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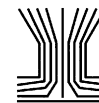
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Numerical Investigation of Sea Salt Aerosol Size Bin Partitioning in Global Transport Models: Implications for Mass Budget and Optical Depth

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In this study the importance of sea salt aerosol (SSA) size representation in a global transport model is investigated. For this purpose the Navy Aerosol Analysis and Prediction System (NAAPS) model is employed in a number of SSA simulations. A new dry deposition velocity parameterization is implemented into NAAPS in order to more physically represent deposition processes in the model. SSA size distribution is divided into size bins using two different partition procedures: the previously used iso-log method and the iso-gradient method, which relies on size-dependence of deposition processes. The global SSA simulations are analyzed in terms of the total sea salt mass and the average SSA optical thickness. The results indicate that there is a large dependence of the total mass and average aerosol optical depth on the number of size bins used to represent the aerosol size distribution. The total SSA mass is underestimated by 20% if 2 instead of 15 (reference) size intervals are used. The average aerosol optical depth underestimation is even higher and reaches over 35%. Such large differences can have substantial implications on the accuracy of SSA radiative forcing simulations in climate models. A comparison of the two division procedures shows that the simulations with the iso-gradient intervals are more accurate than the iso-log ones if at least 6 size bins are used. This result indicates that the more physically based division scheme can offer better performance and reduce computational cost of global aerosol transport models.

1. INTRODUCTION

Sea salt aerosol (SSA) is one of the most abundant components of the marine atmosphere. It exerts a strong influence on a broad range of geophysical and chemical processes occurring over oceans. It affects atmospheric chemistry, cloud physics,

atmospheric radiation, remote sensing, and air quality. The importance of SSA on Earth's radiation budget has recently been recognized (Winter and Chylek 1997; Haywood et al. 1999; Jacobson 2001; Dobbie et al. 2003; Satheesh and Lubin 2003; Ma et al. 2008; Ayash et al. 2008). SSA particles scatter solar radiation and affect the microphysics of marine clouds by serving as condensation nuclei for cloud drops (Feingold et al. 1999; O'Dowd et al. 1999a,b; Mason 2001; Rosenfeld et al. 2002; Lohmann and Feichter 2005). Estimates of the global mean clear-sky direct SSA radiative forcing range between -0.15 and -2.2 (Wm^{-2}) (Grini et al. 2002; Ma et al. 2008; Ayash et al. 2008; Dobbie et al. 2003) and are based on model simulations. A more extreme value based on simplified emission assumptions is reported by Haywood et al. (1999), who estimate SSA forcing for "high" concentration case to be as high as -5.03 Wm^{-2} (ocean averaged). The evaluation of the SSA indirect effect is even more complicated due to the complex microphysical interactions between aerosols and clouds. Some recent model studies by Ma et al. (2008) indicate the first indirect SSA radiative forcing to be -1.34 (Wm^{-2}) and the total indirect forcing to be -2.9 (Wm^{-2}) if climatic feedbacks are taken into account. A much smaller assessment of the indirect effect equal to -0.38 (Wm^{-2}) was reported by Ayash et al. (2008). Such a large spread of direct and indirect forcing estimates is mainly related to the uncertainties in sea salt emission modeling (Ayash et al. 2008). The uncertainties of emission function formulation and inadequate simulation of wind velocity and its variability in global models are considered to be the main culprits (Lewis and Schwartz 2004; Pierce and Adams 2006; Ma et al. 2008). However, another aspect that contributes to these uncertainties which has not been so far sufficiently studied is the representation of the SSA size distribution in numerical models.

Accurate representation of aerosol size distributions is important in modeling aerosol dynamics, chemistry and radiative forcing because of strong size-dependence of these processes. Two alternative approaches are typically employed in air quality models: (a) the modal approach (Whitby and McMurry 1997; Ackermann et al. 1998; Vignati et al. 2004; Sartelet et al. 2006)

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and (b) the sectional approach (Jacobson 1997; Meng et al. 1998; Pilinis et al. 2000; Gong et al. 2003; Ma and von Salzen 2006). Recently von Salzen (2006) proposed a hybrid approach, which incorporates advantages of both modality and sectioning in representing aerosol size distribution in numerical models. This hybrid method was further employed in studies of the sea salt aerosol direct and indirect radiative forcing by Ma et al. (2008).

A limited number of studies were focused on comparing results of the modal and bin approaches (Zhang et al. 1999). With the use of a one-dimensional version of a climate-aerosol-chemistry model Zhang et al. (2002) investigated the impact of aerosol size representation on aerosol-cloud interactions. Korhonen et al. (2003) studied the impact of size distribution representation on atmospheric particle formation and growth. Foret et al. (2006) investigated the influence of the size bin number and the size bin scheme selection on mineral dust concentrations and optical thickness using a simple box model. As an extension to the study by Foret et al. (2006), Menut et al. (2007) tested sensitivity of mineral dust concentrations to the model size distribution using a mesoscale transport model. However, uncertainties remain in terms of the influence of the size bin selection on model results in global three-dimensional simulations. It is unknown how these uncertainties might affect aerosol radiative properties and the simulated global energy budget. A common opinion in the aerosol modeling community is that the more size bins are used to represent the size distribution the better. On the other hand introducing many size intervals is computationally expensive, especially when simulations on a global scale are performed. There is usually a trade-off between the number of size bins and the availability of computational resources.

SSA size distributions are often represented in numerical models based on the sectional approach. The particle size distribution is divided into a limited number of intervals, which are usually defined to have equal ranges in logarithmic progression (Takemura et al. 2000; Guelle et al. 2001; Grini et al. 2002; Gong et al. 2002). Such splitting and the resulting so-called “isolog” size bins introduce additional uncertainties in model simulations, dependent on the number of bins used to represent the size spectrum and the width of the distribution. Recently Foret et al. (2006) introduced an alternative “iso-gradient” method of dividing size distributions into bins. This method leads to increased resolution in size ranges where size dependent processes are the most intense and strongly changing for a small variation in particle diameter. The bins are defined according to the dry deposition velocity gradient, minimizing the extent of dry deposition velocity variations within each size interval. Hence, the iso-gradient method accounts for the size dependence in dry deposition processes, which are a driving mechanism affecting transport and removal of aerosol particles.

This study addresses the problem of the impact of the bin number selection on the global SSA budget and average SSA

optical properties. These numerical errors in simulating SSA are expected to have direct influence on radiative forcing predictions in climate models. Furthermore, differences between results obtained with the iso-log and iso-gradient division methods are investigated. Section 2 describes methodology used in this study. The Navy Aerosol Analysis and Prediction System model (Christensen et al. 1997; Witek et al. 2007) is briefly introduced, followed by a description of a deposition velocity scheme that was implemented in the aerosol transport model. This section also introduces the iso-gradient bin sectioning method - a modified version of the scheme introduced by Foret et al. (2006), as well as the SSA optical package that was used to derive the asymmetry parameter and aerosol optical thickness (AOT). Section 3 presents results of the multi-bin SSA simulations. The total SSA mass and average AOT are analyzed. In section 4 some conclusions and summary of the analysis are offered.

2. METHODOLOGY

2.1. Aerosol Transport Model

Navy Aerosol Analysis and Prediction System (NAAPS) (Christensen et al. 1997; Witek et al. 2007) is a global semi-lagrangian aerosol and air pollution model, developed in the Naval Research Laboratory in Monterey, CA. Physical processes that apply to aerosol species include emission from the surface, dispersion and advection by the wind and removal from the atmosphere by wet and dry deposition. The model resolution used in this study is 1 degree in horizontal and 24 vertical terrain-following sigma levels. The average depth of the first layer is around 34 meters and the top layer ends at around 18 km. The integration time step is 15 min for vertical diffusion and 30 min for all other processes. The NAAPS model is driven by meteorological fields obtained from Navy Operational Global Atmospheric Prediction System (NOGAPS) (Hogan and Rosmond 1991). NOGAPS analyses are available every 6 h (at 00, 06, 12 and 18 UTC).

