Modeling sea-salt aerosols in the atmosphere 1. Model development

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Abstract. A simulation of the processes of sea-salt aerosol generation, diffusive transport, transformation, and removal as a function of particle size is incorporated into a onedimensional version of the Canadian general climate model (GCMII). This model was then run in the North Atlantic between Iceland and Ireland during the period of January-March. Model predictions are compared to observations of sea-salt aerosols selected from a review of available studies that were subjected to strict screening criteria to ensure their representativeness. The number and mass size distribution and the wind dependency of total sea-salt aerosol mass concentrations predicted by the model compare well with observations. The modeled dependence of sea-salt aerosol concentration in the surface layer $(\chi, \mu g m^{-3})$ on 10-m wind speed $(U_{10}, m s^{-1})$ is given by $\chi = be^{aU_{10}}$. Simulations show that both a and b change with location. The value a and b range from 0.20 and 3.1 for Mace Head, Ireland to 0.26, and 1.4 for Heimaey, Iceland. The dependence of χ on surface wind speed is weaker for smaller particles and for particles at higher altitudes. The residence time of sea-salt aerosols in the first atmospheric layer (0-166 m) ranges from 30 min for large particles ($r = 4-8 \ \mu m$) to ~60 hours for small particles ($r = 0.13-0.25 \ \mu m$). Although some refinements are required for the model, it forms the basis for comparing the simulations with long-term atmospheric sea-salt measurements made at marine baseline observatories around the world and for a more comprehensive three-dimensional modeling of atmospheric sea-salt aerosols.

1. Introduction

Sea-salt aerosols play a very important role in a variety of processes in the atmosphere. They influence radiative transfer directly by scattering solar radiation and indirectly by altering cloud droplet size distribution and concentration, thereby influencing the albedo of marine boundary layer clouds. In addition, sea-salt aerosol particles are chemical carriers of species containing Cl, Br, I, and S and therefore play a role in the atmospheric cycles of these important elements. The halogens Br and Cl, once mobilized by heterogeneous reactions from sea-salt inorganic forms to reactive gaseous forms (e.g., Br₂, Cl₂) [e.g., *Mozurkiewcz*, 1995], can play a role in atmospheric ozone depletion and destruction of light hydrocarbons [*Jobson et al.*, 1994].

Numerous experimental investigations of sea-salt aerosols have been conducted around the globe in order to determine their abundance and physical/chemical properties [Jacobs, 1937; Woodcock, 1953; Toba, 1965a, b; Lovett, 1978; Prospero, 1979; Podzimek, 1980; Parungo et al., 1986; Marks, 1990; Ikegami et al., 1994; McGovern et al., 1994]. Sea-salt aerosol concentration is a strong function of the state of the sea surface, which is in turn determined by meteorological conditions, especially surface wind speed. Sea-salt particles are distributed from 0.02 to 60 μ m with a bimodal size distribution in the submicron portion [Fitzgerald, 1991]. Since the observations

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can only be performed at certain times and locations, application of the experimental results is limited. It is vital to develop an aerosol transport model to predict its concentration and size distribution on a regional scale as a function of atmospheric processes. Various models have been proposed in the literature [*Gathman*, 1982; *Fairall and Davidson*, 1986; *Erickson and Duce*, 1988; *Fitzgerald*, 1992; *van Eijk et al.*, 1992]. A summary of major functions and assumptions in these models is shown in Table 1. All of these models are limited to the marine boundary layer and do not address the long-range transport of sea salt.

A comprehensive sea-salt aerosol model coupled with a onedimensional climate model (FIZ-C) [*Therrien*, 1993] is presented in this paper. Using the meteorological conditions generated by the FIZ-C, the model includes the following processes: (1) sea-salt generation due to surface wind; (2) vertical transport by turbulence and convection; (3) dry deposition and gravitational settling; and (4) wet removal processes which include both in-cloud and below-cloud scavenging. The model does not at this time include chemical or physical transformation through interactions with other aerosol types such as acidic sulphur. The particle size distribution is modeled by representing the size spectrum as a series of discrete size bins.

2. Physical Model

The fate of sea-salt aerosols, once they are injected into the atmosphere from the ocean source, is governed by a series of physical processes such as transport, coagulation, dry and wet removal, and chemical transformation. Transport of aerosols is

			Size			Ren	noval
Authors	Dimension	Domain	Spectrum	Transport	Source	Dry	Wet
Gathman [1982]	one	MBL	no	eddy	Blanchard [1963]	D*	BC*
Fairall and Davidson [1986]	one	MBL	yes	eddy	Fairall and Davidson [1983]	G† D*	IC* BC†
Burk [1986]	one	MBL	no	eddy	Monahan et al. [1986]	D†	BC*
Stramska [1987]	one	MSL	yes	eddy	Monahan et al. [1986]	D*	BC*
Erickson and Duce [1988]	two	surface	no	eddy	empirical	D† G†	BC†
Fitzgerald [1992]	zero	MBL	yes	eddy	Monahan et al. [1986]	D* C*	BC†
van Eijk et al. [1992]	zero	MBL	yes	eddy	empirical	D* G*	BC* IC*

Table 1. Summary of Sea-Salt Aerosol Models

MBL, marine boundary layer; D, dry deposition; G, gravitational fall; BC, below-cloud scavenging; IC, in-cloud scavenging. *Not included.

†Included.

realized by turbulent diffusion, advection, and vertical convection of the atmosphere, which is represented in this model by the FIZ-C parameterizations which are analogous to those for moisture transport in the FIZ-C. A generalized prognostic mass balance equation for size i of type j aerosol particle can be written as

$$\frac{\partial \rho \chi_{ij}}{\partial t} + d\iota v \rho \chi_{ij} U = S_{ij} + I_{ij}$$
(1)

where χ_{ij} is the mass mixing ratio of the *i*th size range aerosols (kg/kg_{air}), U is the horizontal wind velocity vector (m s⁻¹), ρ is the air density (kg m⁻³), and S_{ij} is the source and sink terms which may include following contributions: (1) surface source (natural and anthropogenic); (2) clear air processes (particle nucleation, particle coagulation, and chemical transformation); (3) in-cloud processes (activation of aerosols, attachment to clouds, and removal of aerosol-attached clouds by hydrometeors); (4) dry deposition; and (5) precipitation scavenging, where particle nucleation, and chemical transformation are processes not required for sea-salt aerosols. Term I_{μ} represents the rate of intersectional transfer process which moves the aerosol mass from one size bin to another. The formation of new aerosol particles through coagulation or breaking of the existing particles is one intersectional transfer process. Since only dry mass of aerosols is entered into the mass balance equation (1), the condensation/evaporation of water on sea-salt aerosol particles will not contribute to the intersectional transfer rate. However, condensation/evaporation may contribute to intersectional transfer rate of other types of aerosols, for example, sulphate aerosols when sulphuric acid vapor condenses on the existing aerosols.

