1. INTRODUCTION

Wet deposition is the most important sink of aerosol particles in the troposphere (Pruppacher and Klett, 1997). Wet deposition processes involve complex microphysical interactions between aerosol particles and hydrometeors. Two processes lead to wet deposition: the nucleation scavenging and the impaction scavenging. The nucleation scavenging is the scavenging of aerosol particles as cloud droplets or ice crystals are forming, respectively, on cloud condensation nuclei and on ice forming nuclei by heterogeneous nucleation. The impaction scavenging is the result of the interactions between aerosol particles and hydrometeors motions including: Brownian diffusion, interception, inertial impaction, thermophoresis, diffusionphoresis, airflow turbulence and electrostatic attraction. The impaction scavenging usually splits in two processes: in-cloud impaction scavenging, which treats the interactions between cloud droplets and raindrops with interstitial aerosol particles and below cloud scavenging, which concerns the collection of aerosol particles by falling raindrops below the cloud base. The relative importance between in-cloud scavenging processes (nucleation and impaction scavenging), also called washout, and below cloud scavenging, also called rainout, depends on meteorological conditions and on the properties of aerosol particles (size distribution and chemical composition) as well as on the stage of cloud development.

This study will focus on the below cloud scavenging (BCS) of aerosol particles although this work is embedded in a largest project, which aims at describing all processes leading to wet deposition of aerosol particles in a three-dimensional cloud resolving model. For BCS, due to the size of raindrops, the collection efficiency depends on Brownian diffusion, interception and inertial impaction (Pruppacher and Klett, 1997). A recent study (Andronache, 2003) shows that the BCS coefficient of aerosol particles increases with the rainfall rate. It has a high value for very small particles (diameters less than 0.01µm) and for the biggest ones (diameters larger than 2µm) whereas it is very low for particles in the intermediate range.

First a BCS module has been developed based upon the collision efficiency parameterization of Slinn (1983) widely used in atmospheric chemistry modeling (Tost et al., 2006). The module uses a generic Gauss quadrature method to integrate over the raindrops and aerosol particle size distributions. Then zero-dimensional tests of the module have been done on a set of rain rates and aerosol particle concentrations data of the COPS experiment, which took place in the Vosges-Black Forest area last July (see http://www.cops2007.de/). Finally, the module has been implemented in the three-dimensional mesoscale/cloud resolving model MesoNH (Lafore et al., 1998) and two preliminary simulations have been performed: an idealized case of a shallow tropical rainband in a 2D kinematic framework from the HaRP campaign (Cohard and Pinty, 2000) and a squall line case from the COPT81 experiment in West Africa (Lafore et al., 1989). For both simula-
tions, sensitivity tests are performed on the initial size distributions of aerosol particles as well as on their initial vertical profile.

2.DEVELOPMENT OF THE BCS MODULE

A BCS module has been developed for the tridimensional model MesoNH in order to describe the wet removal of aerosol particles by precipitation below the cloud base. The process of aerosol particles wet scavenging is related to the aerosol particles size distribution but also to the raindrop size distribution (Andronache, 2003). So, the major difficulty to represent the aerosol particles sink by BCS is due to the polydisperse nature of both distributions. However, for simplification, one assumes a monodisperse distribution for raindrops (with a typical raindrop diameter) in order to determine the BCS coefficient $\gamma(d_p)$ between aerosol particles and falling raindrop (Tost et al., 2006; Loosmore et al., 2004). Because in MesoNH some information on the raindrop size distributions is available, we choose to compute the BCS coefficients by the full integral over raindrop size, in order to get an accurate estimate (Andronache, 2003; Loosmore et al., 2004; Henzing et al., 2006; Tost et al., 2006; Feng, 2006). Then, the BCS coefficient is integrated over the aerosol particles size distribution, to obtain finally, the total size number BCS rate.

Our purpose is to implement the BCS module described below in the tridimensional cloud-resolving model MesoNH, wherein the aerosol particles are represented by the sum of several log-normal distributions $n_{pi}(d_p)$ depending on the number of modes:

$$\sum_{i=1}^{l} n_{pi}(d_p) = \sum_{i=1}^{l} \frac{N_i}{\sqrt{2\pi}d_p\sigma_i} e^{-\left(\frac{\log(d_p/d_{pi})}{\sqrt{2\log\sigma_i}}\right)^2}$$

where $l$ is the number of mode with index $i$, $d_p$ is the aerosol particle diameter, $N_i$, the number concentration, $\sigma_i$, the geometric standard deviation of the log-normal distribution, and $d_{pi}$ is the modal diameter. For the moment, we use a single-moment scheme for aerosol particles in MesoNH (only the number concentration $N$ is prognostic), so $\sigma_i$ and $d_{pi}$ are constants depending on aerosol particles types. Usually, three modes are used in order to represent the three classical modes: the Aitken mode, the accumulation mode and the coarse mode.

