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Shepard and Hardy Multiquadric Interpolation Methods for Multicomponent Aerosol–Cloud Parameterization

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ABSTRACT

This paper presents a novel method based on the application of interpolation techniques to the multicomponent aerosol–cloud parameterization for global climate modeling. Quantifying the aerosol indirect effect still remains a difficult task, and thus developing parameterizations for general circulation models (GCMs) of the microphysics of clouds and their interactions with aerosols is a major challenge for climate modelers. Three aerosol species are considered in this paper—namely sulfate, sea salt, and biomass smoke—and a detailed microphysical chemical parcel model is used to obtain a dataset of points relating the cloud droplet number concentration (CDNC) to the three aerosol input masses. The resulting variation of CDNC with the aerosol mass has some nonlinear features that require a complex but efficient parameterization to be easily incorporated into GCMs. In bicomponent systems, simple interpolation techniques may be sufficient to relate the CDNC to the aerosol mass, but with increasing components, simple methods fail. The parameterization technique proposed in this study employs either the modified Shepard interpolation method or the Hardy multiquadrics interpolation method, and the numerical results obtained show that both methods provide realistic results for a wide range of aerosol mass loadings. This is the first application of these two interpolation techniques to aerosol–cloud interaction studies.

1. Introduction

Atmospheric aerosols are small suspensions of fine solid or liquid particles in the atmosphere, ranging in size from a few nanometers to tens of micrometers. They have an important role in the earth's radiative budget via their direct effect (as they scatter and absorb solar and infrared radiation in the atmosphere) and their indirect effect (as they alter the formation and precipitation efficiency of the clouds; Lohmann and Feichter 2005). The quantification of the aerosol radiative forcing is a major and complex challenge for climate modelers (see, e.g., Houghton et al. 2001 and Forster et al. 2007). This paper is intended to provide the research community with a novel technique aimed at improving the modeling of the aerosol activation process and, thus, of the aerosol indirect effect.

Modeling the simultaneous activation and growth of aerosol species under typical stratiform clouds conditions is very important for a better understanding of the aerosol indirect effect. As noted in Lohmann and Feichter (2005) and McFiggans et al. (2006), establishing the link between aerosol and cloud droplets is probably still the weakest point in the attempt to estimate the aerosol indirect effect. At the moment there are several existing schemes (using several different approaches) for the parameterization of cloud droplet formation. Some schemes (e.g., Jones et al. 1994, hereafter JRS94; Boucher and Lohmann 1995, hereafter BL95; Menon et al. 2002, hereafter MD02) use empirical relationships between aerosol mass or number concentration and cloud droplet number concentration (CDNC). One of the limitations of these schemes is the scarcity of observational data; another is the fact that most of the derived relationships relate only sulfate and sea salt to CDNC. Other parameterizations use lognormal representations of aerosol size distributions and Köhler theory to relate the aerosol size and composition to the number activated as a function of maximum supersaturation (see Abdul-Razzak and Ghan 2000). In Chuang and Penner (1995), the CDNC is parameterized in terms of local aerosol number, anthropogenic sulfate

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number concentration, and updraft velocity, whereas in Glantz and Noone (2000, hereafter GN00) a physically based conversion algorithm for estimating the relationship between aerosol mass and cloud droplet number was proposed. More recently, Nenes and Seinfeld (2003) and Fountoukis and Nenes (2005) have developed parameterizations based on a generalized representation of aerosol size and composition within the framework of an ascending adiabatic parcel, deriving a largely analytical solution to the equations for droplet activation.

Most authors acknowledge today that sulfate aerosols have a significantly reduced role in cloud droplet nucleation compared to previous estimates and that competition between different aerosol species, such as sulfate and sea salt, as cloud condensation nuclei has to be taken into account (see O'Dowd et al. 1999a). In Ghosh et al. (2007), a third component—biomass smoke aerosol—was included in addition to sulfate and sea salt in a detailed microphysical parcel model, and it was concluded that this third component can significantly perturb the activation and growth of both sulfate and sea-salt particles, leading to a much more complex relationship between these aerosol components and the number of cloud droplets activated.

A key issue that must be addressed to improve the modeling of the aerosol activation and cloud droplets formation processes is therefore the competition between the different aerosol species. This study focuses on this issue and proposes a novel numerical technique for the modeling of the complex nonlinear response of the CDNC in a three component aerosol case, in which the three components are sulfate, sea salt, and biomass smoke. The numerical technique is based on interpolating a series of points generated by the detailed microphysical parcel model described in Ghosh et al. (2007). Two such interpolation techniques are presented—namely, the Shepard interpolation and the Hardy multiquadrics methods—both of which provide a global three-component parameterization, which can be adapted to the study of climatic responses in multicomponent aerosol domains.

2. The model

For modeling the nucleation of the aerosol particles into cloud droplets, we use a Lagrangian parcel model with explicit microphysics and fully interactive chemistry based on the one described in O'Dowd et al. (1999b) and Ghosh et al. (2005). This model was adapted and developed to obtain a series of data points that relate the CDNC to the three aerosol masses considered, namely, sulfate, sea salt, and biomass smoke. The microphysical aspect of the model uses the dynamic growth equation (see Pruppacher and Klett 1997) for the growth of aerosol solution droplets by condensation of water vapor on a size-resolved droplet spectrum. The growth law includes curvature and solution effects and is corrected for the breakdown of the continuum approximation close to the droplet surface. The model also includes mass transport limitations based on Schwartz (1986) and treats the nonideal behavior of solution droplets based on the Pitzer calculations (see Pitzer 1991).

