

5. Aerosol model (MASINGAR mk-2)

Atmospheric aerosols influence the climate by perturbing the Earth's radiation budgets in several ways. A direct radiative effect is caused by the direct scattering and absorption of atmospheric radiation by aerosols. An indirect radiative effect is caused by aerosols also acting as cloud condensation nuclei, which affects properties of clouds such as cloud albedo (Twomey, 1974; Twomey, 1991), precipitation formation, and cloud lifetime (Albrecht, 1989). Absorptive aerosols such as black carbon or mineral dust warm the atmosphere and reduce solar radiation at the surface, which increases atmospheric stability. Absorptive aerosols can locally inhibit cloud formation or reduce cloud cover by heating cloud droplets. This effect is called the semi-direct aerosol effect (Hansen et al., 1997). Moreover, absorptive aerosols deposited on a snow surface reduce the albedo of the snow surface and enhance the melting of the snow (Hansen and Nazarenko, 2004).

In MRI-ESM1, atmospheric aerosols are calculated with a global aerosol model called MASINGAR mk-2 (Model of Aerosol Species in the Global Atmosphere), which is coupled with the Scup coupler library (Yoshimura and Yukimoto, 2008). MASINGAR mk-2 is an improved version of the MASINGAR aerosol model (Tanaka et al., 2003) modified to work as a component of MRI-ESM1. The model treats five aerosol species: non-sea-salt sulfate, black and organic carbon, sea salt, and mineral dust. The grid resolution of the model is variable, and can be different from that of the coupled AGCM.

5.1. Coupling with the atmospheric general circulation model MRI-AGCM3

The aerosol model receives the meteorological fields and surface conditions from the AGCM through the coupler. The meteorological fields include horizontal wind components, air temperature, specific humidity, convective mass flux, precipitation and evaporation with convective and large-scale clouds, the vertical eddy diffusion coefficient, and surface pressure. The surface conditions include near-surface wind speed, surface air temperature, ground temperature, snow amount, land-use type, vegetation amount, and leaf area index. The MASINGAR mk-2 aerosol model sends the concentrations of the five aerosol species and the deposition fluxes of absorptive aerosols (black carbon and mineral dust) to MRI-AGCM3 to be used in calculations of the direct and indirect radiative effects of the

aerosols and of the snow albedo. The sea-salt aerosol is calculated in 6 size bins in the aerosol model, but is sent in 2 bins (smaller and larger than 1 μm).

5.2. Coupling with the chemistry climate model MRI-CCM2

The interaction of aerosols with atmospheric chemistry is incorporated into MRI-ESM1 by coupling of the MASINGAR mk-2 aerosol model with the MRI-CCM2 atmospheric chemistry model, which is described in section 6. MASINGAR mk-2 receives concentrations of OH, H₂O₂, HO₂, O₃, NO₃, and O(³P) for the calculation of chemical reactions involving the sulfate aerosol. If MRI-CCM2 is not coupled with MRI-ESM1, then MASINGAR mk-2 uses monthly averaged concentrations of the chemical species pre-calculated by MRI-CCM2. On the other hand, MASINGAR mk-2 sends the three-dimensional surface area density of the aerosols to MRI-CCM2, enabling it to incorporate heterogeneous chemical reactions on the aerosol surface.

5.3. Processes in the global aerosol model

5.3.1. Atmospheric transport

Atmospheric transport is calculated using a semi-Lagrangian advection scheme and schemes for sub-grid turbulent vertical diffusion and convective transport. The vertical eddy diffusion coefficient is taken from that of water vapor, calculated in the AGCM. Convective transport is calculated using the updraft mass flux from the cumulus convection scheme (AS-type or Yoshimura scheme) in MRI-AGCM3.