In accordance with the FIZ-C convention, the prognostic equation is rewritten in the form of

$$\frac{\partial \chi_{y}}{\partial t} = \frac{\partial \chi_{y}}{\partial t} \Big|_{\text{dynamics}} + \frac{\partial \chi_{y}}{\partial t} \Big|_{\text{surface}} + \frac{\partial \chi_{y}}{\partial t} \Big|_{\text{clear air}} + \frac{\partial \chi_{y}}{\partial t} \Big|_{\text{dry}} + \frac{\partial \chi_{y}}{\partial t} \Big|_{\text{m-cloud}} + \frac{\partial \chi_{y}}{\partial t} \Big|_{\text{below-clouds}}$$
(2)

In (2), the aerosol concentration change has been divided into tendencies for dynamics, surface, clear air, dry deposition, incloud and below-cloud processes. The dynamics includes resolved motion as well as subgrid turbulent diffusion and convection. The surface processes include surface emission rate of both natural and anthropogenic aerosols and serve as boundary conditions for the model. Particle nucleation, coagulation, and chemical transformation are included in clear-air process. In the current version, coagulation is not included for sea-salt aerosols. However, for purposes of predicting total mass of sea-salt aerosols for comparison with observations, this is not a major drawback, since Na and Cl which dominate sea-salt aerosol mass are not affected substantially by coagulation. The aerosol model is structured in such a way that any process which affects the concentration and size distribution of sea-salt aerosols can be modified or added to the model as new or better parameterizations for such processes become available. Some detailed physical parameterizations for sea-salt aerosols will be presented in the following sections.

2.1. Aerosol Growth With Relative Humidity

Experimental evidence [*Fitzgerald*, 1991] has shown that there exists a size distribution of sea-salt aerosols of radii ranging from 0.02 to 60 μ m. A knowledge of size distribution is necessary in order to calculate the effect of the aerosol on climate as well as on chemical reactions in the atmosphere.

In this model, the aerosol size distribution is simulated by a method similar to that developed by *Gelbard and Seinfeld* [1980]. The whole size range of aerosol particles is divided into a number of sections (or size bins), and each size section is represented by one prognostic equation (i) which is a mass continuity equation averaged for that size range. The calculation of sea-salt generation is carried out by integrating the production over each size bin while the physical properties such as dry deposition and wet removal rates are calculated by using the average size in each size bin. It is noted that in the model, the prognostic equation is written for dry size only. Being very hygroscopic, sea-salt aerosols will change size as the ambient relative humidity changes. By assuming that the par-

ticles are always in stable equilibrium with the surrounding relative humidity, the actual size of sea-salt aerosols is related to their dry size by the empirical relationship of *Gerber* [1985]:

$$r = \left[\frac{C_1 r_d^{C_2}}{C_3 r_d^{C_4} - \log S} + r_d^3\right]^{1/3}$$
(3)

where r_d is the dry particle radius (centimeters), S the saturation ratio (or relative humidity), and C_1 , C_2 , C_3 , and C_4 are parameters for different types of the aerosol particles (Table 2). Equation (3) can be used in a relative humidity ranging from 0% to 100%.

Physical tendencies, for example, coagulation, dry deposition, gravitational settling, or wet removal, are computed for each dry size bin using the aerosol size in equilibrium with ambient water vapor. The relative humidity profile is calculated from temperature and moisture predicted by the climate model.

As the particles grow with relative humidity, the density of the particles is also altered by incorporating more water. The average density of sea-salt aerosols in each size bin is calculated based on the size difference between the dry and wet particles. The radius of the particles in equilibrium with ambient moisture is estimated by (3).

2.2. Generation of Sea-Salt Aerosols

The generation of sea-salt aerosols has been attributed to various physical processes [Blanchard, 1983]. The most prominent mechanism is believed to be the entrained air bubbles bursting during whitecap formations due to the surface wind [Monahan et al., 1986]. Precipitation (both rain and snow) contributes to the production but only on an intermittent and local scale. A summary of various mechanisms are shown in Figure 1.

Physically, the production of sea-salt aerosols by wind is proportional to the whitecap coverage. Continuous supply of excessive energy by the wind to the sea surface results in wave breaking. Wu [1986] developed a relationship between the whitecap coverage and wind speed by considering that the energy lost by wave breaking is balanced by the energy gained from the wind:

$$W \propto \dot{E} = \tau V \sim \tau u_* = (C_{10} U_{10}^2) (C_{10}^{1/2} U_{10}) \sim U_{10}^{375}$$
(4)

where W is the fraction of the sea surface with whitecap coverage, \dot{E} is the energy supply rate by the wind to a unit area of the sea surface, τ is the wind stress, V is the sea surface drift current, u_* is the wind-friction velocity, U_{10} is the wind speed at 10-m level, and C_{10} is the wind stress coefficient which is proportional to $U_{10}^{1/2}$ [Wu, 1969]. On the basis of shipboard photographic observations of sea surface [Monahan, 1971;

 Table 2.
 Constants for Growth Equation (3)

Aerosol Model	C_1	C_2	<i>C</i> ₃	C_4
Sea salt	0.7674	3.079	2.573×10^{-11}	-1.424
Rural	0.3926	3.101	4.190×10^{-11} 5.415×10^{-11}	-1.404 -1.399
$(NH_4)_2SO_4$	0.4809	3.082	3.110×10^{-11}	-1.428

Reproduced from Table 1 of *Gerber* [1985]. The temperature dependence of (3) is found primarily in C_3 due to the sensitivity of the Kelvin effect to temperature; C_3 can be temperature corrected with the expression: $C_3(T) = C_3[1 + 0.004(298 - T)]$, T in kelvin.



Figure 1. Mechanisms for sea-salt aerosol generations (adapted from *Monahan et al.* [1986]). Two mechanisms are presented (a) indirect production by bubbles and (b) direct production by spumes.

Toba and Chean, 1973] and wave tank measurements of the sea-salt aerosol production rate per unit area of whitecaps, *Monahan et al.* [1986] have substantially refined the formulation for the sea-salt aerosol generation and included both indirect (bubbles) and direct (spume) mechanisms in the model. The density function dF/dr (particles $m^{-2} s^{-1} \mu m^{-1}$), which expresses the rate of sea-salt droplet generation per unit area of sea surface, per increment of droplet radius, is given (1) for indirect mechanism (through bubbles):

$$\frac{dF_0}{dr} = 1.373 U_{10}^{3\,41} r^{-3} (1 + 0.057 r^{1\,05}) \times 10^{1\,19e^{-R}}$$
(5a)

where $B = (0.380 - \log r)/0.650$ and (2) for direct mechanism (through spume):

$$\frac{dF_1}{dr} = \begin{cases} 0 & r < 10 \ \mu\text{m} \\ 8.60 \times 10^{-6} e^{2 \ 08U_{10}} r^{-2} & 10 \ \mu\text{m} \le r \le 75 \ \mu\text{m} \\ 4.83 \times 10^{-2} e^{2 \ 08U_{10}} r^{-4} & 75 \ \mu\text{m} \le r \le 100 \ \mu\text{m} \\ 8.60 \times 10^{6} e^{2 \ 08U_{10}} r^{-8} & r \ge 100 \ \mu\text{m} \end{cases}$$
(5b)

The total generation rate of sea-salt aerosols via whitecaps and breaking waves is then

$$\frac{dF}{dr} = \frac{dF_1}{dr} + \frac{dF_0}{dr} \tag{6}$$

A total flux of sea-salt aerosols for each size bin is obtained by integrating this formula over the size range of this bin. This sea-salt generation model is of essential importance in setting up the lower boundary conditions for our sea-salt aerosol model. It is noted that the applicable range of radius for (5a) is $0.8-10 \ \mu m$ (at 80% relative humidity (RH)) within which the empirical relationships are based. It may introduce some uncertainties if it is used for the generation of submicron sea-salt aerosols. For particles larger than 10 $\ \mu m$, (5b) should be used with (5a). However, it generates too much big sea-salt particles at high wind speeds compared to observations and is therefore neglected in the simulation.