The initial model from O'Dowd et al. (1999b) and Ghosh et al. (2005) was modified to include biomass smoke along with sulfate and sea-salt aerosols. For biomass smoke particles to activate into cloud droplets, it is essential that these particles have some water-soluble components. Although sulfates, water-soluble salts, and inorganic acids are known to be very efficient cloud condensation nuclei (CCN), the ability of biomass smoke particles to act as CCN is currently relatively poorly understood. Several studies (e.g., Van Dinh et al. 1994; Novakov and Corrigan 1996) have suggested that although aerosols from biomass burning are primarily composed of organics, some of their CCN activity may actually be due to coresident inorganic constituents. Accounting for all chemical species in biomass smoke particles in a numerical model may be an intractable problem. However, because we are mainly concerned with the physico-chemical properties that significantly affect the activation of these particles into cloud droplets, we assume that inorganic sulfate comprises the major contributor to the amount of soluble material in biomass smoke particles. Therefore, as in Ghosh et al. (2007), the biomass smoke is assumed to be internally mixed with sulfate particles. The amount of soluble material to be expected within the biomass burning particles can be estimated from observational studies of these particles (e.g., Yamasoe et al. 2000).

A series of model runs provides a set of data points relating the CDNC to a wide range of subcloud aerosol mass loadings. For example, if N such runs are performed, then the following dataset is obtained: (x_i, y_i, z_i, f_i), $i = 1, \ldots, N$, where x, y, and z represent the sulfate, sea-salt, and biomass smoke aerosol mass loadings, respectively, and f values are the corresponding CDNC values. The aim of the techniques presented in this paper is to obtain a smooth function F, F: $\mathbb{R}^3 \rightarrow \mathbb{R}$, such that $F(x_i, y_i, z_i) = f_i$, for $i = 1, \ldots, N$.

For this parameterization to be incorporated into GCMs, two very important requirements have to be satisfied: (i) it should have a minimized computational cost and (ii) it should involve an exact fit of the data and a "no nonsense" progression of the surface from a data point to other data points in the vicinity (see Hardy 1990). As explained in Hardy (1990), for more or less obvious reasons, trigonometric series and polyno-

mials cannot satisfy the second requirement for sparse, scattered data, and therefore more complex schemes have to be considered.

The distribution of aerosol mass concentration over the globe is very diverse and, thus, aerosol–cloud parameterizations have to be able to deal with different extreme scenarios. Examples include sea salt from storm surges, sulfates from volcanic eruptions, or biomass smoke from forest fires. Because of the diverse mass ranges encountered, robust interpolation methods have to be applied to provide a global parameterization, and the two such methods that are proposed in this paper are described in the following sections.

3. Shepard interpolation

The main idea of this technique was introduced by Donald Shepard in 1968 for irregularly spaced data in two dimensions (see Shepard 1968) and consists in defining the function F based on a weighted average of the values at all data points, where the weighting is a distance function; that is,

$$F(x, y, z) = \begin{cases} \dfrac{\sum_{i=1}^{N} d_i^{-\mu} f_i}{\sum_{j=1}^{N} d_j^{-\mu}}, & \text{if} \quad (x, y, z) \neq (x_i, y_i, z_i) \quad \text{for all} \quad i = 1, \ldots, N, \quad \text{and} \\ f_i, & \text{if} \quad (x, y, z) = (x_i, y_i, z_i) \quad \text{for some} \quad i = 1, \ldots, N, \end{cases} \tag{1}$$

where $d_i = d_i(x, y, z) = \sqrt{(x - x_i)^2 + (y - y_i)^2 + (z - z_i)^2}$ is the Euclidian distance and $\mu > 0$ (typically $\mu = 2$, but other values may also be used). In our specific three-component study, these weighting functions ensure that the CDNC value at a new point in the three-dimensional space defined by the three aerosol mass concentrations is more influenced by the CDNC values at the points which are closer to this new point, as opposed to other more remote points.

From a practical point of view, the above pure inverse-distance weighting formulation has two main shortcomings, as shown in several studies (e.g., Shepard 1968; Gordon and Wixom 1978; Franke and Nielson 1980; Renka 1988; Nielson 1993). These shortcomings are (i) the fact that the method is global and therefore computationally inefficient and (ii) the fact that a flat spot occurs at each data point as a result of the zero derivatives at these points. This flat spot feature means that in the vicinity of each data point, all the new CDNC values are too much influenced by that data point and this leads to a nonrealistic progression of the CDNC values between two data points.

Thus, to overcome these drawbacks, various modifications have been proposed in the literature. The most widely accepted version is the one introduced in Franke and Nielson (1980), which is called the modified Shepard method. According to this method, the function F is defined as follows:

$$F(x, y, z) = \frac{\sum_{i=1}^{N} w_i(x, y, z) q_i}{\sum_{j=1}^{m} w_j(x, y, z)}, \tag{2}$$

where

$$w_i(x, y, z) = \left[\frac{(R_w - d_i)_+}{R_w d_i} \right]^2, \tag{3}$$

in which R_w is a radius of influence about the node (x_i, y_i, z_i), which means that the data at (x_i, y_i, z_i) only influences interpolated values at points within the radius of influence R_w, and

$$(R_w - d_i)_+ = \begin{cases} R_w - d_i, & \text{if} \quad d_i < R_w, \quad \text{and} \\ 0, & \text{if} \quad d_i \geq R_w. \end{cases} \tag{4}$$

The radii of influence R_w take different values for every corresponding node (x_i, y_i, z_i), being chosen to be just large enough to include N_w data points, where N_w is a positive integer. The introduction of these radii of influence ensures that the interpolation becomes significantly less expensive from a computational point of view.