Aerosol particles are subject to gravitational settling relative to air motion and are assumed to fall with the terminal velocity V_s . The terminal velocity V_s is calculated under the assumption that the particles are spherical as

$$V_s = \frac{2C_c(\rho_p - \rho_a)gr_p^2}{9\mu}, \quad (5.1)$$

where ρ_a is the air density; ρ_p and r_p are the density and radius of the particle, respectively; g is gravitational acceleration; and μ is the viscosity of the air. C_c is the Cunningham slip-flow correction, expressed as

$$C_c = 1 + Kn[A + B\exp(-C / Kn)] \quad (5.2)$$

where Kn is the Knudsen number for air, and the coefficients A , B , and C are set to 1.257, 0.4, and 1.1.

5.3.2. Dry and wet deposition processes

Dry deposition is parameterized by the resistance-in-series model (Seinfeld and Pandis, 1997), which includes turbulent impaction and gravitational settling. Wet deposition is distinguished between in-cloud and below-cloud scavenging and is also categorized by cloud type (convective or large-scale) and species (aerosol or gas; accommodation with water droplets). For in-cloud scavenging by large-scale precipitation, we used the parameterization developed by Giorgi and Chameides (1986). Both the dry and wet deposition schemes for sea salt and mineral dust aerosols are particle-size dependent.

For water-soluble gases such as SO_2 , wet scavenging is calculated simultaneously with the aqueous-phase chemistry. The fraction of a water-soluble gas that is in liquid water is assumed to follow Henry's law of equilibrium, which is temperature and pH dependent. The rate of wet deposition is calculated as the ratio of the new rainwater formation rate to the total water in the grid box (cloud water plus rainwater formation). At temperatures below -20 °C, all cloud droplets are assumed to be frozen and precipitation is assumed to be snow; thus, wet scavenging of water-soluble gases is ignored, whereas aerosol particles are assumed to be scavenged by either rain or snow. Evaporation of rainwater is considered when calculating the wet deposition rate. When evaporation occurs, a fraction of the trace elements is released back to the air. This fraction of released trace elements is assumed to be proportional to the amount of evaporated rainwater.

5.3.3. Emission processes and chemical reactions

For the emission processes of anthropogenic sulfur compounds and carbonaceous aerosols, prescribed emission inventories are used. The emissions of oceanic DMS, sea salt, and mineral dust are calculated from the meteorological and surface conditions.

Sulfate

The emission and formation of sulfate aerosol is based on Tanaka et al. (2003), but some modifications are applied. Sulfate aerosols are mainly formed from gaseous and aqueous phase reactions of SO₂ and DMS. Emissions of SO₂ from fuel consumption, ships, and biomass burning are taken from prescribed inventories. The global source distribution from volcanos is taken from GEIA (Andres and Kasgnoc, 1998).

Emission of DMS from the ocean is calculated as a function of the surface seawater DMS concentration and its piston velocity. Climatological monthly averaged surface seawater DMS concentrations are adopted from Kettle et al. (1999). The piston velocity of DMS is calculated as a function of the 10-m wind velocity and the Schmidt number of DMS at the sea-surface temperature. This model implements two parameterizations of DMS piston velocity. First, the widely used empirical scheme of Liss and Merlivat (1986) is employed as the default scheme:

$$k_{\text{DMS}} = k_u \left(\frac{\text{Sc}_{\text{DMS}}(T_{\text{SST}})}{\text{Sc}_{\text{CO}_2}(20)} \right)^\alpha \quad (5.3)$$

where α is an empirical parameter that is $-2/3$ if wind speed at 10 m height (U_{10}) is below 3.6 m s^{-1} and $-1/2$ otherwise. $\text{Sc}_{\text{CO}_2}(20)$ is the Schmidt number of CO₂ at the sea surface temperature of 20 °C (= 595), $\text{Sc}_{\text{DMS}}(T_{\text{SST}})$ is the Schmidt number of DMS at sea-surface temperature T_{SST} (°C) calculated by the formula of Saltzman et al. (1993),

$$\text{Sc}_{\text{DMS}} = 2674.0 - 147.12T_{\text{SST}} + 3.726T_{\text{SST}}^2 - 20.038T_{\text{SST}}^3 \quad (5.4)$$