Another mechanism of marine aerosol generation, which has not received much attention during the last 3 decades, is the production of sea-salt aerosols by wet precipitation. This mechanism is mentioned here for two reasons. First, since it is well recognized that a major self-cleaning process of the atmosphere is the removal of particles by clouds and precipitation, any attempt to model aerosol particles in the marine atmosphere without incorporating wet removal mechanism is not realistic. Second, in the case of wet precipitation, if only the removal process is considered, which is the case for all the present marine aerosol models, aerosol concentrations may be underestimated, especially near the sea surface.

Unfortunately, literature on this subject is very limited. Only two papers have been found that deal exclusively with it [Blanchard and Woodcock, 1957; Marks, 1990]. In the first paper, Blanchard and Woodcock [1957] suggest two mechanisms of marine aerosol generation by raindrops: (1) direct production through the impact of the rain drops and (2) from bubbles produced in the surface water as a result of impact. This process occurs when a rain drop hits the sea surface, it generates not only sea-salt aerosols and bubbles but also secondary drops due to the splash. The secondary drops fall back into the sea and generate more bubbles. It is believed that in both cases the production of bubbles is a function of droplet size [Blanchard and Woodcock, 1957]. The generation of sea-salt aerosol particles follow the bursting of bubbles.

Marks [1990] carried out an experimental investigation of the marine aerosols under both dry and wet (rain) conditions from the Dutch research platform Noordwijk, 9 km offshore in the North Sea. Total sea-salt concentrations were measured at three heights of 4.5, 12, and 18.3 m and sea-salt size distributions at 12-m elevation. Compared to dry conditions, a reduction in the aerosol concentration at 4.5 m by rain was found at wind speeds of 8 and 10 m s⁻¹. In other words, the net effect of rain on the total aerosol concentrations at these wind speeds is negative, an indication that the wet removal process surpasses the generation of sea-salt aerosols by rain. When the wind speed at 4.5 m reached 15 m s⁻¹, the sea-salt aerosol concentration during rainy weather became greater than under dry conditions. This means rain generates more sea-salt aerosols than it scavenges provided that wind-mediated sea-salt production remains unchanged during dry or wet conditions. One interesting point from the experiment was that for 12- and 18.3-m elevations at a wind speed of approximately 15 m s⁻¹, the sea-salt aerosol concentrations were not influenced by rain, meaning that a balance between production and washout of sea-salt aerosols by rain occurred under this condition. If this is a true characteristic of the marine boundary layer sea-salt aerosols, it might be used to parameterize processes of rainmediated generation of sea-salt aerosol. In this respect, wet scavenging mechanism which have been studied extensively [Wang et al., 1978; Herbert and Beheng, 1986] would play an important role.

Since the rain intensity was not reported in the paper, a quantitative relationship between the rain-mediated production and scavenging and wind speed can not be inferred from the paper. Therefore this mechanism of sea-salt generation is neglected in our model. One justification for this would be that in comparison to wind-generated sea-salt aerosols, this source is likely a minor one, especially considering the frequency of the precipitation.

2.3. Wet Removal of the Marine Aerosols

For modeling atmospheric aerosols, the inclusion of wet removal process, that is, the scavenging of particles in the air by precipitation and by clouds, is essential. This self-cleaning mechanism maintains the balance between the sources and sinks of atmospheric aerosol particles. In the present model, both below-cloud and in-cloud scavenging processes are considered with a simplified approach suitable for use in a largescale transport model.

2.3.1. Below-cloud scavenging. Below-cloud scavenging as referred to here is the process of aerosol removal from the atmosphere between cloud-base and the ground by precipitation (i.e., rain or snow). The capture of aerosol particles by falling hydrometeors takes place by Brownian and turbulent shear diffusion, inertial impaction, diffusiophoresis, thermophoresis, and electrical effects. Detailed studies of these processes [*Greenfield*, 1957; *Pilat*, 1975; *Wang et al.*, 1978; *Slinn*, 1984; *Herbert and Beheng*, 1986] have revealed that there exists a minimum in the collection efficiency of aerosol particles between sizes ranging from 0.5 to 1 μ m radius, which is sometimes called the "Greenfield gap." According to *Slinn* [1984], the removal rate of aerosols per unit volume can be written as

$$L(r_i) = \psi(r_i)\chi(r_i) \tag{7}$$

where r_i is the averaged actual radius of *i*th size bin and ψ is the scavenging rate which is computed by integration of the following equation over all hydrometeor sizes:

$$\psi(r_{i}) = \int_{0}^{\infty} E(r_{i}, a) A_{x} [V_{i}(a) - V_{g}(r_{i})] N(a) \ da$$

where a is the drop radius, V_t and V_g are the gravitational settling velocity of hydrometeors and particles, $E(r_t, a)$ is the collection efficiency of aerosols of size r_t by hydrometeors of size a, and N(a) and A_x are the hydrometeor number density and effective cross-sectional area. However, the requirement of N(a) and large computer time for the integration makes it unrealistic to use in a large-scale aerosol model. An approximate expression [Slinn, 1984] for the rain scavenging rate is used:

$$\psi(r_i) = \frac{cp\bar{E}(r_i, R_m)}{R_m} \tag{8}$$

where c is a numerical factor of order unity (0.5) and the mean collision efficiency $\bar{E}(r_i, R_m)$ is evaluated by using a mean drop size. The precipitation rate p is also used to obtain the mean drop radius:

$$R_m = 0.35 \text{ mm} \left(\frac{p}{1 \text{ mm/h}}\right)^{1/4}$$
 (9)

For snow scavenging,

$$\psi(r_i) = \frac{\gamma p \bar{E}(r_i, \lambda)}{D_m} \tag{10}$$

where λ is the characteristic capture length, D_m is the characteristic length, and γ is a constant of order unity (0.6). Depending on temperature, λ and D_m may have different values [*Slinn*, 1984] because of different type of snows (Table 3).

2.3.2. In-cloud processes. In-cloud scavenging processes are reviewed by *Barrie* [1991]. Scavenging of chemical pollutants in clouds by three phases of water was discussed in that

paper. A three-process model was developed to describe the in-cloud processes of the aerosol particles. The first process is the activation of sea-salt aerosols, that is, served as cloud condensation nuclei (CCN), depending on the critical super-saturation and particle dry size. The mechanism of this process has been studied in detail by cloud physicists [e.g., *Twomey*, 1977] and is represented by the so-called Köhler curves. For sea salt (NaCl) and sulphate ((NH₄)₂SO₄), the relationship between the critical radius (r_c) of dry soluble particle and supersaturation (S_c in percent) is given approximately by [*Twomey*, 1977]

Sea salt

$$r_c = 1.16 \times 10^{-6} S_c^{-2/3}$$

Sulphate

$$r_c = 1.40 \times 10^{-6} S_c^{-2/3}$$

which simply describes the minimum dry radius which would enable nucleation to occur upon it at a supersaturation S_c . The activation process has two major impacts: (1) because of the larger size of CCN they will be subject to a different removal scheme for particles with size greater than the critical radius r_c and hence the size distribution spectrum of sea-salt aerosols will be modified and (2) a link is established between aerosols and their indirect effect on climate for the activation will alter the cloud number density, liquid water content, and possibly cloud albedo.