The other key modification of the pure inverse-distance weighting formulation is the introduction of the nodal functions q_i, which are local approximations to f_i at (x_i, y_i, z_i) defined as follows:

$$q_i(x, y, z) = f_i + \bar{a}_{i2}(x - x_i) + \bar{a}_{i3}(y - y_i) + \bar{a}_{i4}(x - x_i)^2 + \bar{a}_{i5}(x - x_i)(y - y_i) + \bar{a}_{i6}(y - y_i)^2, \tag{5}$$

where the coefficients $\bar{a}_{ij}$, $j = 2, \ldots, 6$ minimize the following expression:

$$\sum_{k=1, k\neq i}^{N} \omega_k(x_i, y_i, z_i)[f_i + a_{i2}(x_k - x_i) + a_{i3}(y_k - y_i) + a_{i4}(x_k - x_i)^2 + a_{i5}(x_k - x_i)(y_k - y_i) + a_{i6}(y_k - y_i)^2 - f_k]^2, \tag{6}$$

with

$$\omega_k(x, y, z) = \left[\frac{(R_q - d_k)_+}{R_q d_k}\right]^2, \tag{7}$$

where R_q is a radius of influence about the node (x_i, y_i, z_i). It is observed that q_i is a bivariate quadratic function that satisfies $q_i(x_i, y_i, z_i) = f_i$ and fits the values of f on a set of nearby nodes in a weighted least squares sense within a radius R_q of the point (x_i, y_i, z_i). In a similar manner as with R_w, R_q is chosen to be just large enough to include N_q data points, where N_q is a positive integer. The introduction of these nodal functions eliminates the nonrealistic flat spot feature of the interpolating function F that is observed when a pure inverse distance weighting formulation is adopted.

Note that to eliminate the sensitivity of the interpolation with scaling, we have mapped the whole data onto a unit cube; that is, $(x_i, y_i, z_i) \in [0, 1] \times [0, 1] \times [0, 1]$, for all $i = 1, \ldots, N$.

The numerical results illustrated in this paper employ the modified Shepard method described above, as implemented in the Numerical Algorithms Group (NAG) FORTRAN library subroutine E01TGF (see Numerical Algorithms Group 2006), which was derived from the routine QSHEP3 described in Renka (1988). It should be mentioned that applications of Shepard-based interpolation methods can be found in the literature in various applications [e.g., computer science (scientific visualization), chemical physics, computational fluid dynamics etc.] but until now the method has not been applied to aerosol–cloud parameterizations.

4. Hardy multiquadric interpolation

The second technique proposed in this paper is the multiquadric (MQ) method for interpolating scattered data, which was discovered in 1968 by Rolland Hardy and was published in Hardy (1971). Since then, the method has been adopted as an efficient tool by scientists from various areas because of its accuracy and simplicity. A good review of the method and its applications presented in an evolutionary, chronological order can be found in Hardy (1990).

The main idea of the Hardy MQ interpolation method is based on constructing the interpolation function F by superpositioning some quadric surfaces. Thus, using a particular example of a multiquadric surface, the function F is defined as follows:

$$F(x, y, z) = \sum_{i=1}^{N} c_i[(x - x_i)^2 + (y - y_i)^2 + (z - z_i)^2 + \Delta^2]^{1/2}, \tag{8}$$

where c_i, $i = 1, \ldots N$ are unknown coefficients and Δ^2 is a nonzero input shape parameter.

The coefficients c_i are calculated by solving the $N \times N$ system of equations given by the conditions

$$F(x_i, y_i, z_i) = f_i, \tag{9}$$

for $i = 1, \ldots N$. It was proved in Micchelli (1986) that for distinct data, this system of equations (and thus the MQ interpolation) is always solvable. A standard direct Gaussian elimination method is employed in this paper to find the solution of this system of equations.

The choice of the input shape parameter Δ^2 changes the sharpness of the quadric surfaces; that is, a small Δ^2 generates "sharp nosed" quadric surfaces, whereas a large Δ^2 generates "broad nosed" quadric surfaces (see Hardy 1990). It should be mentioned that finding a method for computing the optimal value for Δ^2 is still an open theoretical problem. In Carlson and Foley (1991) it has been shown that the optimal value depends primarily on the function values f_i and is almost independent of the data points (x_i, y_i, z_i). In the same paper, an empirical relation for computing this optimal value was also developed.

Another approach proposed by some authors consists in allowing the input shape parameter Δ^2 to vary with the basis function number, generating in this way a diverse collection of differently shaped quadric surfaces. In Kansa (1990) and Kansa and Carlson (1992), it was reported that this approach leads to a significant improvement in the accuracy of the method. Kansa (1990) also mentioned that an essential factor in obtaining accurate results is the conditioning of the matrix of coefficients c_i, with significantly better results being obtained for a low condition number (i.e., a well-conditioned matrix). Therefore, Kansa notes that only a monotonic variation of the input parameter Δ^2 should be permitted, and he suggests that exponential variations, as opposed to linear variations, produce some better conditioned coefficient matrices.