In NAAPS, the size dependant sea-salt emission function is given by (Monahan et al. 1986)

$$\frac{dF}{dr_{80}} = 1.373 U_{10}^{3.41} r_{80}^{-3} (1 + 0.057 r_{80}^{1.05}) 10^{1.19 \exp(-B^2)}, \quad [1]$$

where r_{80} is the particle radius in μm at 80% RH, U_{10} is the wind speed at 10 m above the sea surface, and $B = [0.38 - \log(r_{80})]/0.65$. dF/dr_{80} , given in units of $\text{m}^{-2}\text{s}^{-1}\mu\text{m}^{-1}$, describes the areal flux of the number of sea salt particles with the radius $r \pm dr$ produced over the ocean surface. Originally Monahan et al. (1986) defined this source function for particles with radii ranging from 0.8 to 8 μm at 80% RH. The study by Gong (2003) and laboratory results obtained by Martensson et al. (2003) indicate that this formulation can be extended down to 0.2 μm without introducing much uncertainty. This revised size range, 0.2–8 μm , is used in this

study. The dry sea salt mass emitted within a size bin is obtained by noting that $dF/dr_{dry} = 2dF/dr_{80}$ and integrating over the mass of dry particles within the size limits of the bin. The exact equation is given further in the text (Equation (17)).

2.2. Dry Deposition Velocity Model

The parameterization of particle dry deposition is based on the concept of dry deposition velocity V_d (Slinn and Slinn 1980; Slinn 1982; Zhang et al. 2001; Lewis and Schwartz 2004). This approach has already been described in several publications, however, due to some modifications made specifically for NAAPS and for the sake of clarity, the concept is described herein.

The theory originally developed by Slinn and Slinn (1980) divides the lowest part of the atmosphere into two virtual layers: the surface layer (up to about 10 meters above the surface) and the viscous sublayer (first few centimeters of the atmosphere). Particles mix between the upper and lower layer, but actual deposition takes place only in the lower layer. In each layer different processes influence aerosol transport and depletion. In the viscous sublayer, where the actual ground deposition takes place, the key processes involve Brownian diffusion, impaction, and gravitational settling. In the surface layer, the dominant processes are turbulent mixing and gravitational settling, which force particles to move towards the lower viscous sublayer. All these mechanisms are parameterized by corresponding transfer velocities which strongly depend on particle size and meteorological conditions.

The overall size-dependent dry deposition velocity V_d [m/s] in the model is described by the following equation:

$$V_d = \frac{V_V V_S}{V_V + V_{turb}}, \quad [2]$$

where $V_V = V_{gV} + V_{diff} + V_{imp}$ and $V_S = V_{gS} + V_{turb}$ are the cumulative deposition velocities [m/s] in the viscous sublayer and the surface layer, respectively. V_{diff} is the Brownian diffusion velocity, V_{imp} is the impaction velocity, V_{turb} is the turbulent diffusion velocity, V_{gV} and V_{gS} are the gravitational sedimentation velocities in the viscous sublayer and the surface layer, respectively. These two sedimentation velocities are distinguished because of different relative humidities in the two layers. A constant 98% relative humidity (RH) is assumed in the viscous sublayer over oceans, whereas the RH in the surface layer and in the viscous sublayer over land is computed on the basis of water vapor mixing ratio, temperature and pressure.

The gravitational settling velocity in each layer is represented by the Stokes velocity:

$$V_g = \frac{2r_a^2 \rho_a g C_c}{9\mu_{air}}, \quad [3]$$

where r_a [m] is the particle radius at the ambient RH (particles are assumed to be spherical), ρ_a [g/m³] is the particle density,

g [m/s²] is the acceleration due to gravity, C_c is the Cunningham correction factor and μ_{air} [Pa s] is the dynamic viscosity of air. The Cunningham factor is not negligible only for small particles (r_a smaller than 1 μ m) and is expressed as (Seinfeld and Pandis 1998):

$$C_c = 1 + \frac{6.6 \times 10^{-8}}{r_a} [1.257 + 0.45 \exp(-1.667 \times 10^7 r_a)]. \quad [4]$$

The air dynamic viscosity, according to the Sutherland equation (Sutherland 1893), is a function of air temperature and is given by (White 1991):

$$\mu_{air}(T) = \frac{bT^{3/2}}{T + S}, \quad [5]$$

where $b = 1.458 \times 10^{-6}$ [kgm⁻¹s⁻¹K^{-1/2}] is a constant, $S = 110.4$ [K] is the Sutherland constant, and T [K] is air temperature. Both r_a and ρ_a depend on RH; r_a can be derived with the use of the growth function: $r_a(rh) = r_{dry} f(rh)$. $f(rh)$, defined as the ratio of particle radius at rh (fractional relative humidity) to its dry equivalent, is described as (Lewis and Schwartz 2004):

$$f(rh) = \begin{cases} 2 \times 0.67 / (1 - rh)^{1/4} & rh \leq 0.93, \\ 2 \times 0.54 / (1 - rh)^{1/3} & 0.93 < rh \leq 0.98. \end{cases} \quad [6]$$

The change in particle density with rh is approximated with the following second order polynomial $\rho_a(rh) = -0.11rh^2 - 0.45rh + 1.61$. Such approximation was chosen to ensure proper behavior of the product $\rho_a(rh) f(rh)^2$ (a part of the Stokes velocity equation) for the full RH range.

The Brownian diffusion velocity V_{diff} is typically parameterized in terms of the Schmidt number:

$$V_{diff} = u_* Sc^{-p}, \quad [7]$$

where u_* [m/s] is the wind friction velocity and p is a coefficient which accounts for the surface type (the value 1/2 is used for open water and snow/ice areas, whereas 2/3 is used for all other surface categories). Sc (dimensionless) represents the ratio of the kinematic viscosity of air to the Brownian diffusivity of an aerosol particle. In practice, Sc can be approximated as (Lewis and Schwartz 2004):

$$Sc \approx 3 \times 10^{12} \frac{(r_a / \mu m)}{C_c}. \quad [8]$$

The impaction accounts for the inability of a particle to respond rapidly to a non-uniform flow near the surface. It plays considerable role only for larger particles ($r_a > 1 \mu$ m). The inertial impaction velocity V_{imp} is parameterized as:

$$V_{imp} = u_* 10^{-3/S_i}, \quad [9]$$

where St is the dimensionless Stokes number. For numerical purposes, the Stokes number can be approximated by (Lewis and Schwartz 2004):

$$St \approx 0.004 \times 10^{12} \left(\frac{r_a}{\mu\text{m}} \right)^2 \left(\frac{U_{10}}{\text{ms}^{-1}} \right)^2. \quad [10]$$

The turbulent transfer or turbulent diffusion velocity V_{turb} accounts for mixing processes in the surface layer which transport particles towards the lower viscous sublayer. An accurate parameterization of this transfer velocity is important since, in general, it is a very efficient mechanism for particle removal. The turbulent transfer mostly depends on atmospheric stability and surface roughness. For neutral atmospheric conditions (assumed here), the expression for V_{turb} is:

$$V_{turb} = \frac{ku_*}{\ln(z/z_0)}, \quad [11]$$

where $k = 0.4$ is the von Karman constant, z is the reference height for wind velocity (10 m) and z_0 [m] is the roughness length of the surface.