The second in-cloud process is the attachment of the aerosol particles in size range *i* to the existing cloud droplets at a rate α_{iT} . The attachment of aerosols to the cloud droplets is assumed to be by two processes: Brownian and turbulent diffusion; that is, $\alpha_{iT} = \alpha_{iB} + \alpha_{iTB}$. The contribution by Brownian diffusion to α_{iT} is estimated by [Dingle and Lee, 1973]

$$\alpha_{iB} = \frac{kT}{3\eta} \left(\frac{1 + al/r_i}{r_{cl}} + \frac{1 + al/r_{cl}}{r_i} \right) (r_i + r_{cl}) M_c \quad (11)$$

where r_i is the averaged aerosol particle radius at size bin *i* and r_{cl} is the cloud equivalent droplet radius; *a* is the Cunningham correction factor; *l* is the mean free path of air molecules; M_c is the number density of cloud droplets; *k* is Boltzmann's constant; *T* is the absolute temperature; and η is the air viscosity. The turbulent contribution [Levich, 1962] is approximated as

$$\alpha_{iTB} = 14.1(r_i + r_{cl})^3 \left(\frac{\varepsilon}{\nu}\right)^{1/2} M_c$$
 (12)

where ε is the rate of energy dissipation and ν is the kinematic viscosity. One approximation in formulating (11) and (12) is that the cloud size spectra is replaced by an equivalent droplet radius (r_{cl}) , which is estimated by the cloud scheme of the climate model.

The final process involves the removal of the aerosolcontaining cloud droplets produced by the first two processes by large falling hydrometeors at a rate β_i [Dingle and Lee, 1973; Slinn, 1984]. Assuming that an activation rate is Λ_i for size range *i* and that α_{iT} and β_i are constant within the integration interval, the in-cloud processes can be expressed by following ordinary differential equations:

 Table 3.
 Parameters for Snow Scavenging

Crystal Type		D_m , cm	
Graupel particles Rimed plates and stellar den Powder snow and spatial den Plane dendrites Needles	drites drites	$\begin{array}{c} 1.4 \times 10^{-2} \\ 2.7 \times 10^{-3} \\ 1.0 \times 10^{-3} \\ 3.8 \times 10^{-4} \\ 3.8 \times 10^{-3} \end{array}$	
Crystal Type	λ, μm	α	
sleet, graupel rimed crystals powder snow dendrites tissue paper camera film	1000 100 50 10 50 1000	2/3 2/3 2/3 1	

$$\frac{d\chi_{ia}}{dt} = -(\alpha_{iT} + \Lambda_{i})\chi_{ia} - \gamma_{ia}\chi_{ia}$$

$$\frac{d\chi_{ic}}{dt} = (\alpha_{iT} + \Lambda_{i})\chi_{ia} - \beta_{i}\chi_{ic}$$
(13)

where χ_{ia} is the interstitial concentration of the aerosol particles of *i*th size bin, χ_{ic} is the concentration of the aerosols in cloud water, and γ_{ia} is the rate of direct aerosol removal by large hydrometeors which can usually be ignored when compared to α_{iT} and β_i . The activation rate Λ_i is determined by the critical radius of aerosol particles. The total aerosol concentration, $\chi_{it} = \chi_{ia} + \chi_{ic}$, is obtained by solving (13) for χ_{it} subject to $\chi_{it}|_{t=0} = \chi_{i0}$.

$$\chi_{ii} = \frac{\chi_{i0}}{\beta_i - (\alpha_{iT} + \Lambda_i)} \left[\beta_i e^{-(\alpha_i T + \Lambda_i)t} - (\alpha_{iT} + \Lambda_i) e^{-\beta_i t} \right]$$
(14a)

In the case of nonactivation, that is, $r_i < r_c$, $\Lambda_i = 0$,

$$\chi_{\iota\iota} = \frac{\chi_{\iota 0}}{\beta_{\iota} - \alpha_{\iota T}} \left[\beta_{\iota} e^{-\alpha_{\iota} \tau} - \alpha_{\iota T} e^{-\beta_{\iota} t} \right]$$
(14b)

and for the activation case, that is, $r_i \ge r_c$, if we assume $\Lambda_i = \infty$,

$$\chi_{ii} = \chi_{i0} e^{-\beta_i i} \tag{14c}$$

In deriving (14c), an implicit assumption is made that once an aerosol particle is activated, all the particles with the same dry radius will form cloud droplets ($\Lambda_i = \infty$), and they will be removed from the atmosphere if there is precipitation. The removal rate by large hydrometeors, β_i , is calculated by setting $\beta_i = \psi(r_{cl})$ in (8). If no precipitation is experienced during the time step, the aerosol particle remains in the atmosphere and total concentration does not change.

2.3.3. Collection efficiency (E). In order to calculate the scavenging rate of aerosols or clouds by both rain and snow (equations (8) and (10)), the collection efficiency of aerosol particles by hydrometeors (E) is required. The collection efficiency is defined as the ratio of the total number of collisions occurring between droplets and particles to the total number of particles in an area equal to the droplet's effective cross-sectional area. According to previous investigations [Slinn and Hales, 1971; Pilat, 1975; Wang et al., 1978; Herbert and Beheng, 1986], E is a result of the combined action of Brownian diffusion, inertial impaction, interception, thermophoresis and diffusiophoresis, and electric forces. Detailed formulation of these processes in an atmospheric aerosol model is a prohibi-



 K_{α} - drift velocity

Figure 2. Schematic illustration of two-layer model used for dry deposition of aerosol particles on water surface. It is assumed a zero surface concentration and a bounce-off factor f_{BO} .

tively difficult undertaking. Neglecting phoresis and electric effects, *Slinn* [1984] derived the following semiempirical expression for the particle/drop collection efficiency:

$$E = \frac{4}{\text{ReSc}} \left[1 + 0.4 \text{Re}^{1/2} \text{Sc}^{1/3} + 0.16 \text{Re}^{1/2} \text{Sc}^{1/2} \right] + 4 \Re \left[\omega^{-1} + (1 + 2 \text{Re}^{1/2}) \Re \right] + \left[\frac{\text{St} - \text{S}_*}{\text{St} - \text{S}_* + 2/3} \right]^{3/2}$$
(15)

where $\text{Re} = R_m V_t / \nu$ is the Reynolds number of hydrometeors, Sc = ν/D and St = $[\tau(V_t - V_g)]/R_m$ are the Schmidt and Stokes numbers of particles. The coefficient τ is the characteristic relaxation time of the particles, D is the diffusion coefficient of particles, V_t and V_g are the terminal velocity of hydrometeors and particles, and R_m and r_t are the radius of hydrometeors and particles, respectively. $\mathcal{H} = r_t/R_m$, $\omega = \mu_w/\mu_a$, and μ_w and μ_a are the viscosity of water and hydrometeors.

$$S_* = \frac{12/10 + (1/12)Ln(1 + Re)}{1 + Ln(1 + Re)}$$

For snow scavenging, the efficiency is estimated as

$$E = \left(\frac{1}{\text{Sc}}\right)^{\alpha} + \left\{1 - \exp\left[-(1 + \text{Re}_{\lambda}^{1/2})\right]\frac{r_{i}^{2}}{\lambda^{2}}\right\} + \left[\frac{\text{St} - \text{S}_{*}}{\text{St} - \text{S}_{*} + 2/3}\right]^{3/2}$$
(16)

Depending on the type of snow crystals, α and λ values from Table 3 are used in (16).