The disadvantage of the variable (Δ^2) multiquadric (VMQ) as opposed to the constant multiquadric (CMQ) is that in the VMQ case the coefficient matrix is no longer symmetric and does not yield a biharmonic interpolant. However, for the aerosol–cloud parameterization proposed in this paper, we found that this drawback of the VMQ is a small price to pay for the benefits of the vastly improved accuracy, as noted in Kansa and Carlson (1992). The VMQ strategy was therefore adopted, with the function F defined as follows:

$$F(x, y, z) = \sum_{i=1}^{N} c_i[(x - x_i)^2 + (y - y_i)^2 + (z - z_i)^2 + \Delta_i^2]^{1/2}, \tag{10}$$

with

$$\Delta_i^2 = \Delta_{\min}^2 \left(\frac{\Delta_{\max}^2}{\Delta_{\min}^2} \right)^{(i-1)/(N-1)}, \quad (11)$$

where $i = 1, \ldots N$, and $\Delta_{\min}$ and $\Delta_{\max}$ are two fixed nonzero input parameters.

As with the Shepard interpolation, all data were mapped to a unit cube to eliminate the scaling sensitivity of the interpolation.

Finally, we mention that, although the Hardy MQ method has not yet been employed for aerosol–cloud parameterizations, MQ-based methods have been successfully applied to various problems in geodesy, geophysics, remote sensing, signal processing, geography, hydrology, etc. More recently, MQs were also employed in the development of mesh-free methods for the numerical solution of various partial differential equations.

5. Implementation into GCMs

The incorporation of this new scheme for the parameterization of aerosol–cloud interactions into GCMs is based on two steps. The first step involves the choice of the aerosol mass concentration ranges and of the initial conditions for which the parcel model is run to obtain the points that define the function relating CDNC to aerosol mass concentrations. With respect to the actual GCM runs, these parcel model runs are done offline.

The second step involves the interpolation of the points from the previous step to predict new values of the CDNC in terms of some given aerosol mass concentrations. This step is done online within the GCM every time a CDNC calculation is needed.

Ideally, because the first step is done offline, it is desirable to perform a large number of parcel model runs to produce a large set (x_i, y_i, z_i, f_i), $i = 1, \ldots, N$. This would mean that the process of particle activation in the aerosol–cloud domain is represented with a high resolution (i.e., the larger the values of N, the higher the resolution). However, the complexity, and therefore the computational efficiency, of the second step is strongly related with the size of this set (i.e., a higher computational cost for a larger N). Therefore, the performance of this new parameterization depends on the choice of N, which should be large enough to account for a good representation of the aerosol–cloud domain but small enough to allow a computationally efficient online interpolation.

The methodology presented in this paper is directly applicable to the Hadley Centre Global Environmental Model version 2 (HadGEM2), being an improvement of its current aerosol–cloud scheme. However, it is expected that similar methodologies could be applied to improve the parameterizations of the aerosol–cloud interaction processes in other GCMs.

TABLE 1. The spectral parameters for aerosol lognormal input distributions.

	Median radius (nm)	Standard deviation	Density (kg m^{-3})
Sulfate	95	1.4	1769
Biomass smoke	120	1.3	1350
Sea salt (film mode)	100	1.9	2165
Sea salt (jet mode)	1000	2.0	2165

6. Numerical results

This section focuses on evaluating the results generated by the new parameterization. We first compare the noninterpolated model results with the existing scheme currently used in HadGEM2. Then we evaluate the performance of the two interpolation methods described in the previous two sections, and finally we compare the CDNC generated by this new parameterization with CDNC values generated by some existing schemes.

A set of N data points (x_i, y_i, z_i, f_i) is generated by N separate runs of the detailed microphysical parcel model briefly described in section 2. The model inputs allow for the specific choice of several parameters describing the channels of the aerosol species considered, the initial dynamic conditions, and the concentrations of the various gas-phase species modeled. The three dry input aerosol species were considered to appear in four modes, namely ammonium sulfate particles, aged biomass smoke particles internally mixed with the sulfate particles, sea-salt film mode particles, and sea-salt jet mode particles. The ammonium to sulfate molar ratio was inferred from volatility analysis to be 0.8. The channels for the four aerosol modes were chosen according to the lognormal distributions defined by the spectral parameters currently used by HadGEM2 (see Table 1). It should also be mentioned that the sea-salt split between its two modes (i.e., the film and the jet modes) was calculated from the parameterization functions (based on observational data) relating the number concentrations of the two modes to wind speed (see O'Dowd et al. 1997, 1999a).

The model was initialized with the following dynamical input parameters: updraft velocity 0.2 m s^{-1}, relative humidity 98.9%, temperature 10.24°C, and pressure 879.8 mb. Also, the input trace gas concentrations were as follows: CO_2 350 ppm, NH_3 0.3 ppb, O_3 30 ppb, H_2O_2 1.0 ppb, SO_2 5.0 ppt, HCl 10^{-5} ppt.

The model generates the predicted hydrated aerosol distribution after undergoing physical and chemical processing in the cloud. Of these hydrated aerosol particles, the ones that have grown to a radius larger than 1 μm are counted as cloud droplets. In this way, after one model run, one data point relating a CDNC to a certain combination of sulfate, sea-salt, and biomass smoke mass concentration is obtained. In mathematical terms, we can say that the ith model run generates a set $\{x_i, y_i, z_i, f_i\}$, for which $F(x_i, y_i, z_i) = f_i$.

Ten values were considered for each of the three aerosol masses within the following ranges: $x \in [0.001, 75]$, $y \in [0.001, 275]$, and $z \in [0.001, 100]$, corresponding to the sulfate, sea-salt, and biomass smoke aerosol mass concentrations (in μg m^{-3}), respectively. The limits of these ranges were chosen to include both clean and very polluted conditions. However, the distribution chosen for the ten values within the interval considered in each dimension is nonuniform, with most points taken in the aerosol mass concentration ranges that are most common. Therefore, most of the values (i.e., 7 from the total of 10) are within the following ranges: $x \in [0.001, 1.5]$, $y \in [0.001, 155]$, and $z \in [0.001, 2.3]$.