The raindrop size distribution $n_D(D_d)$ in MesoNH is modeled by a generalized Gamma distribution. It is reduced in this study, to the classical Marshall-Palmer law:

$$n_D(D_d) = N_0 \exp(-\lambda R D_d)$$

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

The method used to calculate the BCS coefficient follows the concept of the collision efficiency between an aerosol particle and a raindrop (Slinn, 1983; Pruppacher and Klett, 1997; Seinfeld and Pandis, 1998). The collision efficiency $E$ expresses the number of aerosol particles collected by collision with the falling raindrop, in the swept out volume of the raindrop. A value of $E = 1$ implies that all particles in the geometric volume swept out by a falling drop will be collected. All the recent studies on BCS use this concept (Andronache, 2003; Loosmore et al., 2004; Henzing et al., 2006; Tost et al., 2006; Feng, 2006).

While theoretical solution of the Navier-Stokes equation for the prediction of $E$ is not available, the accepted method uses dimensional analysis coupled with experimental data. By nondimensionalizing the equation of motion for air and for aerosol particles and raindrops, Slinn (1983) found that $E$ depends on five dimensionless parameters:

- $Re$ is the raindrop Reynolds number:
  $$Re = \frac{D_d U_t(D_d) \rho_a}{2 \mu_a}$$
  where $U_t$ is the terminal velocity of raindrop, $\rho_a$ is the air density and $\mu_a$ is the air viscosity.

- $Sc$ is the aerosol particle Schmidt number:
  $$Sc = \frac{\mu_a}{\rho_a \mathcal{D}}$$
  where $\mathcal{D}$ is the aerosol particle diffusivity.
St is the aerosol particle Stokes number:

\[ St = \frac{2U_t(D_d)\tau_a}{D_d} \]

where \( \tau_a \) is the characteristic relaxation time of the collected aerosol particle.

- \( \phi \) is the ratio of diameter:
  \[ \phi = \frac{d_p}{D_d} \]

- \( \omega \) is the viscosity ratio:
  \[ \omega = \frac{\mu_w}{\mu_a} \]

where \( \mu_w \) is the viscosity of water.

The particle diffusivity \( D \) is defined by:

\[ D = \frac{kTC_c}{3\pi \mu_a d_p} \]

where \( k \) is the Boltzmann constant, \( T \) is the temperature, and \( C_c \) is the Cunningham slip correction factor, which depends on the aerosol particle diameter and on the mean free path of air (Seinfeld and Pandis, 1998).

Then, Slinn (1983) propose the following analytical expression for the collision efficiency \( E \) based on correlation with experimental data:

\[ E(D_d, d_p) = \frac{4}{ReSc} \left[ 1 + 0.4Re^{1/2}Sc^{1/3} + 0.16Re^{1/2}Sc^{1/2} \right] + 4\phi \omega^{-1} + (1 + 2Re^{1/2})\phi \]

\[ + \left( \frac{St - St^*}{St - St^* + 0.667} \right)^{3/2} \left( \frac{\rho_p}{\rho_w} \right)^{1/2} \]

where \( St^* \) is the critical Stokes number expressed as:

\[ St^* = \frac{1.2 + (1/12ln(1 + Re))}{1 + ln(1 + Re)} \]

In our calculations, we assume that the aerosol has density \( \rho_p \) equal to 1 \( gm^{-3} \).

The analytical expression of the collision efficiency \( E \) has three terms with distinct physical contributions according to the aerosol particle diameter \( d_p \):

1. The first term in Eq.(1) illustrates the Brownian diffusion that dominates for aerosol particles with \( d_p < 0.01 \mu m \).

2. The second term is related to collection by interception process and concerns aerosol particles with diameter between 0.01 and \( 1 \mu m \). In this range, \( E \) has a minimum often referred to as the "Greenfield gap" (Seinfeld and Pandis, 1998).