The surface roughness z_0 in each of the model grids is computed according to the formula:

$$z_0 = \sum \text{veg}_i z_{0i}, \quad [12]$$

where veg_i is a fraction of vegetation type in the grid and z_{0i} is the prescribed roughness length corresponding to each vegetation type. Eight surface types (Table 1) are defined in the model. Roughness lengths for these vegetation types vary with season and are taken after Voldner et al. (1986) with some modifications after Zhang et al. (2001). All values in Table 1 are prescribed except for the open water category, where the following equation is applied:

$$f(u_*, U_{10}) = 10 \exp\left(-\frac{U_{10}k}{u_*}\right). \quad [13]$$

TABLE 1

Surface roughness length z_{0i} [m] for 8 land categories during 4 different seasons

	Winter	Spring	Summer	Fall
1. Coniferous forest	0.90	0.80	0.80	0.90
2. Deciduous forest	0.55	0.75	1.05	0.95
3. Cultivated land	0.02	0.05	0.10	0.02
4. Grassland	0.02	0.05	0.10	0.05
5. Urban	1.00	1.00	1.00	1.00
6. Swamp	0.02	0.03	0.15	0.10
7. Open water	$f(u_*, U_{10})$			
8. Snow/ice	0.01	0.01	0.01	0.01

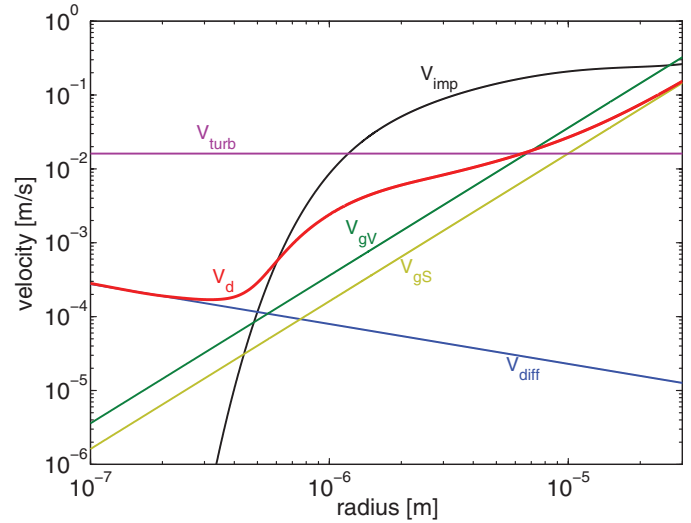


FIG. 1. Average dry deposition velocity and different transfer velocities calculated from the described model as a function of the ambient aerosol radius. Averaging was done over values based on the NOGAPS meteorological fields (180×360 data points). For a description of the symbols see the text.

This parameterization is derived from the logarithmic wind profile under neutral atmospheric stability.

Figure 1 pictures all components of the dry deposition velocity (averaged over model domain 180×360) computed with the use of sample meteorological fields from the NOGAPS global circulation model (1 July 2006, 00 UTC). Many deposition velocities exhibit large, up to several orders of magnitude, spreads within each size category (only average values are presented). This is due to the large variability of meteorological conditions derived from the NOGAPS atmospheric model. The average trends agree well with those presented by Lewis and Schwartz (2004) and Foret et al. (2006). Average V_d exhibits a strong dependence on the particle size and may change by three orders of magnitude from $0.1 \mu\text{m}$ to $30 \mu\text{m}$. There is a minimum in V_d at about $0.3 \mu\text{m}$. As small velocity increase towards smaller sizes is caused by the Brownian diffusion mechanism, the velocity increase towards larger sizes is caused mainly by the gravitational sedimentation and impaction. In general, Figure 1 shows that the described parameterization can reasonably resolve the dry deposition velocity under variable meteorological conditions taken from a global atmospheric circulation model.

One of the characteristics of the described parameterization is the difference in dry deposition velocity over the land and over the ocean, which is clearly visible in Figure 2. This difference is caused by the assumption that in the viscous sublayer the ambient RH over the ocean is 98%, whereas over the land is the same as in the surface layer, which is usually lower than 98%. This assumption causes particles to have larger radius over the ocean than over the land. This, in turn, enhances the impaction and gravitational settling and results in a higher deposition velocity over the ocean. However, this mechanism does not explain a

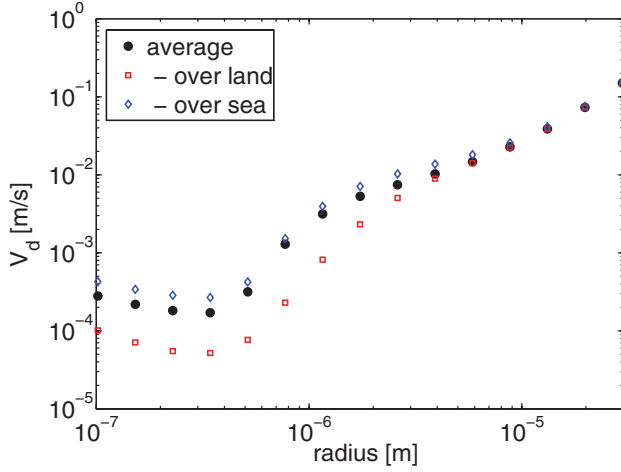


FIG. 2. Average dry deposition velocities in the model as a function of the ambient aerosol radius. The land and ocean values are indicated by the open squares and the open diamonds, respectively. The solid circles indicate averaged land-ocean values.

larger difference in the deposition velocity in the lower sub-micron regime (particles smaller than 0.5 micron in radius). In this size regime, the impaction and gravitational settling are much less effective than the Brownian diffusion, which plays a key role in particles deposition. The observed trend can be explained by the fact that different Brownian velocity parameterizations are used for the open water area and all other surface categories. Coefficient p in Equation (7) is set to $1/2$ for the open water (Slinn and Slinn 1980; Zhang et al. 2001) and to $2/3$ for other land types (Slinn 1982; Foret et al. 2006).

2.3. Size Bin Division Method

A typical approach in aerosol transport models is to divide size spectra in terms of the iso-log size intervals, i.e., following a constant geometric progression of the particle radius. Foret et al. (2006) showed limitations of such an approach and proposed an alternative iso-gradient size segregation procedure. This method relies on the size-dependency of the dry deposition velocity. Foret et al. (2006) applied this method to mineral dust aerosol and showed its usefulness in a simple box model numerical study. Menut et al. (2007) found large benefits of using the iso-gradient size-segregation procedure in simulating mineral dust concentrations in a three-dimensional mesoscale transport model.

The method described here follows the iso-gradient concept introduced by Foret et al. (2006), but uses a different procedure of determining the size ranges of different bins. Conceptually, this proposed method minimizes differences in the dry deposition velocity at the edges of each size interval. In other words, constrained by a limited number of size bins, the method forces the dry deposition velocity to be as much uniform as possible

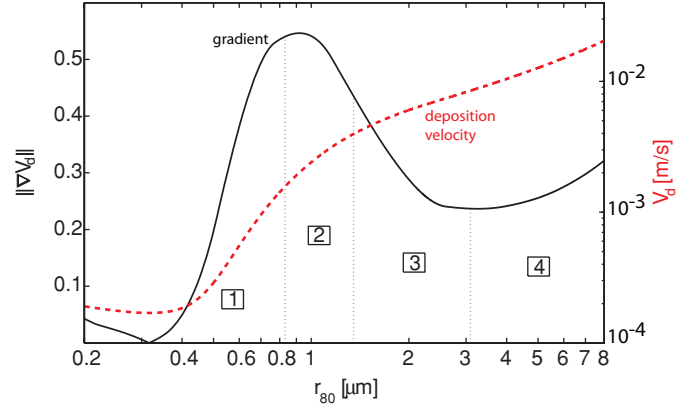


FIG. 3. Right axis: average dry deposition velocity (dashed line) derived numerically from the model described in 2.2. Left axis: normalized gradient of the dry deposition velocity (solid line). Dotted vertical lines divide the area below $\|\nabla V_d\|$ into four equal segments.

within the limits of each bin. This minimizes the deposition variability of particles in a given bin.

Determination of the limits of size bins in the iso-gradient division method is performed using the procedure described below. This technique can be applied to any given size range and any shape of the deposition velocity function. In the present study the procedure is described for a size range spanning from $r_{min} = 0.2 \mu\text{m}$ up to $r_{max} = 8 \mu\text{m}$. This choice corresponds to the size range of the Monahan's emission function at 80% RH (Monahan et al. 1986).