2.4. Dry Deposition

In modeling particle dry deposition, a two-layer hypothesis is usually assumed [*Slinn and Slinn*, 1981; *Giorgi*, 1986]. Figure 2 shows a typical two-layer model for the water surface. The transport of particles in the top layer, that is, the surface layer extending between h and δ , is dominated by turbulence and gravitational settling, respectively. Viscous effects and the nature of the surface roughness are considered only in the interfacial layer which is from surface to height δ . Assuming steady state, the dry deposition flux of aerosol particles through the Earth-atmosphere is expressed by *Giorgi* [1986]:

$$\phi_h = \nu_{dep} n_h \tag{17}$$

where n_h is the particle concentration at height h and v_{dep} , the particle dry deposition velocity, is given by *Giorgi* [1986] as:

$$\nu_{dep} = K_{dep} + K_{dh}$$

where the definition of K_{dh} is given in Figure 2 with

$$K_{dep} = \frac{K_{SL}(K_{IL}f_{BO} + K_{d\delta} - K_{dh})}{K_{SL} + K_{IL}f_{BO} + K_{d\delta}}$$
(18)

where K_{SL} and K_{IL} are the transfer velocity in surface and interfacial layer, respectively, and are assumed to be of the following forms [*Giorgi*, 1986]:

$$K_{SL} = C_D U_h A_P \frac{C_{D0}^{1/2}}{C_{D0}^{1/2} - C_D^{1/2}}$$

$$K_{IL} = C_{D0}^{1/2} u_* G$$
(19)

where C_D and C_{D0} are the drag coefficients relative to h and δ , U_h is the wind speed at h. C_D is predicted in the climate model as the drag coefficient for the first layer above the surface and C_{D0} is calculated in the dry deposition module according to the surface type, that is, z_0 . The model distinguishes between the following surfaces: water (ocean or lake), sea-ice, snow and vegetative canopies, and computes the roughness length z_0 for them respectively. The friction velocity u_* is calculated as

$$u_* = C_D^{1/2} / U_h$$

G is a function accounting for Brownian diffusion, interception, and impaction of particles on the surface elements. Given the surface type, wind speed, air density, temperature at the ground level, and the surface momentum drag coefficient, the dry deposition velocity of aerosol particles can be estimated.

Slinn and Slinn [1981] derived a similar expression for dry deposition of particles on a natural water surface. Hygroscopic growth of particles was taken into account in the saturated viscous sublayer near the surface. As a first approximation, this effect was neglected because of high relative humidity (>80%) in open ocean marine boundary layer.

2.5. Vertical Turbulent Transport

Vertical transport of aerosol particles in the free troposphere due to turbulence is realized using eddy diffusivity formulations in the free atmosphere and drag coefficient formulations at the surface. This method is used in the Canadian climate model for the transport of momentum, heat, and moisture [*McFarlane et al.*, 1992]. The eddy diffusivities for momentum and heat are of the form

$$(K_m, K_h) = l^2 |\partial \mathbf{V} / \partial z| (f_m, f_h)$$
(20)

where V is the horizontal velocity vector and z the distance above the local terrain. The Richardson number (Ri) dependent functions (f_m, f_h) have the form

$$(f_m, f_h) = \begin{cases} 1 - 10 |Ri| / (1 + 10 |Ri/87|^{1/2}) & Ri < 0\\ (1 - 5\varepsilon Ri)^2 / (1 + 10(1 - \varepsilon)Ri) & 0 \le Ri \le 1/5\varepsilon \\ 0 & Ri > 1/5\varepsilon \end{cases}$$
(21)

where

$$l = kz/(1 + kz/\lambda)$$

where k is the von Kármán constant and the asymptotic mixing length (λ) is specified as 100 m. Like moisture in the climate model, the aerosol particles have a diffusivity equal to that for heat. The flux in the free atmosphere A_{VT} is written as

$$A_{VT} = -\rho K_a \frac{\partial \chi_i}{\partial z}$$
(22)

where $K_a = K_e = K_h$; χ_i and ρ are defined as in (1). The flux at and near the surface $(A_{VT})_s$ is expressed as

$$(A_{VT})_{s} = -\rho_{s} |\mathbf{V}_{L}| (\chi_{L+1/2} - \chi_{g}) C_{a}$$
(23)

where $C_a = C_h$; $\chi_{L+1/2}$ is the lowest prognostic level aerosol mass mixing ratio. The drag coefficient (C_h) is related to the surface-layer bulk Richardson number (Ri_B) by following equations:

$$C_h = C_{DH}F_h$$

where $F_h(Ri_B)$

$$= \begin{cases} 1 - 10 |Ri_{\rm B}| / (1 + 10 |Ri_{\rm B} / (87A_{h}^{2})|^{1/2}), & Ri_{\rm B} < 0\\ (1 - 5\varepsilon Ri_{\rm B})^{2} / [1 + 10(1 - \varepsilon) Ri_{\rm B}], & 0 \le R\iota_{\rm B} \le 1/5\varepsilon\\ 0, & Ri_{\rm B} > 1/5\varepsilon \end{cases}$$
(24)

 $A_h = [(z_0/z_L)^{1/2}]k^2/C_{DH}, z_0$ is the roughness height which is calculated inside the climate model, and ε is a constant accounting for the effects of lack of homogeneity. In the free atmosphere $\varepsilon = 3$, and at the surface over water while over land and ice covered surface, it is set to zero. Transport of aerosol particles by vertical advection and convective mixing is not included in this column version simulation.

3. Sea-Salt Aerosol Simulation by the Model

In order to test the aerosol model parameterization, a single one-dimensional, time variant column version (FIZ-C) [Therrien, 1993] of the Canadian general climate model GCMII is chosen to generate a climate with necessary meteorological parameters for the aerosol model. This local climate model is driven by dynamic tendencies $(\partial U/\partial t, \partial V/\partial t, \partial q/\partial t, \partial T/\partial t, \partial$ $\ln(ps)/\partial t$ precalculated by the GCMII at its lateral boundary conditions, where U and V denote the horizontal velocities, while q, T, and ps are the specific humidity, temperature, and surface pressure, respectively. In this version, dynamic tendencies are updated every 24 hours during the simulation. The complete GCM physics is recalculated in FIZ-C with 20-min time step to produce time variant fields such as wind, temperature, relative humidity, cloud coverage, and precipitation. The FIZ-C has 10 vertical levels stretching from surface to ~34 km and represent one grid cell $(3.75^\circ \times 3.75^\circ)$, that is, $\sim 400 \times 400$ km) of the GCM. Since the dynamic tendencies for the tracers (aerosols) are not yet available from the GCM, the horizontal



Figure 3. The geographic location for the test run in the North Atlantic.

advection of aerosols is neglected. In other words, it is assumed that there is an infinite homogeneous domain in the horizontal and the solution for aerosol concentration represents a quasisteady state balance between local production and local removal.