3. The third term is due to the inertial impaction. It is efficient for large particles (\( d_p > 2 \mu m \)). This term is included only when \( St > St^* \).

Wet deposition is a 1st order decay process,

\[ \frac{\partial \psi(d_p, t)}{\partial t} = -\gamma(d_p)\psi(d_p, t) \]  \( (2) \)

where \( \psi(d_p, t) = n_p(d_p, t) \) if considering the conservation of aerosol particles number distribution \([particles/\mu m^{-1}m^{-3}]\) or \( \psi(d_p, t) = g(d_p, t) \) if considering the conservation of aerosol particles mass distribution \([\mu gm^{-1}m^{-3}]\), and \( \gamma(d_p) \) is the BCS coefficient of particles of diameter \( d_p \) already mentioned.

To compute the BCS coefficient \( \gamma(d_p) \), the collision efficiency \( E \) is integrated over all raindrop diameters \( D_d \):

\[ \gamma(d_p) = \int_0^\infty \frac{4\pi}{3}D_d^2 U_t(D_d)E(D_d, d_p)n_D(D_d)dD_d \]  \( (3) \)

To solve the integral of Eq.(3), a Gauss quadrature method is employed (Press et al., 1992) after a change of variable. Quadrature method consists in a discretization of the integral over a number \( n \) of optimized abscessas with an assigned weight (the sum of the weights is 1). A Gauss-Laguerre variant is used to integrate over the raindrop size distribution:

\[ \int_0^\infty x^\beta \exp(-x) f(x) dx \approx \sum_{i=1}^n w_i f(x_i) \]
where \( x = \lambda_R D_d \) and \( x_i \) is the discreted abscissa.

Finally, the BCS coefficient is integrated over all aerosol particle diameters \( d_p \) to obtain the total size number BCS rate \( \frac{\partial N}{\partial t} \):

\[
\frac{\partial N}{\partial t} = \frac{\partial}{\partial t} \int_0^\infty n_p(d_p) \, dd_p = \int_0^\infty \frac{\partial n_p(d_p)}{\partial t} \, dd_p = -\int_0^\infty \gamma(d_p) n_p(d_p) \, dd_p 
\]

(4)

For the integration over the aerosol particles diameters \( d_p \), a Gauss-Hermite quadrature formula is used:

\[
\int_0^\infty \exp(-x^2) f(x) \, dx \simeq \sum_{i=1}^n \omega_i f(x_i)
\]

where \( x = \frac{\log(d_p/d_0)}{\sqrt{2} \log \sigma} \) and \( x_i \), the abscissas. After several tests we choose to compute the sums with \( n = 20 \).

The BCS coefficients calculated with this method are consistent with those of Andronache (2003), Slinn (1983) and Feng (2007).

Figure 1 shows that BCS coefficient \( \gamma(d_p) \) varies significantly with the rain rate and the aerosol particle size. This figure illustrates the three distinct regimes of BCS corresponding to the three different physical processes involved in the collision efficiency \( E \) already described. So, coarse aerosol particles are the most efficiently scavenged, Aitken mode is moderately scavenged and for the accumulation mode, the scavenging is low.

The Figure 1 also demonstrates that, when the rain rate increases, the corresponding BCS coefficient increases. These results are in agreement with those of the previous studies (Andronache, 2003; Loosmore et al., 2004; Tost et al., 2006; Feng, 2006).

3. 0D APPLICATION ON COPS EXPERIMENT

COPS (Convective and Orographically-induced Precipitation Study) was a 3 month international field campaign with part of a program aiming to improve precipitation forecasts and headed by the German Research Foundation. COPS took place in Summer 2007 in south-western Germany and north-eastern France. One of the goals of COPS was to collect time series of high, spatial and temporal, resolution surface flux including as many aerosol, cloud, and precipitation variables as possible, to be used as lower boundary conditions for mesoscale models.

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

- Mean rainfall rate [mm/hr] calculated each 10 min from precipitation measurements with an Optical Rain Gauge (ORG)
- Time evolution of aerosol particle concentrations sampled by a Grimm optical particles counter with a time step of 1 min. The aerosol concentrations are provided for 15 classes of particles: size cut radius (\( \mu m \)) are [0.15; 0.2; 0.25; 0.325; 0.4; 0.5; 0.8; 1; 1.5; 2; 2.5; 3.75; 5; 7.5; 10; 20]
After comparing the two types of COPS data (precipitation intensity and aerosol particle concentrations), some sequences have been selected for which the aerosol depletion could be attributed to the wet removal by precipitation. The sequences have duration no longer than one day, and each of them contains several rainfall events. Figure 2 shows the COPS aerosol concentration data for Julian day 184.38 to 184.8 as well as the precipitation rate. This figure 2 attests the importance of scavenging on big aerosol particles.