First, a deposition velocity V_d has to be established within specific size limits. Here we use an average V_d derived numerically from the model described in 2.2. This function is presented in Figure 3 with the dashed line (right axis). The solid line (left axis) in Figure 3 shows the change in normalized deposition velocity as a function of radius, $\|\nabla V_d\|$. Note the difference in vertical scales on the right and left axes. Normalization is defined as:

$$\Psi = \int_{r_{80min}}^{r_{80max}} \nabla V_d(r_{80}) d \log(r_{80}), \quad [14]$$

where index 80 denotes referential RH for the radii, and then:

$$\|\nabla V_d\| = \frac{\nabla V_d}{\Psi} \Rightarrow \int_{r_{80min}}^{r_{80max}} \|\nabla V_d\| d \log(r_{80}) = 1. \quad [15]$$

It is important to notice that integration over $\log(r_{80})$ rather than r_{80} is performed. This allows for a better representation of the sub-micron size range, where deposition velocity changes considerably. The area below $\|\nabla V_d\|$ shown in Figure 3 integrates to unity and can be divided into a desired number of equal area segments using a simple procedure. Mathematically

the limits of equal area parts are derived progressively from the equation:

$$\int_{r(i)}^{r(i+1)} \|\nabla V_d\| d \log(r) = \frac{1}{n}, \quad [16]$$

where n is the number of size bins, $i = 1, 2, \dots, n+1$, and $r(1) = r_{min}$, $r(n+1) = r_{max}$. Figure 3 is an example of the division into four equal segments (cf. vertical dotted lines). These division lines define the limits of the size bins.

The sea salt mass emission within a size bin is derived according to the equation:

$$F_{dry}(i) = \frac{4}{3} \pi \rho_{ss} \int_{r_i}^{r_{i+1}} 2 \frac{dF}{dr_{80}} r_{dry}^3 dr_{dry} \text{ [g / m}^2\text{s]}, \quad [17]$$

where $\rho_{ss} = 2.2 \times 10^6 \text{ [g / m}^3\text{]}$ is the dry sea salt density and dF/dr_{80} is the Monahan's SSA emission function (Monahan et al. 1986). The equivalent radius of particles assigned to a bin (r_{eqv} [μm]) is obtained by using a weighting function in the form $v_d(r) r_{dry}^3 dF/dr_{80}$. It is expressed by:

$$r_{eqv}(i) = \exp \left\{ \left(\int_{r_i}^{r_{i+1}} \log(r) v_d(r) \frac{dF}{dr_{80}} r_{dry}^3 dr \right) / \left(\int_{r_i}^{r_{i+1}} v_d(r) \frac{dF}{dr_{80}} r_{dry}^3 dr \right) \right\}, \quad [18]$$

where $v_d(r)$ is given by Equation (2). Such an arbitrarily adopted weight function accounts for the deposition velocity variability and the emitted mass size distribution.

Figure 4 (top panel) presents the average size dependent dry deposition velocity and the size bins limits (lower panel) for a various number of divisions. Size intervals for both the iso-gradient (thick lines) and the iso-log (dotted lines) division methods are presented for comparison. Additionally, a sample of r_{eqv} and dry sea salt mass emission values for 2, 4, and 6 size intervals obtained with the iso-gradient method are presented in Table 2.

For two size bins, the division is at about 1 micron regardless of the method used, which can be interpreted as the fine and coarse aerosol mode selection. As more sections are introduced, the iso-log procedure generates equal-length bin intervals in the logarithmic progression. In the case of the iso-gradient division procedure, it can be observed that as the number of size intervals increases the regions of highly variable V_d are more accurately covered with new bins. This is reflected by the tightening of the bins in the critical area at around 1 micron size range (Figure 4). On the other hand, all particles smaller than $0.5 \mu\text{m}$ are enclosed in one size interval even when as many as 22 size bins are used. This can be explained by the fact that all these particles,

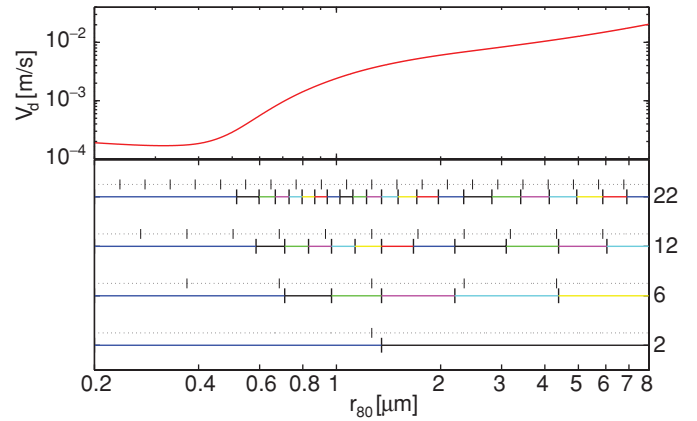


FIG. 4. Top panel, average dry deposition velocity (Section 2.2). Lower panel, size limits for a set of 2, 6, 12, and 22 size bins derived with the iso-gradient size division procedure (solid lines). The dotted lines denote a similar set of boundaries but for the iso-log size intervals.

according to the shape of the V_d function, deposit at a similar rate. The enumerated features of this bin division technique highlight its advantage over the previous iso-log method. The generated iso-gradient size intervals are characterized by the minimal variability in the deposition rate of all particles enclosed in a bin. This, in turn, is expected to improve the accuracy of model simulations. This statement will be verified in next sections.

2.4. Sea Salt Optical Properties Model

The knowledge of aerosol optical properties is essential for radiative forcing computation. In modeling studies many assumptions and simplifications, with respect to optical properties, are often made that affect accuracy of the assessment of aerosol influence on climate. Aerosol size-binning introduces yet additional numerical uncertainties. It is therefore important to properly derive SSA optical properties to be able to assess the significance of size-bin selection on radiative forcing calculations.

In this section, a SSA optical properties calculation method in each size interval is discussed. The optical properties of importance for the computation of shortwave radiative forcing in the commonly used two-stream approximation include the extinction coefficient σ_{ext} , the single scattering albedo ω , and the asymmetry parameter g . Since the present study is limited to the analysis of sea salt optical depth only a method of calculating σ_{ext} is presented.

For the case of highly hygroscopic SSA particles, the change in size of a solution droplet with ambient RH needs to be taken into account in the derivation of optical properties. Therefore, a transformation relation into a different RH that conserves the total number of particles needs to be formulated. This transformation takes the form:

$$n(r_a) dr_a = \tilde{n}(r_{80}) dr_{80}, \quad [19]$$

TABLE 2

Dry mass m_{vp} , extinction cross-section $\sigma_{e,vp}$ (at 550 nm wavelength), and equivalent radius r_{eqv} of virtual particles, as well as dry sea salt mass emission within a bin F_i , defined for 2, 4, and 6 iso-gradient size intervals. The shape of the size distribution function is based on Monahan et al. (1986) emission parameterization