A grid point from the GCMII in the North Atlantic (latitude 61.2° north, longitude 352.5°) was selected for the test run (Figure 3). The simulation was run for 90 days starting January 1. The aim was to evaluate the performance of the model by comparing model predictions with available experimental observations from that region. Because of the one-dimensional nature of the FIZ-C used with the aerosol model, the comparison shall not serve as a validation analysis which will be performed when the aerosol model is coupled to a three-dimensional regional or global version of GCMII but rather it will serve to illustrate qualitatively how realistic are the parameterizations of various processes affecting sea-salt aerosols.

Figure 4 shows simulation results for aerosols in the four lowest atmospheric layers (layer depth: 0-166 m, 167-722 m, 723-1767 m, and 1767-3594 m) together with the wind speed at 10 m and precipitation for January, February, and March. Predictions for the first 10 days were omitted. This is the spin-up time for the model. Note that the simulation region in the North Atlantic is on the storm track of cyclones during winter. The periodic variation of wind and precipitation shows a succession of synoptic disturbances with a period of 3-5 days characteristic of winter storms. The variation of all meteorological variables is consistent with those in the GCM. Comparison of the two graphs immediately reveals that the sea-salt concentration is regulated more strongly by surface wind than by precipitation, especially the surface layer (0-166 m). Detailed discussion of the results will be given in the following sections.

3.1. Observation Available for Model Validation

A summary of observational data for sea-salt aerosols is listed in Table 4 for comparison with the model prediction. Experimental location, measurement method, sampling height as well as the maximum wind speed during the measurement are given in the table. There are basically two parameters for determining the sea-salt aerosols, that is, their size and mass. Among the observations, *Lovett* [1978], *Kulkarni et al.* [1982],



Figure 4. Simulation results for the test location. (a) Variation of total sea-salt aerosol concentrations in the first four model layers extending from the surface to 3594 m. (b) The wind speed and precipitation for the same period.

and *Marks* [1990] measured the total sea-salt mass as a function of wind speed. Their methods involve the collection of marine particles on a filter and then analyze the sodium content on it. While Marks obtained the total sodium by neutron activation method, the others measured only the soluble sodium. The total Na measurement eliminates the artifacts caused by non-sea-salt particles. Particle size distribution was measured by Woodcock [1953], Exton et al. [1986], Gras and Ayers [1983], and Marks [1990] by different techniques. Sea-salt content was obtained by analyzing the sodium [Woodcock, 1953] and by using the sea-salt density and particle volume [Exton et al., 1986; Gras and Ayers, 1983]. The latter technique for obtaining sea-salt contents may be subject to interference by non sea salt in the particles. An important assumption in this particle size to sea salt mass calculation is that the collected aerosol sample is close to 100% marine origin.

The location of experiment can strongly affect the representativeness of observations of open marine aerosols modeled here. Measurements conducted on a ship over the ocean [Tsunogai et al., 1972; Lovett, 1978; Marks, 1990] are closer to ideal marine aerosols than those performed inland [Kulkarni et al., 1982] or on a coast [Gras and Ayers, 1983; Exton et al., 1986]. The inland (1.8 km) observation by Kulkarni et al. was certainly affected by coastal surf as well as by dry depositional losses and hence is not appropriate for comparison with our model. Even if the measurement was made over open ocean, the particle size is not completely characteristic of sea-salt aerosols. Anthropogenic sulphates and formation of DMS oxidation products can contribute substantial into non-sea-salt fine particles, especially during high biogenic activity periods. Therefore the total sea-salt mass from particle size measurements [Exton et al., 1986; Gras and Avers, 1983] may be subjected to some uncertainties. It also seems that the measurements by Gras and Ayers and Exton et al. may suffer from beach surf effects, sedimentation, and turbulent deposition losses to the beach as their probes were several hundred meters away from the tide line.

Most of the measurements in Table 4 were made below 100 m, except *Woodcock* [1953] whose data were taken at cloud levels (~600 m). Accordingly, the observations by *Tsunogai et al.* [1972], *Lovett* [1978], and *Marks* [1990] are chosen for comparison with our model predictions for the surface layer (0–166 m), while that by *Woodcock* [1953] is used for comparison to simulation in layer 2 (167–722 m). The total sea-salt by sodium measurements and marine test locations by these investigators match closely to the conditions used in the simulation.

3.2. Size Distribution of Sea-Salt Aerosols

The particle radius ranging between 0.03 μ m and 8 μ m is divided into 8 size bins according to Table 5. In this size range, the main mechanism of sea-salt generation is attributed to the

Table 4. Published Constants for the Relationships Between Airborne Sea-Salt Concentration χ and Wind Speed U_{10} by Using Equation (25)

Authors	Latitude	Method of Measurements	Location and Sampling Height	Maximum Wind, m s ⁻¹	$a, s m^{-1}$	<i>b</i> , μg m ⁻³
Woodcock [1953]	20°N	glass slide (Na and size)	cloud base over Pacific Ocean (600 m)	35	0.16	2.57
Tsunogai et al. [1972]	40°N-50°N	membrane filter (na)	ship on Pacific Ocean (12-14 m)	18	0.62	0.33
Lovett [1978]	50°N-60°N	membrane filter (na)	weather ships in Atlantic Ocean (5–15 m)	20	0.16	4.26
Kulkarni et al. [1982]	19°N	membrane filter (na)	W. Indian coast 1.8 km inland	8	0.27	5.35
Exton et al. [1986]	57°N	ASAS and CSAS (size)	Hebridean beach facing Atlantic (15 m)	20	0.17	14.30
Gras and Ayers [1983]	41°S	round jet collector (size)	Cape Grim (94 m)	20	0.124	2.52
Marks [1990]	N/A	neutron activation (impactor Na)	ship on North Sea (12 m)	24	0.23	1.10
Marks [1990]	N/A	neutron activation (filter Na)	ship on North Sea (12 m)	24	0.23	1.13

Concentration is in $\mu g m^{-3}$; wind speed is in meters per second.

Table 5. Aerosol Size Discretization Scheme

Bin	Size Range	
1	0.03-0.06	
3	0.13-0.25	
4	0.25-0.50	
6 7	1–2 2–4	
8	4–8	

The radius is in micrometers.



Figure 5. The sea-salt aerosol size distribution of mass concentrations. (a) Model predictions at three wind speeds; (b) experimental observations [*Marks*, 1990] for both dry and wet conditions; (c) comparison with *McGovern et al.* [1994]. The vertical dashed line in Figure 5c indicates the smallest experimental radius at which (5) was based.

a) PREDICTIONS

indirect scheme, that is, via bubbles. The predicted mass size distribution of sea-salt aerosol (Figure 5a) is compared with experimental results of *Marks* [1990] in the North Sea and *McGovern et al.* [1994] at Mace Head on the west coast of Ireland. Several features are revealed by the comparison. The size distribution (1) of the sea-salt mass concentration compares well within the observational data (2) in terms of the shapes of the distribution curves and the magnitudes of the mass concentrations (Figure 5c). This agreement indicates that the parameterization schemes adapted in the model are performed reasonably well. It is also seen from Figure 5a that

b) MARKS OBSERVATIONS



Figure 6. (a) As a function of wind speed, the number size distribution of sea-salt aerosol particles predicted by the model. (b) The predicted sea-salt surface flux distribution (equation (5)) corresponding to results in Figure 6a. (c) Comparison of observations [from *Fitzgerald*, 1991, Figure 1] with predictions at two wind speeds. The vertical dashed line in Figures 6b and 6c indicates the lowest experimental radius at which (5) was based.

some detailed structures of the distribution at submicron ranges are resolved by the simulation. There is a hint of bimodal mass size distribution in the model results. This feature is even more pronounced when the predictions are plotted as number size distribution (Figure 6). The bimodal distribution from the model is not as clear as for the observations. This is likely due to lower size resolution in the model than observations as well as the fact that processes of cycling the aerosol through cloud formation and evaporation are not considered in the present one-dimensional model. They will, however, be taken into account in the three-dimensional model to be run later.