For the moment, only the first precipitation event (between 184.38 and 184.5) is examined. During this rainfall event, aerosol concentrations of classes 1 to 8, are not perturbed. In contrast, aerosol concentrations of classes with larger diameters (classes 9 to 11) vary and are depleted in correlation with the rainfall event. This depletion seems to be partly explained by BCS process because, as shown on Fig. 1, the BCS rate is highest for large diameter and the first classes correspond to the "Greenfield gap". Thus, the data show the potential importance of the BCS process on the evolution of aerosol particles in the troposphere. Therefore, the implementation of this process in the aerosol continuity equation is necessary in the tridimensional model MesoNH. Looking at other rainfall events, for instance (beginning at 184.7), an increase of particle concentrations is observed for all the classes (even for the large ones) during the course of the event. Thus this event illustrates the complexity of the processes implied in the evolution of aerosol concentrations including dynamical transport, microphysical processes, sedimentation and scavenging.

In order to simulate the depletion of the aerosol particles by BCS using the COPS data of the first precipitation event, a mean particle diameter $\bar{d}_m$ has been defined for each class and the BCS coefficient $\gamma(\bar{d}_m)$ is calculated for each $\bar{d}_m$. The method consists to initialize the concentrations at the first time step $t_0$, with the measured values. Then the data of mean rainfall intensity are incorporated in the BCS module for a selected rainy episode. The temporal evolution of the particle concentration of each class of size $N_i(x,y,z,t)$ is only driven by the calculated BCS rates.

For each class, the percentage of aerosol particles concentrations depleted during the rainfall event is calculated by comparing aerosol particles concentrations at the beginning and at the end of the event. This is done for simulated and measured data. The resulting simulated percentage does not predict the entire depletion for each concentration class of COPS measurements, nevertheless, the influence of BCS is reproduced in function of diameter (as shown in Fig. 1). For instance, for the class number 3 and number 10 (defined in legend of Fig. 2), the simulated percentage of depletion is respectively 0.1% and 87% whereas measured one is respectively 10.8% and 54%. The discrepancy between the measured and simulated trends is attributed to other processes such as turbulent transport, microphysics, etc.

4. 2D SIMULATIONS WITH MesoNH

The numerical experiments are set to test
and to evaluate the impact of the BCS on a multimodal population of particles. The particles are transported by a moist flow in which a warm or mixed-phase microphysical cycle produces rainfalls along the course of the simulation.

4.1 Warm shallow convection: the "HaRP" test case

The "HaRP" test case aims at simulating a precipitating cell forced by a time-varying non-divergent circulation during 50 min. The numerical experiments are performed with the MesoNH model using a standard Kessler scheme for the microphysics and a highly performant PPM scheme for the transport of the scalar fields (thanks to T. Maric, now at Univ. of Washington, Seattle, WA). Several simulations are run with multiple marine aerosol modes (fine, accumulation and coarse) which characteristics are given in Table 1 of Andronache (2003) and not recalled here. Each mode is advected in the 2D vertical plane and locally scavenged by rain according to the size distribution parameters of the raindrops and aerosol particles. The computational domain extends over 180 × 60 gridpoints with a spacing of 50 m in x and z directions. The time scale is 5 sec.

At the initial state, several mixtures of particle modes, leaving to the "Marine_A, B, C" cases, are filling the lowest 250 m of the atmosphere. Figure 3 illustrates the scavenging efficiency of the "Marine_B" case after 35 min of simulation. Figure 3a refers to the coarser mode ($d_{p1} = 12\mu m$) which is completely scavenged in the updraft, Fig. 3b shows that the intermediate mode, here still a coarse mode ($d_{p2} = 2\mu m$), is partially scavenged. The finest mode, here with accumulation mode characteristics ($d_{p3} = 0.2\mu m$), remains nearly unaltered by the rainshaft (Fig. 3c). The concentrations of the particle modes (0.05, 3 and 70 cm$^{-3}$, respectively) are continuously resplishing through the lateral boundaries of the lowest 250 m.