N	bin	r_{dry} range [μm]	m_{vp} [g]	$\sigma_{e,vp}$ [m^2] (RH = 80%)	r_{eqv} [μm]	F_i [$\text{g} / \text{m}^2\text{s}$]
2	1	0.1–0.675	$2.5913 \cdot 10^{-13}$	$2.1975 \cdot 10^{-12}$	0.547	9.877×10^{-12}
	2	0.675–4.0	$3.3279 \cdot 10^{-12}$	$6.0212 \cdot 10^{-12}$	2.207	1.2685×10^{-10}
4	1	0.1–0.415	$7.6124 \cdot 10^{-14}$	$1.1068 \cdot 10^{-12}$	0.349	2.9017×10^{-12}
	2	0.415–0.675	$1.8301 \cdot 10^{-13}$	$1.0907 \cdot 10^{-12}$	0.573	6.976×10^{-12}
	3	0.675–1.548	$1.1828 \cdot 10^{-12}$	$3.3821 \cdot 10^{-12}$	1.153	4.5085×10^{-11}
	4	1.548–4.0	$2.1451 \cdot 10^{-12}$	$2.6391 \cdot 10^{-12}$	2.641	8.1758×10^{-11}
6	1	0.1–0.354	$5.1401 \cdot 10^{-14}$	$9.1621 \cdot 10^{-13}$	0.284	1.9593×10^{-12}
	2	0.354–0.484	$6.0392 \cdot 10^{-14}$	$4.9832 \cdot 10^{-13}$	0.431	2.3021×10^{-12}
	3	0.484–0.675	$1.4734 \cdot 10^{-13}$	$7.8295 \cdot 10^{-13}$	0.593	5.6163×10^{-12}
	4	0.675–1.099	$5.3018 \cdot 10^{-13}$	$1.9073 \cdot 10^{-12}$	0.909	2.021×10^{-11}
	5	1.099–2.192	$1.4569 \cdot 10^{-12}$	$2.7775 \cdot 10^{-12}$	1.634	5.5536×10^{-11}
	6	2.192–4.0	$1.3408 \cdot 10^{-12}$	$1.3364 \cdot 10^{-12}$	3.028	5.1108×10^{-11}

where $n(r) = dN(r)/dr$ [$1/(\text{m}^3 \mu\text{m})$] is a SSA number concentration function, that is the number of SSA particles per unit volume in a unit interval of r . Here and throughout this study the reference RH of 80% for particle radius is used (Lewis and Schwartz 2004). $\tilde{n}(r_{80})$ is the number concentration function at that reference RH. The index a denotes ambient RH. Equation (19) can be rewritten as:

$$n(r_a) = \frac{\tilde{n}(r_{80})}{\xi_a}, \quad [20]$$

where the ratio $\xi_a = dr_a/dr_{80}$ is equivalent to the growth function (Equation (6)).

The extinction coefficient can be written as:

$$\begin{aligned} \sigma_{ext}(RH) &= \int_{r_{min,a}}^{r_{max,a}} \pi r_a^2 Q_{ext}(RH) n(r_a) dr \\ &= \int_{r_{min,80\xi_a}}^{r_{max,80\xi_a}} \pi [r_{80\xi_a}]^2 Q_{ext}\left(\frac{2\pi r_{80\xi_a}}{\lambda}, m(RH)\right) \\ &\quad \times \frac{\tilde{n}(r_{80})}{\xi_a} dr, \end{aligned} \quad [21]$$

where Q_{ext} is the nondimensional extinction efficiency, m is the sea salt refractive index, and λ [m] is the wavelength of scattered light. For spherical particles, Q_{ext} can be derived from the Lorentz-Mie theory (Boren and Huffman 1983; van de Hulst 1957). Such a formulation allows for the derivation of σ_{ext} at any RH, given that the SSA number concentration $\tilde{n}(r_{80})$ at 80% RH is known. It should be noted that the transport model simulates the SSA dry mass instead of the number concentration $\tilde{n}(r_{80})$. Additionally, the use of a limited number of size

bins precludes modeling a continuous $\tilde{n}(r_{80})$ function. In order to reconstruct the $\tilde{n}(r_{80})$ function, the emission source function dF/dr is used. That information can be employed assuming that the shape of the size-dependent source function describes entirely the distribution of aerosol particles within a bin. One weakness of this assumption is that it prevents smooth and continuous changes in distribution within and between bins, which is especially relevant at coarse division settings. Nevertheless, it allows for a direct and accurate derivation of the aerosol optical properties based on the model output.

To facilitate this approach, we will introduce the concept of a virtual particle which will help reconstruct $\tilde{n}(r_{80})$. Such a virtual particle is constructed as a single unit which is described by some predefined size distribution probability function $n_{vp}(r)$ [$1/\text{m}$] defined for radii between r_1 and r_2 and which fulfills the relation:

$$\int_{r_1}^{r_2} n_{vp}(r) dr = 1. \quad [22]$$

The area and mass of a virtual particle are computed as integrals of second and third moments of its size distribution:

$$A_{vp} = \pi \int_{r_1}^{r_2} n_{vp}(r) r^2 dr, \quad m_{vp} = \frac{4}{3} \pi \rho \int_{r_1}^{r_2} n_{vp}(r) r^3 dr. \quad [23]$$

Additionally, the extinction cross-section $\sigma_{e,vp}$ is derived from equation:

$$\sigma_{e,vp} = \int_{r_1}^{r_2} \pi r^2 Q_{ext}\left(\frac{2\pi r}{\lambda}, m\right) n_{vp}(r) dr \quad [\text{m}^2]. \quad [24]$$

The predefined size distribution probability function which describes a virtual particle can be any function that satisfies the condition 22. Here it is assumed to have the same shape as Monahan's emission source function. To fulfill the condition 22, the emission function dF/dr has to be normalized, which is given by:

$$n_{vp}(r) = \left(\frac{dF}{dr} \right) / \int_{r_1}^{r_2} \frac{dF}{dr} dr. \quad [25]$$

Let us notice that function $n_{vp}(r)$ is defined at 80% RH. For any other humidity values, the $n_{vp}(r)$ is scaled according to Equation (20) with the use of the growth function ξ_a . In the case of a virtual particle encompassing the range of sizes $r_{80} \in (0.2, 8) [\mu\text{m}]$, that is the sizes for which Monahan's function is defined, the extinction cross-section at 80% RH ($\xi_a = 1.0$) is:

$$\begin{aligned} \sigma_{e,vp} &= \int_{0.2}^{8.0} \pi r^2 Q_{ext} \left(\frac{2\pi r}{\lambda}, m(80\%) \right) n_{vp}(r) dr \\ &= 8.2186 \times 10^{-12}. \end{aligned} \quad [26]$$

The mass of the pure sea salt of such a virtual particle, without the addition of water, is derived from combined Equations (23) and (20) (the growth parameter $\xi_a = 0.5$ for dry particles):

$$m_{vp} = \frac{4}{3} \pi \rho \int_{0.1}^{4.0} r^3 \frac{n_{vp}(r)}{0.5} dr = 3.587 \times 10^{-12} [\text{g}], \quad [27]$$

where the salt density ρ is set to $2.2 \times 10^6 [\text{g/m}^3]$. Table 2 presents pairs of extinction cross-section and dry mass for several different size bins. The limits of the size bins were derived using the iso-gradient division procedure described in Section 2.3.

Having the mass of a virtual particle, the NAAPS simulated sea salt mass concentration within a bin can be transformed into the total number of such particles:

$$N_{vp} = \frac{\rho_{sea\ salt}}{m_{vp}}. \quad [28]$$

In the next step, the actual extinction coefficient is computed by using a simplified version of Equation (21):

$$\sigma_{ext} = N_{vp} \sigma_{s,vp}. \quad [29]$$

Equations (28) and (29) complete the derivation of the SSA optical properties. The aerosol optical depth is then computed according to the equation:

$$\tau = \sum_{i=1, nlev} \sigma_{ext}(z_i) \delta z_i, \quad [30]$$

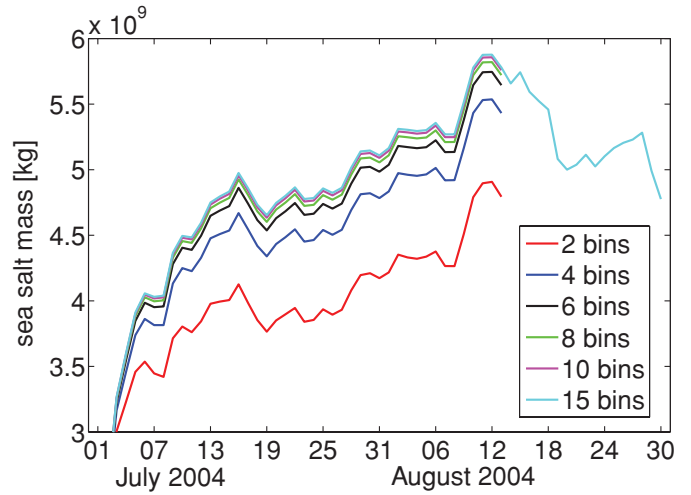


FIG. 5. Total sea salt dry mass as simulated by the model with various numbers of size bins. All simulations are initiated with the zero aerosol mass (cold-start) and are for the time period of 1 July–13 August, 2004. The simulation with 15 size bins extends until August 31, 2004.

where δz_i is the height of the i 'th model layer and $\sigma_{ext}(z_i)$ is the predicted extinction coefficient in the i 'th model layer.