The coefficient k_u is expressed as a function of U_{10} :

$$k_u = \begin{cases} 0.17U_{10} & \text{for } U_{10} \leq 3.6 \text{ m s}^{-1} \\ 2.85U_{10} - 9.65 & \text{for } 3.6 < U_{10} \leq 13 \text{ m s}^{-1} \\ 5.9U_{10} - 49.3 & \text{for } U_{10} > 13 \text{ m s}^{-1} \end{cases} \quad (5.5)$$

To reduce computational costs, hydrogen sulfide (H₂S), carbon disulfide (CS₂), methane sulfonic acid (MSA), dimethyl sulfoxide (DMSO), and dimethyl sulfone (DMSO₂) are not considered.

For the formation of stratospheric sulfate aerosols, carbonyl sulfide (OCS) and SO₃ are added. Because the lifetime of OCS is very long and its mixing ratio is almost stable, the mixing ratio of OCS is assumed to be a constant at 500 pptv near the ground.

Black and organic carbon

Carbonaceous aerosols are classified as BC or OC. We assume that 80% of BC and 50% of OC are emitted as hydrophobic material, following Cooke et al. (1999). We also assume that the hydrophobic carbonaceous aerosols become hydrophilic over time, with an e -folding time of 1.2 days. Hydrophobic carbonaceous aerosols are scavenged by dry deposition only, whereas hydrophilic carbonaceous aerosols are scavenged by both dry and wet deposition. No chemical sinks for carbonaceous aerosols are assumed in this model. The particle densities of OC and BC are assumed to be 1.5 and 1.25 g cm⁻³.

Sources of carbonaceous aerosols are roughly divided into anthropogenic emissions, biomass burning, and volatile organic compound (VOC) gas emissions. Emissions of BC and OC from fuel consumption, ships, and biomass burning, and emission of BC from aircraft are taken from prescribed inventories. In the production of OC aerosol from the terrestrial emission of VOCs, we assume that terpene is converted to OC by gas-to-particle conversion. The monthly mean terpene emission inventory is adopted from the 1° × 1° resolution maps of GEIA databases (Guenther et al. 1995). We assume that 10% of emitted terpene is converted to OC aerosol. This secondary source of OC accounts for 11.8 TgC yr⁻¹.

Sea salt

Sea-salt aerosol is logarithmically divided into size bins ranging from 0.2 to 20 μm in diameter. The number of size bins is reduced from 10 of previous version (Tanaka et al. 2003) to 6, considering the computational cost of long-term climate simulations. Strong winds over the ocean surface produce sea salt, indirectly by bursting air bubbles during whitecap formation and directly by spume production. We adopted the empirical parameterization of the production mechanisms developed by Gong (2003), who improved the parameterization of Gong et al. (1997) and Monahan et al. (1986). The density function dF/dr (particles m⁻² s⁻¹ μm⁻¹) of the sea-salt production rate by indirect mechanisms is

$$\frac{dF_0}{dr} = 1.373U_{10}^{3.41}r^{-A}\left(1 + 0.057r^{3.45}\right) \times 10^{\left(1.607\exp(-B^2)\right)} \quad (5.6)$$

where U_{10} is the wind speed at 10 m (m s⁻¹) and r is the particle radius (μm). The parameters A and B are defined as

$$A = 4.7(1 + \Theta r)^{-0.017r^{-1.44}} \quad (5.7)$$

$$B = (0.433 - \log r) / 0.433 \quad (5.8)$$

Following Gong (2003), the parameter Θ is set to 30. Because the size range of the direct mechanism is larger than the maximum size bin (20 μm), the direct mechanism is neglected. The sea-salt emission flux of each size bin is calculated by integrating the formula (5.6) over the size range of each size bin.

Mineral dust

In this model, the dust emission flux into the atmosphere is calculated according to saltation-bombardment theory and source areas are determined by vegetation cover, snow cover, land-use type, and soil type. Here, we briefly describe the calculation of the dust emission flux. Tanaka and Chiba (2005, 2006) give more detailed descriptions for the treatment of mineral dust aerosol.