Figure 3: Concentration of very coarse (top), coarse (middle) and accumulation (bottom) particle modes after 35 min for the "HaRP" case. Rain mixing ratio contours (log scale) and flow structure (arrows) are superimposed.
The contrasted behaviour of the "Marine A, B, C" cases shows up when considering the peak value of aerosol particle mass that is scavenged by the rain episode, after 50 min of simulation. For "Marine A, B, C", we get an instantaneous value of 1g, 2.5g and 0.01 mg.m$^{-3}$, respectively. The rainfall rate reaches 25 mm.h$^{-1}$ at the same time. The very poor scavenging efficiency of the "Marine C" case is explained by the low $d_p$ (0.033, 0.110 and 0.540 µm) and the narrow character ($\sigma$=1.40, 1.41 and 2.02) of the size distributions.

4.2 Tropical squall line: the "COPT" test case

The "COPT" test case is typically a 12 hour simulation of a tropical squall line with a scale-resolved internal circulation, a 2D turbulence scheme and mixed-phase microphysics. The model is initialized with 3 successive layers of aerosol of 2 km depth starting from the ground level. For each layer, the same multimodal population of particles corresponding to the "Dust Layer" case of Table 1 of Andronache (2003) is used. The domain contains $320 \times 44$ gridpoints unevenly spaced in the vertical ($\Delta z = 70$ m at ground level and $\Delta z = 700$ m above 12 km). The horizontal resolution is 1.25 km. The model is integrated with a time step of 10 sec. A gravity wave damping layer is inserted between 17 km and the model top at 22.5 km. A constant speed transformation is used to compensate for the motion of the squall line. No fluxes are considered in the surface layer. Convection is initiated by forming a -0.0.1 K.s$^{-1}$ artificial cold pool in the low levels of a small domain during 10 min.

A series of two simulations, without (SCAV$_0$) and with (SCAV$_1$) scavenging process applied to the particles, are performed. Figures 4a-c show the differences (SCAV$_0$-SCAV$_1$) relative to the coarse mode concentrations of each dust layer after 9 hours of simulation. The figures, ordered for the top, middle and bottom layers respectively, are showing the relative scavenging intensity of each layer, but plotted at

Figure 4: Concentration of scavenged coarse mode particles deduced from "COPT" simulations SCAV$_0$ and SCAV$_1$ and for 3 initial dust layers: [6,4] km (top), [4,2] km (middle) and [2, 0] km (bottom). The glaciated part of the squall line is depicted by a black solid line, the grey line shows the rainshaft contours.
the same scale. The initial concentration of the coarse mode \((d_{p1} = 0.55 \mu m)\) is \(20 \, \text{cm}^{-3}\). The results of Figs 4a-c indicate that all the dust layers are affected by the scavenging process which is efficient enough to eliminate much more than \(1 \, \text{cm}^{-3}\) of coarse mode (here submicron) particles in the heavy precipitating convective region of the squall line. In other words, the peak value of the scavenged particle mass, obtained by summing the particle mass of the 3 modes and keeping \(p_v = 1 \, \text{g cm}^{-3}\), is as high as \(5 \, \text{mg per m}^{-3}\) of air.

Another interesting feature displayed in Figs 4a-c is the fate of the scavenged particles in the upper glaciated region of the squall line, that is, well above 4 km high. As expected the bottom dust layer is the most affected by the scavenging removal but the 4-6 km layer is also affected because raindrops originate from altitude up to 4.5 km that corresponds to the ice melting layer.

The COPT case needs much investigation to analyse all the aspects of particle scavenging by rain in such an organized convective system. Next step will concentrate on the budget of the total mass of the particles which includes transport, scavenging and sedimentation.

5. CONCLUSIONS

A BCS module has been studied for implementation in the mesoscale/cloud resolving model MesoNH. The module is based on the collection efficiencies of Slinn (1983) and contains explicit numerical integrations over the raindrop and particle size distributions. The first numerical experiments performed with the module show that the coarse mode \((d_{p1} > 1 \mu m, \text{approximately})\) of a particle population can be drastically scavenged according to the rainfall intensity and to the particle distribution modal diameters. This effect is expected to remove giant condensation nuclei and ice forming nuclei by precipitation with possible consequences on the nucleating properties of drizzling marine stratus and convective clouds with an ice phase. However more tests are needed as for instance, to examine the sensitivity to the width of the particle size distributions and the initial vertical profile of the particles with respect to the condensation and to the freezing level.

6. REFERENCES