3. MULTI-BIN SIMULATIONS: RESULTS

In this section, the NAAPS simulations carried out with the use of multiple SSA size bins are investigated and analyzed. The performance of the two different bin division techniques—the commonly used iso-log method and the iso-gradient method—is tested. The analyzed quantities include the total SSA mass and the average sea salt optical thickness.

3.1. Total Sea Salt Mass Analysis

Figure 5 presents the sea salt dry mass as simulated by the model with various numbers of size bins. The iso-gradient size division method was applied in these simulations. Mass accumulation can be observed at the beginning of each model run, which is related to the initialization process (cold-start). The vertical axis in Figure 5 is scaled to show only values above 3×10^9 kg. The simulations were performed for the period between July 1 and August 13, 2004. The simulation with 15 size bins, which is extended until August 31, demonstrates that a local maximum in the total mass is modeled on August 12, 2004. This solves a potential controversy regarding the sharp mass increase around August 10, which could be falsely interpreted as an additional mass accumulation in the model. Further analysis will be limited to the date August 13, 2004.

Figure 5 indicates that there is the total mass increase in the model when more size intervals are introduced. However, differences between the simulations become smaller with an increase in the number of size bins and the simulated mass seems to converge. For example, the simulations with 10 and

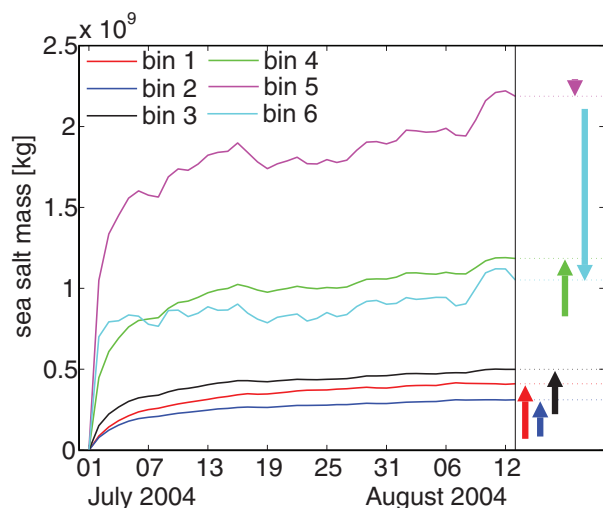


FIG. 6. Sea salt mass in each of the bins in the 6-bin model simulation. Size intervals were derived using the iso-gradient division procedure. Arrows on the right hand side denote relative mass changes at the end of the simulation as compared to the relative mass proportions derived from the emission function.

15 size bins generate very similar sea salt mass and the differences between them can be considered negligible. In this paper the simulation with 15 size bins is selected as reference for various studies.

Figure 6 presents the sea salt mass contributions from each of the size intervals in the 6-bin simulation. The highest contribution, constituting about 40% of the total mass, is modeled in the 5th bin. Somewhat surprisingly the mass in bin 6 is more than twice lower even though there are similar emissions (5.55×10^{12} and 5.11×10^{12} [kg / m²s], Table 2) in both bins. This is caused by stronger deposition processes affecting the larger particles in the sixth bin.

To assess the relative mass change in each size bin during the simulation versus the mass ratios at emission, vertical arrows are drawn on the right hand side of Figure 6. The model output at the last day of the simulation is analyzed. The mass values are extended with dotted lines. In Figure 6 the origin of each arrow is placed at the level determined by the total modeled mass multiplied by a fraction of emission within a bin derived from the emission function. Therefore, they are at the positions which would be observed if all particles were deposited at a constant rate. These positions are proportional to the emission ratios for each of the bins. The arrowheads point to the actual modeled mass values at the end of the simulation. A facing-up arrow indicates a relative mass accumulation in a bin, whereas a downward pointing arrow is interpreted as a relative loss of mass in a bin, caused by deposition excess over emission.

As can be seen in Figure 6, in the first four size bins relative mass accumulation is observed. In the 6th bin, on the other hand, a large relative mass decrease is simulated. The aerosol mass buildup is expected in the case of small particles for which

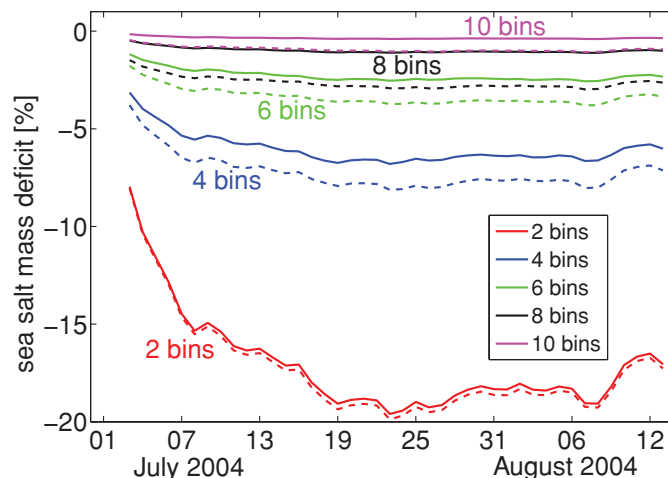


FIG. 7. Mass deficit (in percent) obtained from simulations with various numbers of size bins. The reference simulation has 15 size intervals. Simulations with the iso-gradient divisions are plotted as solid lines; the iso-log divisions are plotted as dashed lines.

the deposition processes are weak. This buildup can result in a situation where a size bin with less efficient mass emission gains in time more mass than a bin where emission is stronger, but deposition plays a more dominant role. Bins 4 and 6 are examples of such situations, where at some point the mass in bin 4 surpasses that in bin 6. Other examples are bins 1 and 2. Even though less mass is emitted in bin 1, the simulated values quickly overtake the mass modeled in bin 2. Again, less efficient deposition processes allow for more mass buildup in bin 1.

Aerosol accumulation in a bin results in a smooth mass change within that bin, which is only slightly affected by temporal variations in global emissions. For example (Figure 5), the mass changes in bins 1, 2, and 3 show almost negligible temporal variability despite substantial spikes in the total mass. Temporal variations are due to larger particles which have a shorter lifetime.

Figure 7 presents relative differences between the total mass for different simulations and the mass obtained with 15 size bins. The iso-gradient and iso-log cases are represented by solid and dashed lines, respectively. The reference 15-bin simulation has iso-gradient divisions. In all cases a mass deficit is observed; there is up to 20% less mass when 2 size bins are used instead of 15. The higher number of size bins, the better accuracy of the simulations - the mass deficit is smaller than 1% in the case of 10 size intervals. Figure 7 shows that simulations with the iso-gradient division method are more accurate than simulations with the iso-log divisions. For instance, 8 iso-gradient size bins are able to reproduce the accuracy obtained with 10 iso-log intervals.

The reasons behind the mass increase with an increasing number of size bins (Figure 5) require further explanation. What processes are responsible for the fact that a finer resolution generates more, not less, sea salt mass? To address these questions

we constructed a simple conceptual model of mass deposition caused by the dry processes.

In this 0-dimensional model, we assume that the mass reduction in a bin is only a function of the deposition velocity multiplied by the emission value:

$$M_{dep,i} = F_i \bar{v}_{d,i}. \quad [31]$$

Here F_i is given by Equation (17) and represents the sea salt mass emitted within a size bin; $\bar{v}_{d,i}$ is the characteristic velocity at which the mass F_i is deposited, and index i denotes a given size bin. The characteristic deposition velocity $\bar{v}_{d,i}$ is strictly related to the equivalent radius of particles assigned to a bin, $\bar{r}_{eqv}$, as derived from Equation (18). Moreover, since most of the deposition occurs over oceans, the effective radius at 80% RH is provided as a parameter to the deposition velocity $\bar{v}_{d,i}$. According to the sea salt growth function, $\bar{r}_{eqv,80} = 2\bar{r}_{eqv}$.