Furthermore, coagulation of aerosols is ignored in this version of the model. As a result, at fine particle size ranging below 0.1 μ m, the prediction yielded a higher concentration than the observation for both mass and number size distribu-

tion. Coagulation would also strengthen the concentration near 0.1 μ m and thereby enhance the bimodal structure. Another effect is that other non-sea-salt aerosols also contribute to the distribution measured in the remote marine environment. However, in the radius range where sea-salt dominates (0.5 to 10 μ m) the model and experimental results agree reasonably well. A comparison of Figures 6a and 6b illustrate that the shape of the number size distribution curves is strongly influenced by the sea surface flux size distribution of sea-salt droplets which is the only source of sea-salt aerosols. As seen from Figure 6b, the flux curve also does not show a distinct bimodal distribution.

It should be pointed out that the empirical sea-salt production function by *Monahan et al.* [1986] (equation (5a)) is based on observations of sea-salt generation for particles larger than about 0.3 μ m in radius (dry). This is the lower limit of particles

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detected in the experiments with which the production function was compared well. Extrapolation beyond the range results in uncertainties since the empirical function begins to deviate from the experimental observations below 0.8 μ m radius (80% RH). From this model simulation, use of *Monahan et al.*'s [1986] production function seems to reasonably simulate production down to a radius of about 0.1 μ m (dry). However, below 0.1 μ m, a large overestimate of both mass and number concentrations occurs (e.g., the prediction curves on the left of a dashed line in Figures 5c and 6c).

3.3. Wind Speed Dependence of Sea-Salt Aerosols

The effect of wind speed on the sea-salt aerosol ambient concentration (χ) has been investigated experimentally by many researchers [*Woodcock*, 1953; *Lepple et al.*, 1983; *Lovett*, 1978; *Gras and Ayers*, 1983; *Exton et al.*, 1986; *Marks*, 1990]. Generally, the dependence is expressed as a log-linear fit to the experimental data between concentration and wind speed:

$$\ln \chi = \ln (b) + a U_{10} \tag{25}$$

In Figure 7b, model predictions of total mass are compared to experimental observations in the surface layer. A general agreement for the wind effect is obtained between the predictions and experiments (Figure 7b). Best fitting curves from the model results are also shown in Figure 7a. A slope (a) of 0.26 and 0.13 and a correlation coefficient of 0.82 and 0.23 are predicted for total and fine particle ($r_d = 0.13$ to 0.25 μ m) mass, respectively. Using a five-stage impactor and filter sampler to measure the sea-salt aerosols at various wind speeds, Marks [1990] obtained the dependence of sea-salt aerosols on wind speed for five size ranges and for total sea salt, respectively. Under dry nonprecipitating conditions, except for the diameter of 0.49–0.95 μ m where the correlation coefficient is 0.31, the slopes for all other four size stages ranging from 0.95 to 30 µm (diameter) was about 0.23 [Marks, 1990, Table 2], which is very close to the model simulated results of 0.26 for total mass. The slope is 0.12 for the observation of particles in diameter of 0.49–0.95 μ m and 0.13 for the simulation of particles in the radii ranging from 0.13 to 0.25 μ m.

Exton et al. [1985, Table 3] found the same trends for three size categories of sea-salt aerosols based on the field measurements off the northwest coast of Scotland. For the ultra large sea-salt ($r = 8-16 \mu$ m) aerosols, the slope and the correlation coefficient were 0.235 (average) and 0.59 (average) respectively. For the large ($r = 0.3-8 \mu$ m) aerosols, they changed to 0.13 and 0.56 respectively. For the small ($r = 0.1-0.3 \mu$ m) aerosols, the slope was reduced even more to 0.08 while the correlation coefficient was 0.4.

Tsunogai et al. [1972] measured sea-salt concentration from a cruise ship on the Pacific Ocean and a slope of 0.62 was obtained for the wind dependence. It is much larger than other observation's. They attributed it to two reasons: (1) the production of sea-salt particles >10 μ m in radius or 10⁻⁸ g in mass is more dependent on the wind speed than the production of smaller ones; (2) the residence time of the larger particles is so short that they contribute only to the weight concentration of sea-salt particles near the surface of the ocean. Lovett [1978] attributed the large value to the moving ship which may generate sea-salt particles. This effect is unlikely true since samples were discarded when a tail wind was recorded. In our opinion, it may be due to the experimental method used to construct the sea-salt wind dependence. It was reported in the paper that the sampling time was from 20 to 50 hours. During the long experimental period, the wind speed might undergo substantial change. Because of the exponential relationship between sea salt and wind speed, the averaged sea-salt concentration at the averaged wind speed could be higher than an averaged sea-salt at the same averaged wind for a short time when the wind speed would not change substantially. This is clearly shown in Figure 8 where the wind speed is assumed to undergo a sinusoidal change from 0° to 180° with a speed of 0-15-0 m s⁻¹ cycle for a designated time and an exponential relationship is assumed between sea salt and wind speed. For sampling through 0° -180° with every degree a sample (duration 180), the root mean square (rms) wind speed for the duration is 10.6 m s⁻¹, and the rms sea-salt concentration is 29.6 μ g m⁻³. For a shorter sampling duration (51) which results in the same rms wind speed (10.6 m s⁻¹), the rms sea-salt concentration is reduced to 22.71 μ g m⁻³. When the wind speed is constant at 10.6 m s⁻¹ for the duration (180 or 51), the sea-salt concentration is 16.37 m s⁻¹ which is given by the assumed exponential relationship. Furthermore, even if the wind speed only changes slightly in a $15-16-15 \text{ m s}^{-1}$ cycle, the long sampling duration (180) still results in an overestimated rms sea-salt concentration when compared with a sampling duration of 51 for the same rms wind speed. Therefore, if wind speed changes, the long-time average of sea-salt and wind speed by Tsunogai et al. [1972] actually overestimates the real relationship between the sea salt and wind speed, that is, higher slope, compared to the sampling time of 0.5 to 3 hours of Marks [1990] and 1-2 to several hours of Lovett [1978].

The sea-salt and wind speed relationship given by *Woodcock* [1953] was obtained at cloud levels (about 600 m) in the Pacific with a slope (a) of 0.16. Although *Lovett* [1978] obtained the same value for an annual average between August 1974 and July 1975 in the Atlantic, the measurements were performed at 5–15 m. It is expected that as altitude increases the dependency of sea salt on wind becomes weaker. This feature is reflected in the model which gives a smaller slope of 0.15 in the layer of 166–722 m than that of 0.26 in the surface layer. It is closer to 0.16 of *Woodcock* [1953].

