure (arrows) are superimposed.

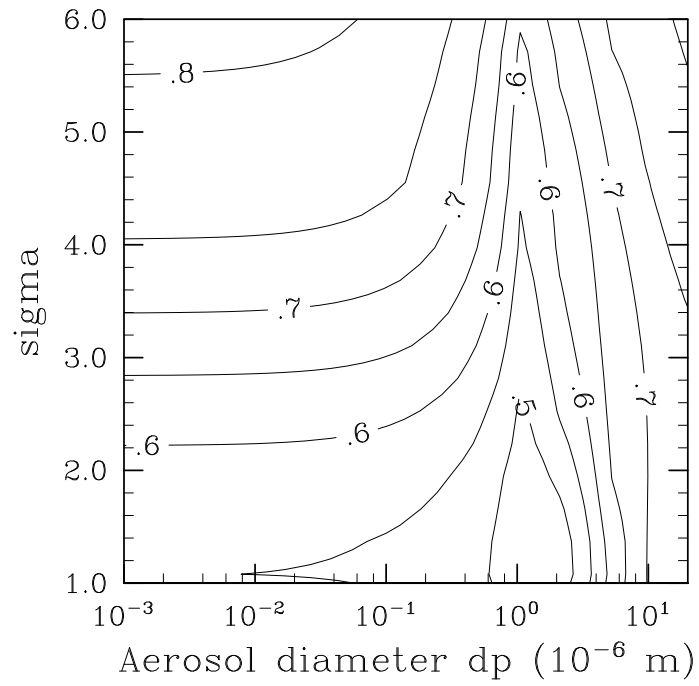


Figure 4: Fraction of scavenged AP as a function of the median diameter $\overline{d_p}$ and of the geometric standard deviation σ of the AP log-normal size distribution. The results obtained for a Marshall-Palmer distribution of the raindrops after 50 min of simulation of the "HaRP" case.