A dataset of $N = 10^3 = 1000$ points, (x_i, y_i, z_i), for $i = 1, \ldots, 1000$ was therefore generated and, after the corresponding 1000 model runs were performed, the input dataset to be interpolated, (x_i, y_i, z_i, f_i), for $i = 1, \ldots, 1000$, was obtained. Each model run employed a number of 10 aerosol sea-salt bins (5 film mode and 5 jet mode) and 10 aerosol non-sea-salt bins (5 sulfate and 5 biomass smoke).

The CDNC values generated by the model can be compared with CDNC values generated by an existing parameterization. Figure 1 illustrates the difference between our model results and the JRS94 parameterization, currently employed in HadGEM2, in cases when the sulfate, sea salt, and biomass smoke masses, respectively, are fixed to three different loadings. In both cases, for the aerosol loading range considered, the predicted maximum CDNC values are approximately 235 cm^{-3}. The minimum values are 0.1 and 5 cm^{-3} for the model results and the JRS94 parameterization, respectively, but we should mention that the latter specifically enforces 5 cm^{-3} as its minimum value. Overall, it is noted that there is a reasonable agreement between the two sets of CDNC predicted values. However, in some regions it is observed that the JRS94 parameterization underpredicts the CDNC by approximately 15%. Also, the fact that the difference between the model results and the JRS94 parameterization is not constant throughout the domain implies that the model suggests the existence of some nonlinear features of the activation process, which is consistent with the observations from Ghosh et al. (2007).

a. Interpolation results

Both the modified Shepard interpolation and the Hardy multiquadric methods are employed to generate a set of 125 000 (50^3) new values for CDNC, corresponding to 50 different values for each of the three aerosol masses uniformly distributed within the ranges specified above, based on the 1000 CDNC values generated by the model runs.

For the modified Shepard interpolation method based on the function defined by Eq. (2) we employed the NAG subroutine E01TGF with its default parameters for N_w and N_q, namely $N_w = 32$ and $N_q = 17$. For the Hardy multiquadric, the VMQ approach as defined by Eq. (10) was adopted, with $\Delta^2_{\min} = 10^{-10}$ and $\Delta^2_{\max} = 10^{-2}$.

Figure 2 illustrates the performance of the two interpolation methods. The top three panels show the predicted CDNC values as generated by the model runs (before interpolation). The middle three and the bottom three panels show the predicted CDNC values after the application of the modified Shepard and Hardy VMQ interpolations, respectively. To allow the visualization of the results, the CDNC values are illustrated when F is a function of only two variables, with the third one being set at some specific values. One such value is considered for each of the three components, obtaining in this way three different cases: $F(1.5, y, z)$ for a fixed value of x (fixed sulfate), $F(x, 154.5, z)$ (fixed sea salt) and $F(x, y, 2.3)$ (fixed biomass smoke). It can be observed that the two interpolation methods provide very similar results and, at the same time, fulfill the main requirements for an appropriate interpolation method. We should also note that whereas at low aerosol loadings the CDNC increases in a fairly linear manner, at high aerosol loadings the cloud droplet activation process presents some nonlinearities that are very well reproduced by both interpolation methods.

Computational cost is an important consideration in choosing one technique over another, especially in the present case in which both interpolation methods proposed produce very similar results. The application of the parameterization into GCMs requires a call of the interpolation routine for each set of aerosol mass concentrations considered at a certain time. This means a GCM run requires a significant number of interpolations. Our investigations, based on 1000 interpolations of 2500 new points using an initial 125-point dataset, showed that on a 3.0-GHz Intel Xeon CPU with 1 GB of RAM the Hardy multiquadric method required approximately 5 s to run, whereas the computational cost