The constant b in (25) is referred as the background sea-salt loading. It is the sea-salt concentration when wind speed reaches zero. From Figure 7a, our prediction yields a value of 1.4 μ g m⁻³ for Heimaey. Compared to the observations (Table 4), b is 1.13 μ g m⁻³ for Marks [1990], 2.57 μ g m⁻³ for Woodcock [1953], 0.33 μ g m⁻³ for Tsunogai et al. [1972], and 4.26 μ g m^{-3} for Lovett [1978]. There are two factors which may explain the observational difference. (1) The three experiments were made at the North Atlantic [Lovett, 1978], the North Sea [Marks, 1990] and the Pacific [Tsunogai et al., 1972] respectively. Different meteorology such as wind speed and precipitation at these sites will result in different patterns for sea-salt generation and removal which influence the constant a and b in (25). Our simulation also shows the variability of constant a and b at various sites (Table 6). The salinity of the ocean may play a role in the difference but may not account for the big discrepancy between 4.26 and 0.218 since the difference between the highest salinity of 35.5% in the North Atlantic and the lowest of 34.2% in the North Pacific cannot contribute such a big difference in sea-salt concentration; (2) the measurement techniques used, for example, the sampling time, average methods and equipment. The sampling duration problem has been discussed above with Tsunogai et al.'s [1972] measurements. When a size distribution is measured by an





Figure 7. Comparison of sea-salt mass concentration as a function of wind speed; (a) model prediction for total mass and radii of $0.13-0.25 \ \mu m$ in the first model layer at Heimaey of Iceland and (b) observations with the model predictions at two sites.

impactor, isokinetic conditions at the inlet have to be maintained to reduce the inlet losses of large particles [Marple and Willeke, 1976].

It is interesting to note that for both observations and predictions, as the particle size decreases, the slope and correlation of the sea-salt vs. wind speed curve decreases. This indicates that for large sea-salt particles (e.g., $r_d = 4-8 \ \mu\text{m}$) the local surface wind is the dominant driving force in determining the concentration. For small particles (e.g., $r_d = 0.1-0.3 \ \mu\text{m}$), however, other factors begin to affect the concentration. One important parameter to characterize this phenomenon is the residence time. For large particles, the residence time is on the order of hours (see next section for details). Any such particles generated by wind will deposit back to the surface very quickly. At any time and location, the sea-salt concentration is maintained mainly by the local wind speed. Hence a stronger wind speed dependency is observed. On the other hand, the smaller particles have a residence time of several days. This implies that at any time the aerosol concentration of smaller particles is controlled not only by the local wind speed but also by long-range transport of fine sea-salt particles. The longer residence time yields a smaller slope and correlation coefficient between sea salt and wind speed. In addition, non sea-salt biogenic sulphur aerosol can contribute substantially to fine particles. Since their concentration is less dependent on wind speed than that of sea salt, the correlation with wind speed is less.

According to our simulations, the dependence of total seasalt aerosols also varies with geographic locations. For four marine locations simulated in this study, the slope a ranges from 0.20 to 0.26 (Table 6). It seems that the climate pattern in a specific site may regulate the removal processes and hence the dependence. Even though different measuring methods may yield a difference in the absolute value of sea-salt concentrations, the dependence on wind speed should be relatively less affected. The variant slopes in Table 4 are indicative of such differences. A global relationship used for the sea-salt dependence on wind speed [*Erickson and Duce*, 1988] may be subject to large uncertainties.

3.4. Residence Time of Sea-Salt Aerosols

Residence time of the particles is an important indicator of particle cycles in the atmosphere. Factors regulating residence time include the production, accumulation rate, removal and growth of the aerosols [Junge, 1974; Bolin and Rodhe, 1973; Slinn, 1984]. Except for horizontal advection, all major physical processes are included in this aerosol model. Therefore it is an ideal tool to investigate the effect of local processes on the residence time of the atmospheric aerosols.

The residence time of particles in each size bin is dynamically calculated according to the following formula:

$$\tau = \frac{\chi}{\left\lceil \frac{\partial \chi}{\partial t} \right\rceil} \tag{26}$$



Figure 8. The effect of sampling duration on the rms sea-salt concentration assuming a sine cycle change of wind speed and an exponential relationship between sea salt and wind speed.

Table 6. Total Sea-Salt Mass Concentration χ Dependence on 10-m Wind Speed U_{10} : $\chi = be^{aU_{10}}$

Location	$a, s m^{-1}$	b, $\mu g m^{-3}$	<i>r</i> ²
Mace Head	0.20	3.1	0.76
Hawaii	0.21	3.4	0.58
Bermuda	0.22	2.5	0.68
Heimaey	0.26	1.4	0.82

where χ is the concentration and $\left[\frac{\partial \chi}{\partial t}\right]$ the tendency due to either addition or removal processes calculated at each time step. Under steady state conditions, this tendency is identical for both of them. In a dynamic model where steady state is hardly reached, a choice has to be made as to which tendency is used. Fortunately, in terms of the averaged tendency over a long period, a quasi-steady state is maintained, that is, an averaged tendency is equal for both addition and removal. Consequently, an averaged residence time is calculated by using the removal tendency. Figure 9 shows the residence times for particles in two size bins for the atmospheric surface layer (0–166 m). For particles in bin 8 ($r_d = 4-8 \ \mu$ m), the averaged residence time is about 30 min (Figure 9a). Because of a large dry deposition and settling velocity, this category of aerosols will not reach the upper atmosphere and engage in long range transports. Smaller particles ($r_d = 0.13 - 0.25 \ \mu m$) have a residence time of about 60 hours (Figures 9b) in the first atmospheric layer (0-166 m). The longer residence time of small particles allows them to reach high altitudes and participate in long-range transport.

It should be pointed out that the residence time in Figure 9 is for the first atmospheric layer only (0-166 m). This is the reservoir where the removal tendency is calculated. It is anticipated that the residence time will vary at different layers as the removal tendency and concentration of aerosols will be controlled by different conditions.

4. Conclusions

The application of this aerosol model to sea-salt aerosols revealed very good agreement between predictions and observations in terms of particle number density, size distribution and wind effects on sea-salt aerosols. The source function by Monahan et al. [1986] produces reasonably sea-salt emission rate for particles down to 0.1 μ m. However, below 0.1 μ m, a large overestimate of both mass and number concentrations occurs probably due to the extrapolation of the Monahan et al.'s formula beyond the particle size range within which the formula is defined. It is found that the total mass of atmospheric sea-salt aerosols is mainly governed by the local wind speed. A regression of the simulated results as a function of wind speed gives a slope (a) ranging from 0.20 to 0.26 s m⁻¹ and an intercept corresponding to background salt loading (b)from 1.4 to 3.4 μ g m⁻³ for the four locations modeled, indicating that the sea-salt wind correlation depends on geographic locations. Small particle concentrations are less dependent on wind speed than large particles. This was true in both experiments and this simulation. The residence time of sea-salt aerosols is found ranging from 30 min ($r_d = 4-8 \ \mu m$) to 60 hours $(r_d = 0.13 - 0.25 \ \mu \text{m})$ in the first atmospheric layer (0-166 m) depending on the size of the particles and local meteorological conditions. The longer residence time for smaller particles



(b) Residence Time of Sea-salt Aerosols [r_d=0.13-0.25 μm]



Figure 9. The residence time of sea-salt aerosols in the first atmospheric layer (0-166 m) for (a) large and (b) small particles. The radii of sea-salt aerosols are shown for dry particles.

explains the less dependency of sea-salt aerosols on the wind speed.

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(a) Residence Time of Sea-salt Aerosols [r_d=4-8 μm]

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