The characteristic deposition velocity $\bar{v}_{d,i}(\bar{r}_{eqv,80})$ is then obtained with the use of Equation (2). In order to derive the total mass deposition, the following summation over all ' n ' size bins has to be evaluated:

$$M_{dep} = \sum_{i=1,n} F_i \bar{v}_{d,i}(\bar{r}_{eqv,80}). \quad [32]$$

For a given emission and dry deposition functions, M_{dep} depends only on the value of n , that is the number of size intervals at which the initial size range is divided. M_{dep} can also be derived in a similar way to that in Equation (32), using continuous forms of emission and deposition functions:

$$M_{dep,a} = \frac{4}{3} \pi \rho_{ss} \int_{r_1}^{r_2} \frac{1}{\xi_{dry}} \frac{dF}{dr_{80}} r^3 \bar{v}_d(r) dr, \quad [33]$$

where $\rho_{ss} = 2.2 \times 10^6 [\text{g}/\text{m}^3]$ is the dry sea salt density, dF/dr_{80} is the Monahan's emission function, and $\xi_{dry} = 0.5$ is the growth function for dry particles.

In Figure 8, the solid line represents the relative difference, with the minus sign, between $M_{dep}(n)$ and the analytically derived $M_{dep,a}$. The dashed and dotted lines on the same figure represent relative mass deficits on 13 August, 2004, obtained with various numbers of size bins. The simulation with 15 size intervals was used as reference. The results of the conceptual model (solid line) indicate that the more size bins are used, the less mass is lost in the model. In other words, a coarser resolution in representing the deposition processes leads to deposition which is too high, or excess of deposition in the model. This relation can be explained by comparing emitted mass F_i and equivalent radius of particles in a bin r_{eqv} for various number of size intervals employed (Table 2). When 2 size bins are used the majority of mass is assigned to the radius of $2.207 \mu\text{m}$. By introducing additional bins, more mass is allocated to particles with smaller radii. The smaller radius indicates, according to

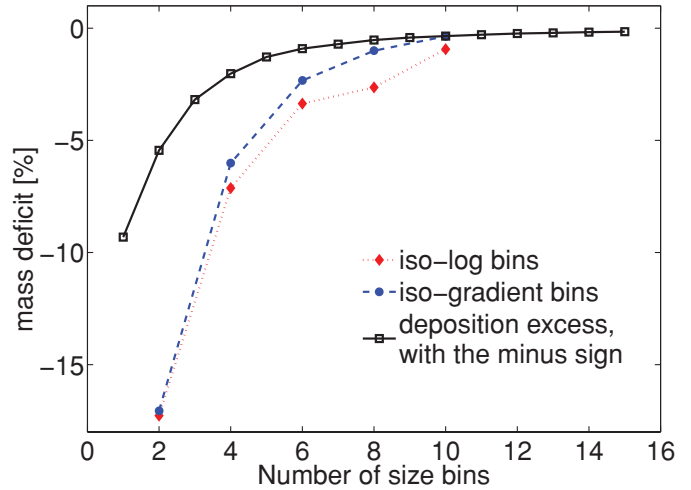


FIG. 8. Relative mass deficit (in percents) on the last day of the simulations as a function of the number of applied size intervals. The dashed and dotted lines represent simulated mass deficit on 13 August, 2004, obtained with the iso-log and iso-gradient size intervals, respectively. The solid line is the conceptually derived mass deposition excess, with the minus sign, computed according to the Equation (32).

Figure 1, the lower dry deposition velocity and lower sea salt mass deposition in the model. In the case of a one size interval, almost 10% more mass is deposited, as compared to the $M_{dep,a}$ value. The results of the conceptual mass deposition model provide a compelling explanation for the total mass increase with the rising number of size bins observed in Figures 5 and 7.

The mass deficit simulated with the global model, as can be observed in Figure 8, is almost three times higher than the deposition excess obtained with the conceptual deposition model. This is mainly due to the mass accumulation in the model, which also leads to the increase in mass differences between various simulations. This is evident in Figure 7 where the mass deficits increase at the beginning, and reach some steady values as the simulations progress. The variability of deposition velocities in the global model due to different conditions, as compared to the 0-dimensional model, can also contribute to the derived differences. However, as the number of size bins increases, global simulations converge to the conceptually derived errors. The simulations with 10 size bins already provide sufficient accuracy, with the error being smaller than 0.5% when the iso-gradient bin division procedure is applied.

3.2. Average Sea Salt Optical Depth Analysis

The NAAPS simulations performed with various numbers of size intervals and the two alternative size division methods were further analyzed with the use of the optical code described in Section 2.4. The global average sea salt optical thickness (AOT) at 550 nm wavelength was investigated.

In Figure 9, the computed AOT is presented with each line representing a simulation with a different number of size bins.

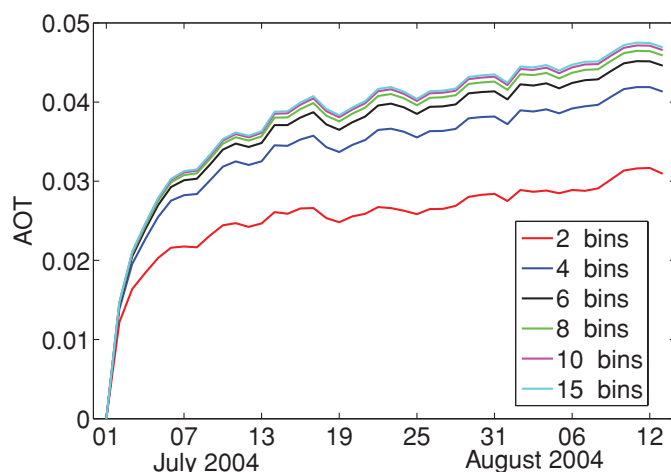


FIG. 9. Trends of computed average AOT at 550 nm wavelength as derived from the NAAPS simulations with various number of size bins. Size intervals are derived using the iso-gradient division procedure.

As it can be seen, the average sea salt AOT reaches about 0.05 for most detailed simulations. Also, a converging trend, similar to the one presented in Figure 5, is observed. An important difference between the trends in total mass (Figure 5) and average AOT (Figure 9) is that the AOT exhibits less temporal variability. For instance, the over 10% mass increase between 6–13 August corresponds to the less than 5% AOT increase during the same period of time. This is related to the relative impact of fractional AOT from individual bins.

Figure 10 illustrates the relative AOT contribution from each bin in case of the 6-bin model simulation. About one-third of the total AOT is generated by the smallest particles (bin 1). The particles with dry radius smaller than 1.1 micron constitute about

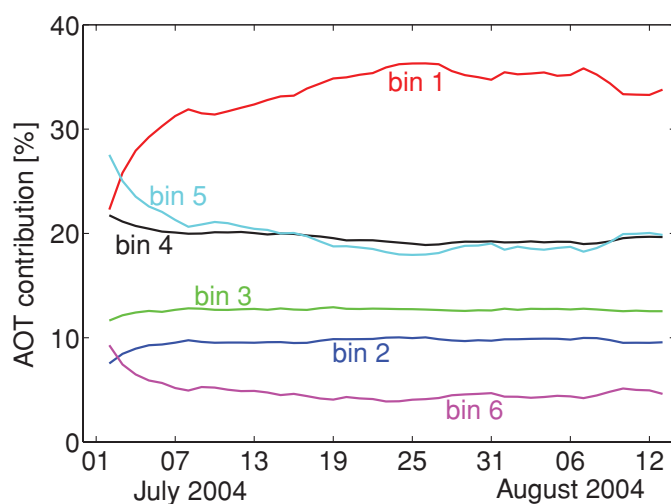


FIG. 10. Trends of AOT contribution, in percent, from different bins in the 6-bin model simulation. Size intervals are derived using the iso-gradient division procedure.