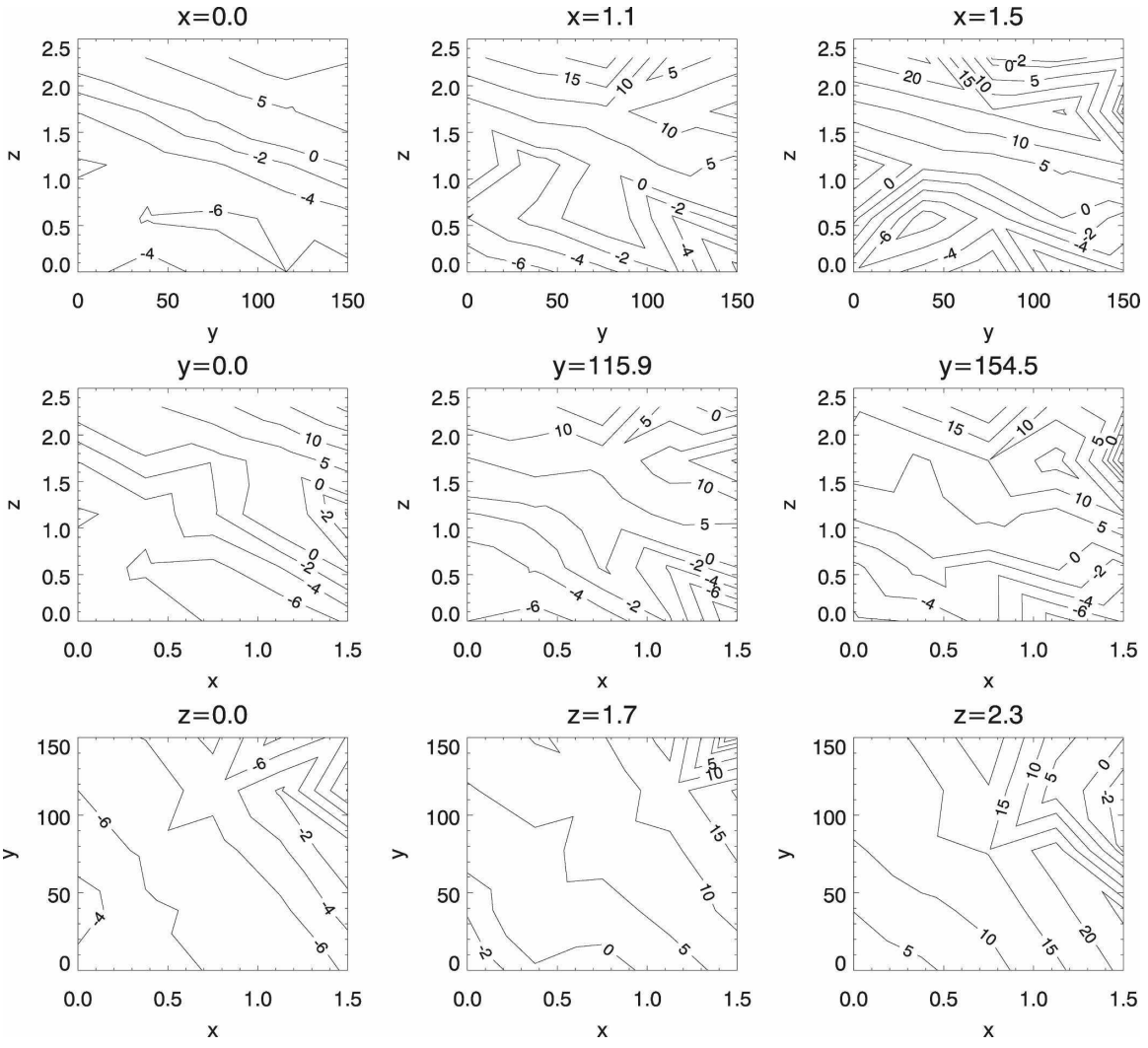

FIG. 1. Difference for the predicted CDNC (N cm^{-3}) between model results and the JRS94 parameterization.

for the modified Shepard method was almost 4 times higher (18.5 s). This implies that for the three-component aerosol–cloud dataset generated by our model, the Hardy multiquadric performs better than the modified Shepard method. The latter, however, has the advantage of being already available to the scientific community because it is incorporated into the NAG library.

b. Comparison with existing parameterizations

One way to evaluate the predicted CDNC values generated by our new parameterization is to consider a particular case that can be parameterized by a series of existing aerosol–cloud schemes. If we assume that all the aerosol particles are only sulfate particles, then we can calculate the predicted CDNC values generated by several existing schemes, such as JRS94, BL95, GN00, MD02, and Quaas and Boucher (2005, hereafter QB05). Figure 3 shows the results obtained for a sulfate aerosol mass concentration ranging between 0 and 1.5 $\mu g\ m^{-3}$. The first thing to note is that the Hardy VMQ method that was used in this case provides a very accurate interpolation of the model results. Also, these model results are very close to those generated by the JRS94, GN00, and QB05 (which is essentially the BL95 parameterization with updated coefficients) parameterizations. The original BL95 scheme and the MD02 scheme seem to generate significantly more and fewer cloud droplets, respectively, than the other three schemes considered.

This comparison suggests that our new scheme is able to reproduce results generated by other existing schemes for the particular case in which sulfate is the only aerosol species considered. However, the strongest

