

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 one-dimensional 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 (χ , $\mu\text{g m}^{-3}$) 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 ($r = 4\text{--}8 \mu\text{m}$) to ~ 60 hours for small particles ($r = 0.13\text{--}0.25 \mu\text{m}$). 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., Br_2 , Cl_2) [e.g., Mozurkiewicz, 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 μm with a bimodal size distribution in the submicron portion [Fitzgerald, 1991]. Since the observations

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 one-dimensional 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

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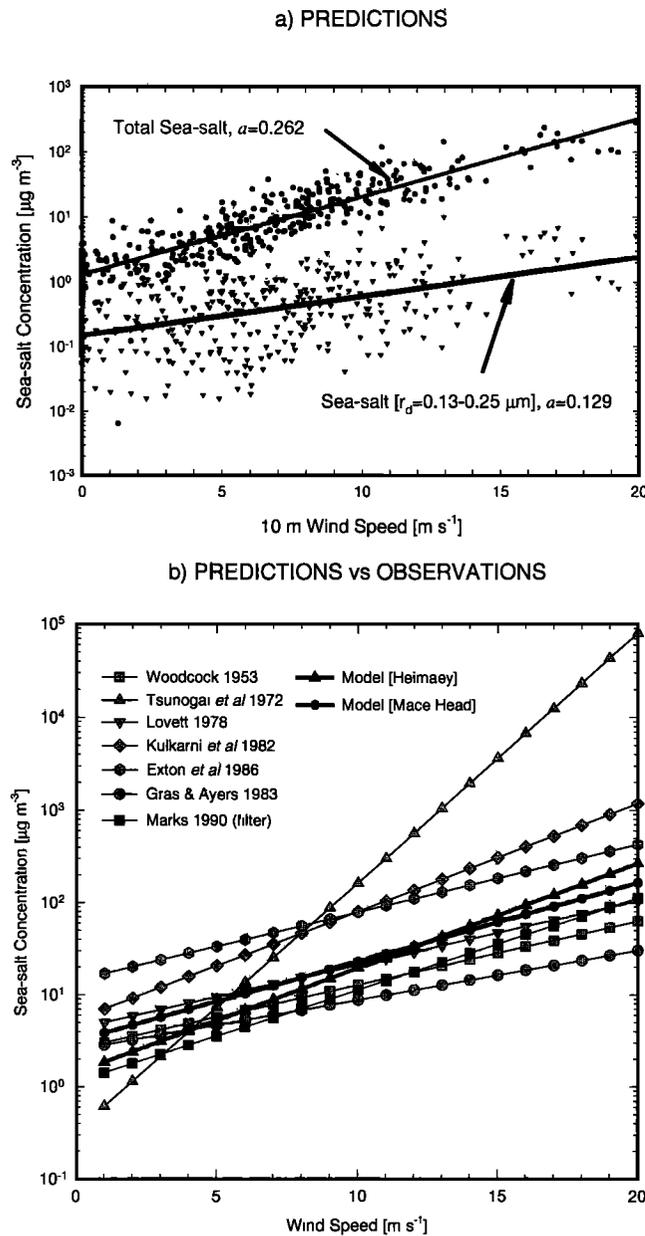


Figure 7. Comparison of sea-salt mass concentration as a function of wind speed; (a) model prediction for total mass and radii of 0.13–0.25 μm in the first model layer at Heimaey of Iceland and (b) observations with the model predictions at two sites.

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., $r_d = 4\text{--}8\ \mu\text{m}$) the local surface wind is the dominant driving force in determining the concentration. For small particles (e.g., $r_d = 0.1\text{--}0.3\ \mu\text{m}$), 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 sea-salt aerosols also varies with geographic locations. For four marine locations simulated in this study, the slope α 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:

$$\tau = \frac{\chi}{\left[\frac{\partial \chi}{\partial t}\right]} \quad (26)$$

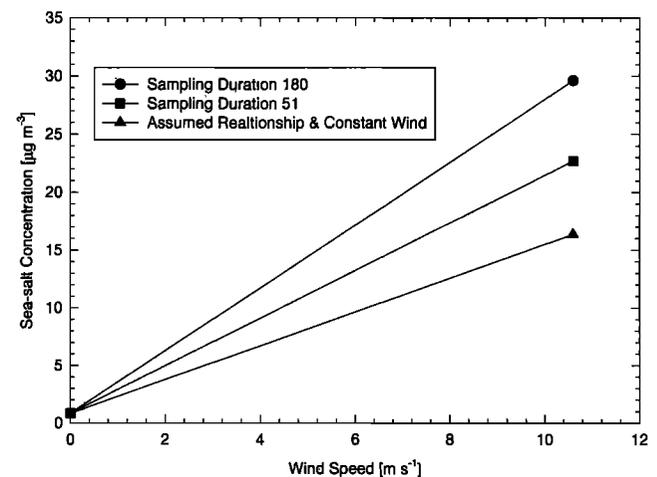


Figure 8. The effect of sampling duration on the rms sea-salt concentration assuming a sine cycle change of wind speed and an exponential relationship between sea salt and wind speed.

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 , $\mu g\ m^{-3}$	r^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 $[\partial\chi/\partial t]$ 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 ($r_d = 4\text{--}8\ \mu\text{m}$), 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 ($r_d = 0.13\text{--}0.25\ \mu\text{m}$) 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 reasonable sea-salt emission rate for particles down to $0.1\ \mu\text{m}$. However, below $0.1\ \mu\text{m}$, 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 $3.4\ \mu g\ m^{-3}$ 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 ($r_d = 4\text{--}8\ \mu\text{m}$) to 60 hours ($r_d = 0.13\text{--}0.25\ \mu\text{m}$) 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

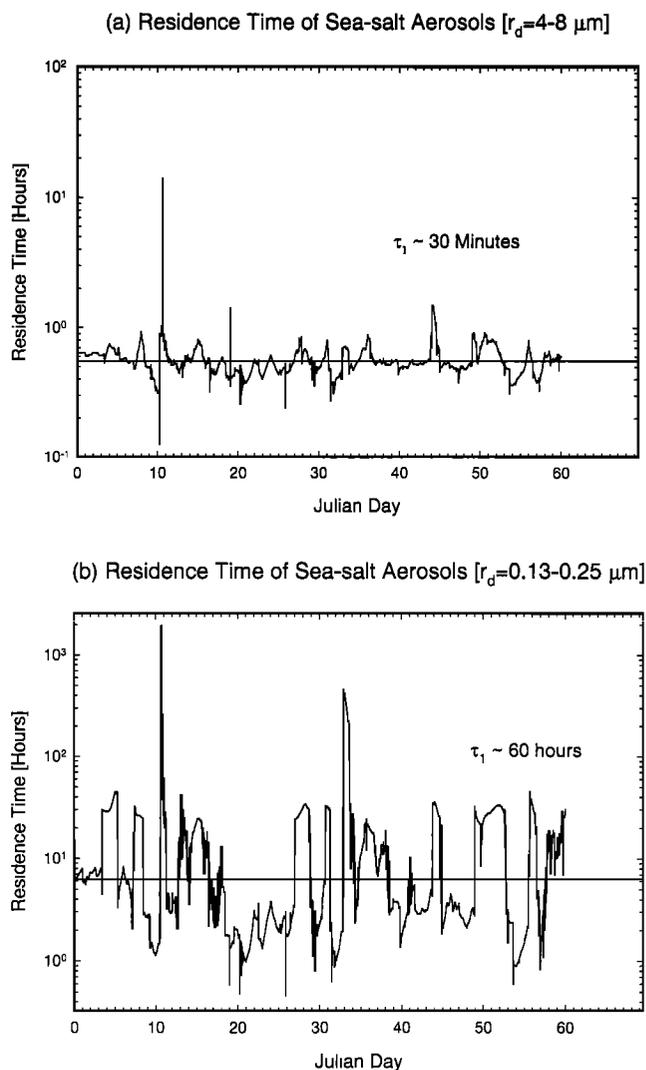


Figure 9. The residence time of sea-salt aerosols in the first atmospheric layer (0–166 m) for (a) large and (b) small particles. The radii of sea-salt aerosols are shown for dry particles.

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

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