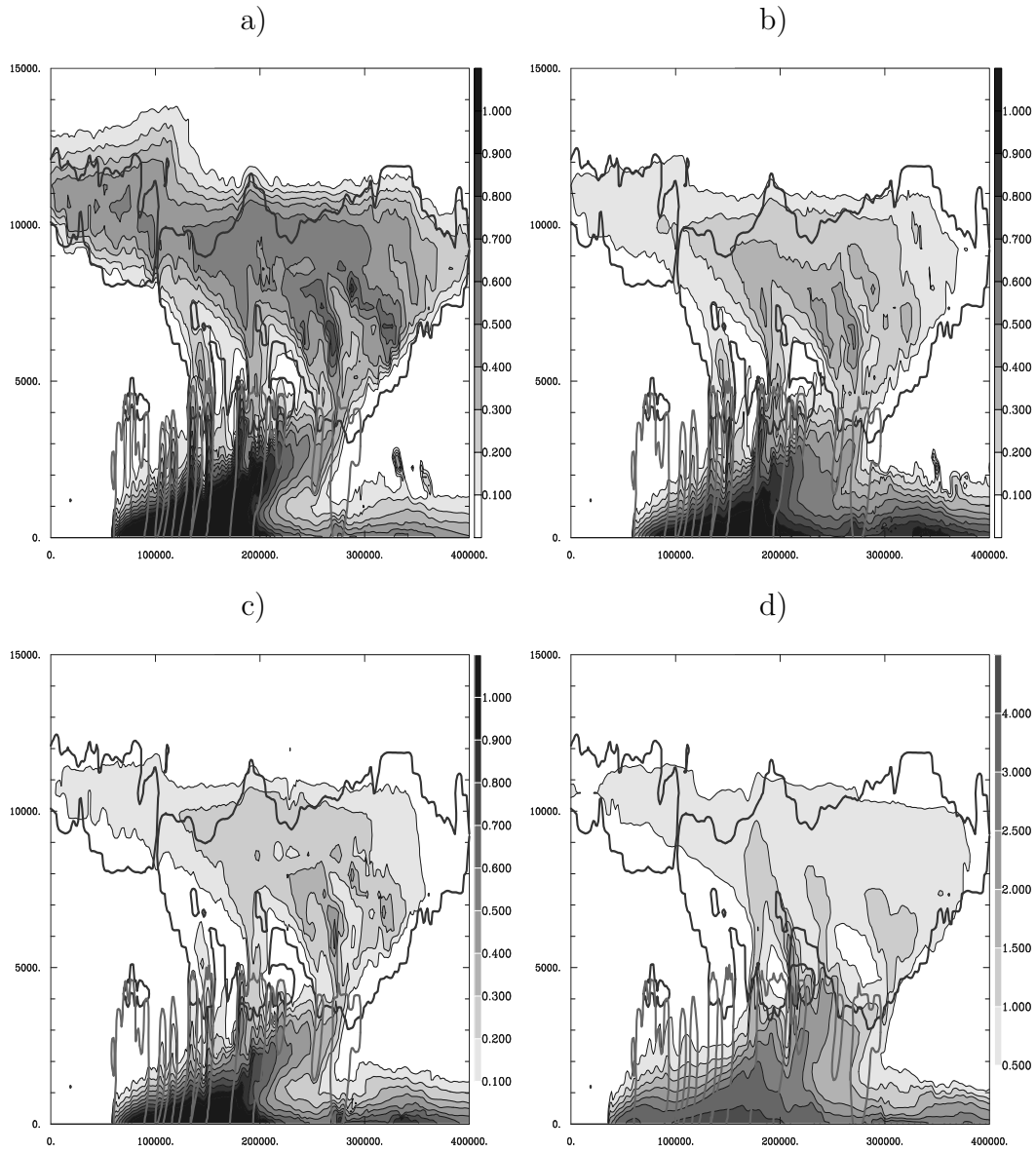


Figure 5: Concentration of the net scavenged coarse mode particles [cm^{-3}] deduced from the "COPT" simulation difference ($\text{SCAV}_0 - \text{SCAV}_1$) after 9 hours and for 3 initial dust layers: a) [0-2] km, b) [2-4] km and c) [4-6] km, and d) the residency time [hr]. The glaciated part of the squall line is depicted by a black solid line, the grey line shows the rainshaft contours for 10^{-5} kg/kg. The "x" and "z"-axes are labeled in meters.

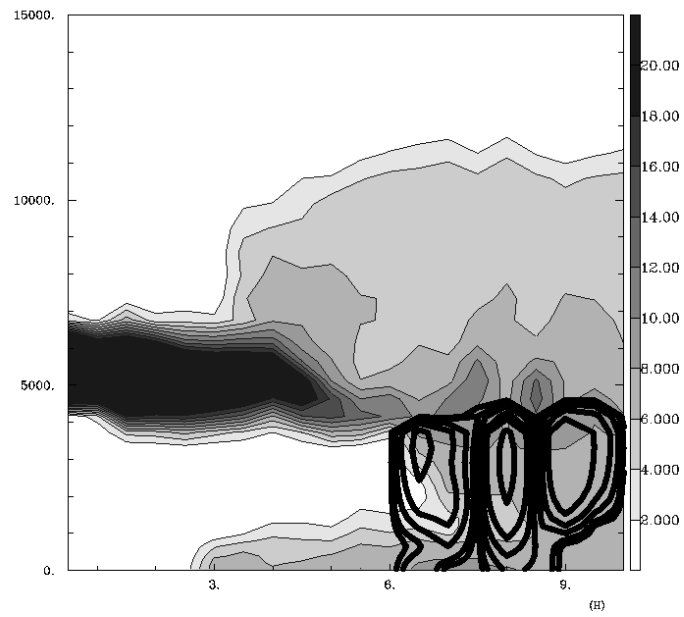


Figure 6: Time evolution of vertical profiles taken at $x=225$ km. The coarse mode particles [cm^{-3}] in the [4-6] km layer case are displayed with the rain mixing ratio with solid contours superimposed in log scale from 10^{-5} to 10^{-3} kg/kg.

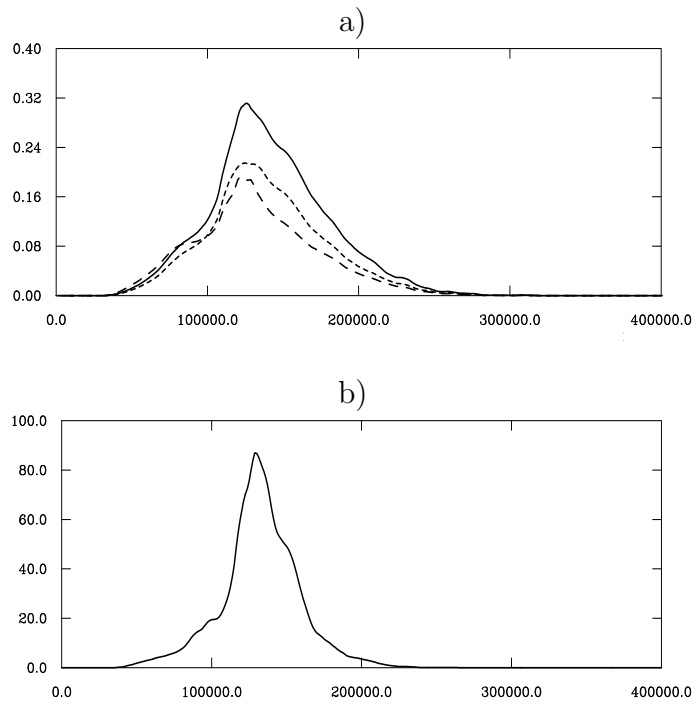


Figure 7: (a) Cumulated mass of scavenged AP by BCS [$\text{kg}\cdot\text{m}^{-2}$] for the [4-6] km layer (dotted line), the [2-4] km layer (solid line) and the [0-2] km layer (dashed line), (b) cumulated precipitation [$\text{mm}\cdot\text{hr}^{-1}$]. The curves are obtained at ground level and after 9 hours of the "COPT" simulation.