3/4 of the modeled AOT. This large contribution of submicron particles explains the relatively smooth temporal trends of the total AOT: the sea salt mass modeled in these bins, due to mass accumulation, exhibits weak temporal variability (Figure 6).

The impact of bin 6 on the total AOT is of the order of 5% despite a relatively wide size range of particles included in that bin. On the other hand, bin 5 contributes about 20% of the total AOT even though the mass emission in both bins is of similar magnitude (Table 2). The AOT difference in bins 5 and 6 can be understood as follows. The mass simulated in bin 5, due to less efficient deposition, is more than twice higher than in bin 6 (Figure 6). This mass difference results in a similar spread in the number of particles, given a comparable weight of virtual particles in both bins (1.46×10^{-12} and 1.34×10^{-12} g). Also, the virtual particle extinction cross-section in bin 5 is more than twice higher than in bin 6 (Table 2). These two factors result in a 4-fold difference in the total AOT contribution of bins 5 and 6.

Figure 11 illustrates the relative differences between the AOT obtained with various size intervals versus the simulation with 15 size bins. Simulations with the iso-gradient and iso-log divisions are presented as solid and dashed lines, respectively. Figure 11 shows that the simulations with 2 size intervals underestimate the average AOT by more than 35% with respect to the reference run. When the AOT averaging is performed only over oceans (results not shown), this difference reaches 50%. A comparison with Figure 7 indicates that the AOT differences are larger than the corresponding total mass differences. Such AOT underestimations can substantially influence radiative forcing estimates and assessment of the planetary albedo.

Figure 11 illustrates that the proposed iso-gradient division scheme offers some advantage for 6 and more bins. A lower

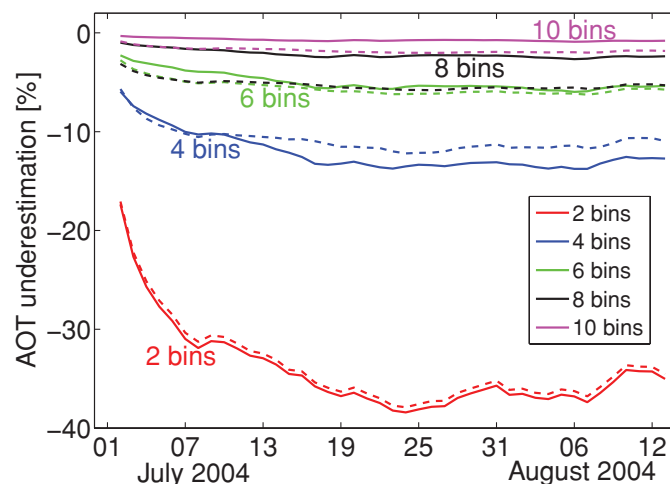


FIG. 11. Average AOT relative difference, in percent, obtained from simulations with various numbers of size bins. The reference simulation has 15 size intervals. The solid lines represent model runs where the iso-gradient size division procedure was applied, whereas the dashed lines represent the corresponding iso-log simulations.

division resolution results in slightly smaller errors for the iso-log simulations. This is because smaller particles, which have dominant impact on AOT, are better represented by the iso-log intervals in the case of a limited number of size bins (Figure 4). For finer divisions, the iso-gradient size division leads to more accurate results, analogically to the total mass case. Also, it should be noted that the simulated AOT exhibits more regular convergence with respect to the number of size bins in the case of the iso-gradient division method. The convergence of the iso-log simulations is less stable, which is expressed by a similar 5% underestimation in both the 6 and 8 division simulations. Performed simulations and analysis show that the iso-gradient size division procedure has advantages over the iso-log scheme.

4. CONCLUSIONS AND SUMMARY

Numerical simulation of atmospheric aerosols has become widespread in a number of geophysical applications, from studying atmospheric chemistry, cloud physics, atmospheric radiation, remote sensing to nutrient transport and air quality control issues. It is indispensable for assessing the aerosol's influence on climate. Many scientific efforts have been undertaken to better understand the aerosol's role in the Earth's atmosphere and to improve aerosol simulation capabilities on a regional and global scale. However, there are still many uncertainties regarding the representation of aerosols in numerical models, in particular the description of emission functions, chemical transformations, and deposition mechanisms. Proper depiction of these processes is difficult because they depend highly on aerosol size, which can vary by several orders of magnitude. Moreover, aerosol size distribution is often poorly represented in numerical models because of computational cost limitations. All these factors result in model errors that affect aerosol simulations and introduce uncertainties in our understanding of aerosols' role on the Earth's energy budget.

There is still large spread in the accuracy of SSA size representation in global aerosol transport models. As an example, a recent model intercomparison framework AeroCom assembles simulations from 16 global aerosol models (Textor et al. 2006). 9 of these models employ a sectional approach: 5 of them use between 1 and 4 size bins, and the remaining 4 models employ 8 to 12 bins to represent a SSA size distribution. Some other sea salt studies use more size intervals, but this often correlates with a coarser horizontal resolution. As an example, Pierce and Adams (2006) use 30 bins and $4^\circ \times 5^\circ$ degree horizontal resolution and Grini et al. (2002) use 16 bins at 5.625° resolution. Comparison of models that use only a few and over 10 size intervals might be hampered by additional numerical uncertainties related to the accuracy of size distribution representation in these models.

In this study the importance of aerosol size representation in a global transport model is investigated. The global SSA simulations are analyzed in terms of the total sea salt mass and

average SSA optical thickness. The numerical experiments were performed with two different size-bin division procedures and with various number of aerosol size bins. The results indicate there is a large dependence of the total mass and average AOT on the number of size bins used to represent the aerosol size distribution. The total mass is underestimated by 20% if 2 instead of 15 (reference) size intervals are used. A simple conceptual mass deposition scheme is constructed to explain the observed differences. It is found that the underestimation of SSA mass in the model is related to the excessive aerosol dry deposition in situations when the size distribution is coarsely resolved. The comparison of simulations with the iso-log and iso-gradient division procedures shows that the iso-gradient method produces more accurate results if more than 2 size intervals are used. Up to 2 fewer iso-gradient size bins are needed to achieve the same accuracy as with the iso-log intervals, which is beneficial for simulation efficiency.

The average AOT underestimation is higher and reaches over 35% between the simulations with 2 and 15 size bins. The iso-gradient simulations are more accurate than the iso-log ones if more than 6 size intervals are used. Such high, 35% AOT differences can have substantial impact on simulating the global mean SSA radiative forcing. Witek (2008) showed, using the simulations presented in this study and employing a highly simplified top of the atmosphere radiative forcing model, that the forcing deficit can reach up to 33% if 2 instead of 15 iso-gradient size intervals are used. It is very likely that the simulated differences in average AOT would foster similar or still substantial uncertainties in forcing calculations even if a more sophisticated radiative transfer code is used.

Finally, numerical experiments conducted in this study are not only relevant to the sea salt aerosol. Similar findings could be readily generalized to other aerosol types (Menut et al. 2007) and also other physical processes regulating aerosol transformation and deposition. For instance, size-dependant wet deposition mechanisms, if they were more variable than dry deposition, could be used instead to derive iso-gradient size intervals. Wet deposition, however, remains uncertain and its actual dependence on particle's size still needs to be studied. Another mechanism, alternative to particles removal by dry or wet deposition, would be aerosol growth and transformation due to microphysics, and corresponding shift of particles from one size class to another. Also, the deposition velocity function used in the present study could be easily substituted with some other size-dependent function, better fitted to deposition variability of other aerosol types. The general approach should be to determine a leading and most variable physical process that controls aerosol transformation or removal and construct size intervals based on the efficiency of this processes at different particle sizes. This iso-gradient procedure leads to more accurate results and can, without much effort, reduce computational cost in aerosol transport models.

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