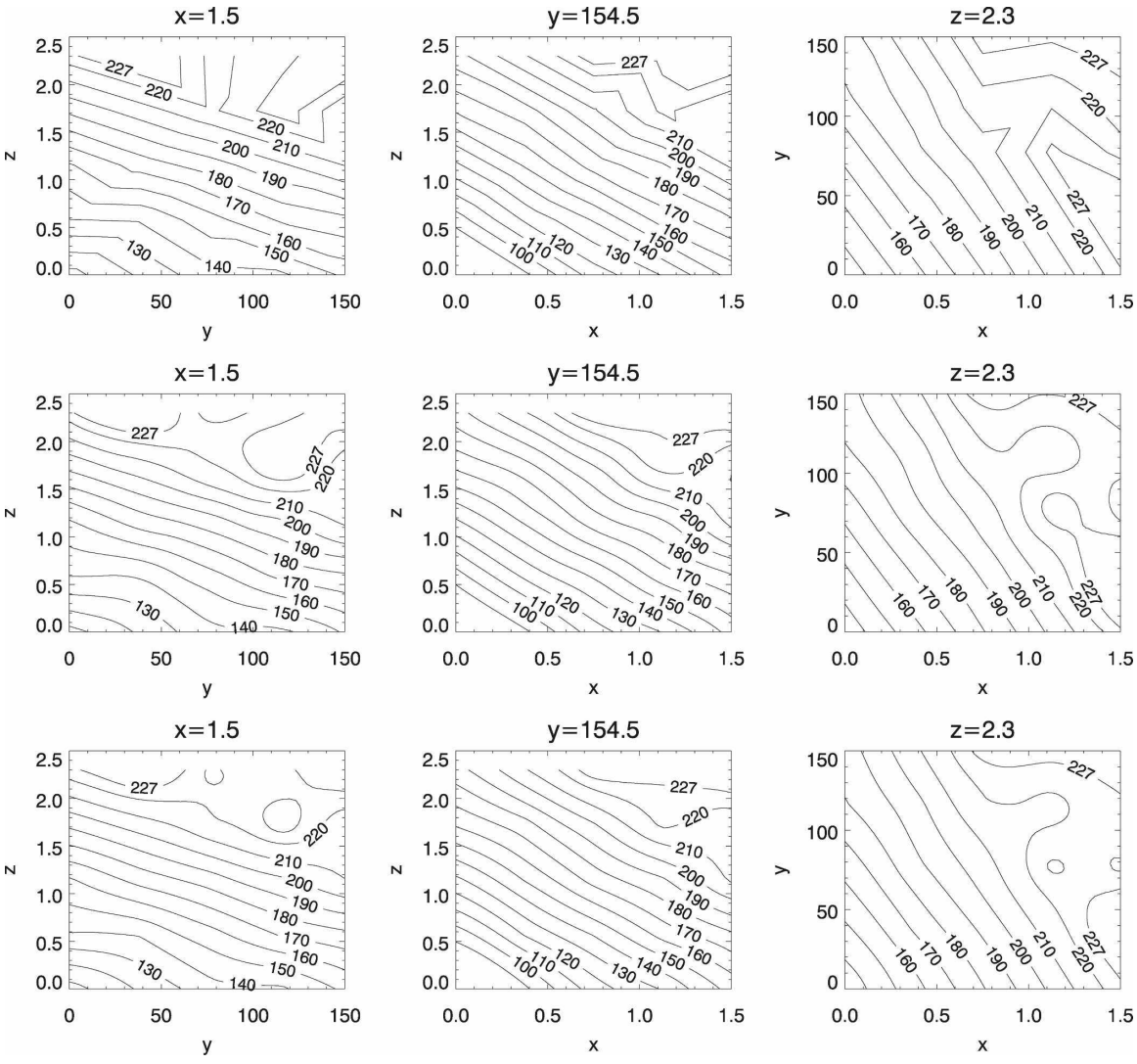

FIG. 2. CDNC values (N cm^{-3}) before interpolation (top three panels), after modified Shepard interpolation (middle three panels), and after Hardy VMQ interpolation (bottom three panels).

point of this new scheme is the possibility to account for three different aerosol species.

As reported in Ghosh et al. (2007), in the three aerosol species case, it is possible that the CDNC decreases when the aerosol mass loadings are increased. This decrease is due to the competition for available water vapor in the air parcel. When there is a preponderance of aerosol particles competing for a limited amount of water vapor, the supersaturation decreases and some of the aerosol particles are not able to grow into cloud droplets, resulting in a fall in CDNC values.

Investigating further the activation process in the case when two specific sulfate and biomass smoke loadings are considered (i.e., 1.3 $\mu g\ m^{-3}$ sulfate and 2.3 $\mu g\ m^{-3}$ biomass smoke), we can plot the contribution of each species to the total CDNC when a variable sea-salt loading is assumed. Figure 4 illustrates the number of aerosol particles that become activated to form cloud droplets; Table 2 shows the percentages of particles activated for each of the three aerosol species. It can be seen that when the sea-salt loading is 0 $\mu g\ m^{-3}$, 70% and 83% of the aerosol particles activate for sulfate and biomass smoke, respectively. When sea-salt particles are added in small loadings (i.e., up to 34 $\mu g\ m^{-3}$), then 72% of them activate to form cloud droplets, without affecting the other two species. This leads to an increase in the total CDNC. With higher sea-salt loadings, the competition for the available water vapor increases and the sea-salt particles suppress the activation of some of the particles from the other two species. The minimum

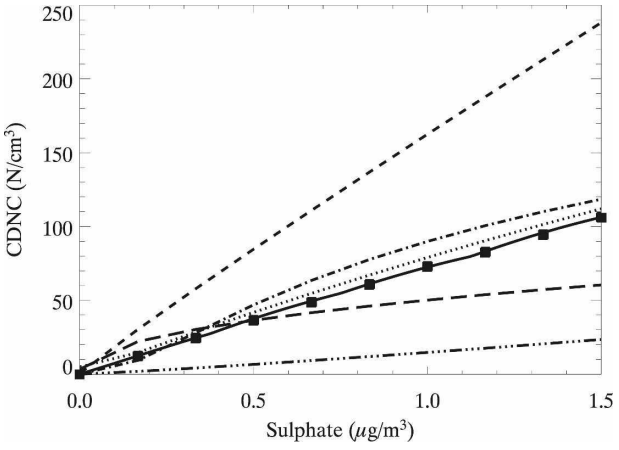

FIG. 3. CDNC values (N cm^{-3}) as a function of sulfate aerosol mass concentration ($\mu g\ m^{-3}$) predicted by the model runs (solid black squares) and the following parameterizations: new scheme (solid), JRS94 (dotted), BL95 (short dashed), GN00 (dashed–dotted), MD02 (dashed–triple dotted), and QB05 (long dashed). This case considers fixed aerosol mass loadings of 0 $\mu g\ m^{-3}$ sea salt and 0 $\mu g\ m^{-3}$ biomass smoke.

CDNC in this case is reached for a sea-salt mass loading of 103 $\mu g\ m^{-3}$, which occurs when the percentages of activated particles decrease from 70% and 83%, recorded for the small sea-salt loadings, to 50% and 80% for sulfate and biomass smoke, respectively. When the sea-salt mass loading is increased even further, the percentage of activated particles increases to the initial value of 83% for biomass smoke but continues to decrease for two species (i.e., sulfate and sea salt). However, although the overall percentage of activated particles decreases, the total CDNC increases because of the high number of available particles.