The mobilization of soil particles is initiated when the friction velocity on a bare surface (u_*) exceeds a threshold value called the threshold friction velocity (u_{*t}), which is calculated using the formula of Shao and Lu (2000) with the soil moisture factor (f_w) of Fécan et al. (1999),

$$u_{*t}(D) = f_w \sqrt{A_N \left(\frac{\rho_p g D}{\rho_a} + \frac{\Gamma}{\rho_a D} \right)} \quad (5.9)$$

where D is the particle diameter, g is gravitational acceleration, ρ_a is air density, ρ_p is particle density, $A_N = 0.0123$, and $\Gamma = 3 \times 10^{-4} \text{ kg s}^{-2}$. The factor f_w is calculated by

$$f_w = \begin{cases} 1 & w \leq w_r \\ \sqrt{1 + a[100(w - w_r)]^b} & w > w_r \end{cases} \quad (5.10)$$

where w is the gravimetric soil water content and w_r is the threshold gravimetric soil water content. The empirical constants a and b are set to $a = 1.21$ and $b = 0.68$.

The dust flux in the size range $[D_i, D_{i+1}]$ is calculated by using the theory of Shao et al. (1996),

$$\Delta F_{i0} = CA \frac{2}{3} \frac{\rho_p \gamma}{\rho_a} \int_{D_i}^{D_{i+1}} \frac{\beta_2(D_d)}{u_{*t}(D_d)^2} p(D_d) dD_d \int_0^\infty \beta_1(D_s) \tilde{Q}(D_s) p(D_s) dD_s \quad (5.11)$$

where D_d is the diameter of emitted dust particles, D_s is the diameter of saltating particles, $\tilde{Q}(D_s)$ is the mass flux of saltating particles, $p(D)$ is the size distribution of the parent soil, A is the erodible areal fraction of the model grid, and C is a global tuning factor, chosen to yield an annual global source strength of about 2100 Tg yr $^{-1}$ (Penner et al., 2001).

Following Shao et al. (1996), $\gamma \sim 2.5$, and β_1 and β_2 appear as functions of D_s and D_d ,

$$\beta_1(D_s) = 0.125 \times 10^{-4} \ln(D_s) + 0.328 \times 10^{-4} \quad (5.12)$$

$$\beta_2(D_d) = \exp(-140.7D_d + 0.37) \quad (5.13)$$

The saltation flux $\tilde{Q}(D_s)$ is calculated with a formula from Owen (1964)

$$\tilde{Q}(D_s) = \begin{cases} \frac{c_s(D_s) \rho_a u_{*s}^3}{g} \left(1 - \frac{u_{*t}(D_s)^2}{u_{*s}^2} \right) & u_{*s} > u_{*t} \\ 0 & u_{*s} \leq u_{*t} \end{cases} \quad (5.14)$$

where

$$c_s(D_s) = 0.25 + \frac{V_s(D_s)}{3u_{*s}}, \quad (5.15)$$

and $V_s(D_s)$ is the gravitational settling velocity of a saltating particle of diameter D_s . The size distribution of the parent soil is specified by using the global soil texture database of Webb et al. (2000).

The erodible areal fraction A is expressed in the following factorial form:

$$A = (1 - A_v)(1 - A_s)(1 - A_w) A_l A_t \quad (5.16)$$

where A_v , A_s , A_w , A_l , and A_t represent the factors of vegetation cover, snow cover, water cover, land-use type, and soil type. We used the soil-type data set from Zobler (1986), which divides the soil into 106 types, with a horizontal resolution of $1^\circ \times 1^\circ$. The soil type ‘‘lithosol’’ is excluded as a possible dust source, because it consists of hard rocks from the mountainous regions, according to Tegen and Fung (1995). The soil-type erodibility factor is $A_t = 0$ for lithosol, and $A_t = 1$ for other soil types.

The size distribution of mineral dust aerosol is logarithmically divided into size bins ranging from diameter of 0.2 to 20 μm . The number of size bins is reduced from 10 of previous version (Tanaka et al. 2003) to 6, considering the computational cost of long-term climate simulations.