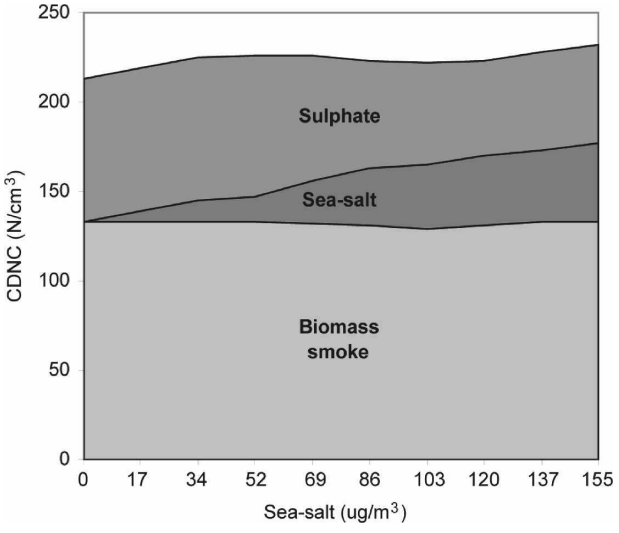

FIG. 4. CDNC values (N cm^{-3}) predicted by the model to nucleate from the three aerosol species as a function of sea-salt aerosol mass concentration ($\mu g\ m^{-3}$). This case considers fixed aerosol mass loadings of 1.3 $\mu g\ m^{-3}$ sulfate and 2.3 $\mu g\ m^{-3}$ biomass smoke.

TABLE 2. Percentages of particles activated from each aerosol species for a fixed aerosol mass loading of 1.3 $\mu g\ m^{-3}$ sulfate and 2.3 $\mu g\ m^{-3}$ biomass smoke, and variable sea-salt mass loadings.

	Sea-salt mass loadings ($\mu g\ m^{-3}$)									
	0	17	34	52	69	86	103	120	137	155
Sulfate (%)	70	70	70	70	62	52	50	46	48	48
Biomass (%)	83	83	83	83	82	81	80	81	83	83
Sea salt (%)	—	72	72	56	72	80	74	68	61	60

Among the five existing schemes considered above, the JRS94 parameterization is the only one that can model this more general case, with three aerosol species. Figure 5 illustrates the CDNC values generated by our new scheme and by the JRS94 parameterization for the case in which 1.3 $\mu g\ m^{-3}$ sulfate and 2.3 $\mu g\ m^{-3}$ biomass smoke loadings are considered. It can be observed that there is a significant difference between the two parameterizations. Whereas the JRS94 parameterization assumes a linear behavior for the activation process, the new scheme is able to interpolate very accurately the model results that suggest the existence of nonlinearities in this process.

7. Summary and conclusions

The aim of this study has been to develop a novel technique for the multicomponent aerosol–cloud pa-

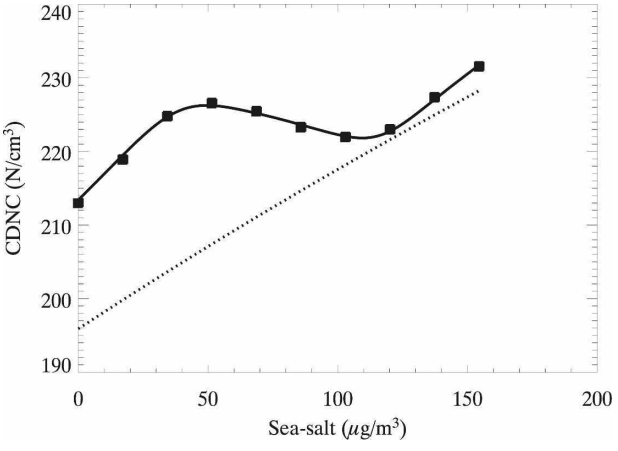

FIG. 5. CDNC values (N cm^{-3}) as a function of sea-salt aerosol mass concentration ($\mu g\ m^{-3}$) predicted by the model runs (solid black squares), the new parameterizations scheme (solid line), and the JRS94 parameterization (dotted). This case considers fixed aerosol mass loadings of 1.3 $\mu g\ m^{-3}$ sulfate and 2.3 $\mu g\ m^{-3}$ biomass smoke.

rameterization in global climate modeling. The approach chosen was based on the interpolation of a set of results generated by a microphysical chemical parcel model. Two different interpolation methods, namely the modified Shepard and the Hardy multiquadric methods, were investigated. By definition, interpolation methods are not precisely accurate, except at the points through which the function has been fitted. However, intuitively, the cloud droplet activation process involves a reasonable progression, rather than strong oscillations, of the CDNC from a data point to another data point in its vicinity. As illustrated in the numerical results section, both methods were able to reproduce accurately the initially known CDNC va1ues and to generate new interpolated values in distributions that are all intuitively reasonable for a wide range of aerosol mass loadings.

Investigations of the computational cost required by the two interpolation methods showed that the Hardy multiquadric technique is almost 4 times faster than the modified Shepard interpolation routine from the NAG library. Therefore, of the two methods, the former is a better candidate to be used within a GCM for aerosol–cloud parameterizations.

The method developed in this study seems to be in very good agreement with other existing schemes, such as JRS94, GN00, and QB05, in the particular case when only sulfate aerosol is considered. However, the main feature of this new scheme is its ability to accommodate three aerosol species and to model the nonlinearities of the activation process. This is expected to have a significant contribution in improving the current understanding and quantification of the aerosol indirect effect.

Future studies should focus on integrating new aerosol components (e.g., soot and/or mineral dust) into the microphysical chemical parcel model and on the development of appropriate interpolation techniques for these more complex cases.

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