Characteristics of Asian Aerosols Transport simulated with a Regional Scale Chemical Transport Model during the ACE-Asia observation

Shinsuke Satake¹⁾, Itsushi Uno²⁾, Toshihiko Takemura²⁾, Gregory R. Carmichael³⁾,Youhua Tang³⁾,David Streets⁴⁾, Nobuo Sugimoto⁵⁾, Atsushi Shimizu⁵⁾, and Mitsuo Uematsu⁶⁾

Department of Earth System Science and Technology, Kyushu University, Kasuga Park 6-1, Kasuga, Japan
 Research Institute for Applied Mechanics, Kyushu University, Kasuga Park 6-1, Kasuga, Japan
 Center for Global and Regional Environmental Research, University of Iowa, Iowa, U.S.A.
 Decision and Information Science Division, Argonne National Laboratory, Argonne, Illinois, U.S.A.
 National Institute for Environmental Studies, Tsukuba, Ibaraki 305-8506 Japan
 Cecan Research Institute, University of Tokyo, Tokyo, Japan

Corresponding author address:

Shinsuke Satake, Department of Earth System Science and Technology, Kyushu University, Kasuga Park 6-1, Kasuga, Fukuoka 816-8580, Japan (e-mail: satake@riam.kyushu-u.ac.jp)

Abstract

The transport and optical thickness of tropospheric aerosols (dust, sulfate, carbonaceous aerosols, and sea salt) during the ACE-Asia intensive observation period (spring 2001) were simulated using a CFORS chemical transport model coupled with a regional meteorological model. Simulated aerosol fields were examined intensively with surface monitoring stations (PM₁₀, sulfate, and total carbonaceous aerosol), Mie Lidar, and satellite observation data. We found that CFORS aerosol fields agree with observations

and reproduced many observed characteristics including the strong correlation between dust and sulfate transports and latitudinal gradients of respective aerosol concentrations. Two-month (March and April) averaged aerosol optical thickness indicated that dust is the largest contributor at the latitudinal band between 35°N and 45°N: it contributed 15% of the total AOT (sum of dust, sulfate, carbonaceous aerosols, and sea salt) over the whole modeling domain. Moreover, AOT of sulfate and carbonaceous aerosols are predominant between the latitudes of 30°N and 40°N. They contributed 25% and 24% of total AOT, respectively. These dominant areas of aerosols were largely related to synoptic weather changes and demonstrated characteristics of aerosol transport. Aerosol horizontal fluxes were also analyzed. The main dust flow is eastward along the 45°N parallel and is located in the free atmosphere. Dominant parts of sulfate and carbonaceous horizontal transport flux are within the boundary layer. They are located as an outflow from the east coast of China. We found another flow of carbonaceous aerosols along about 30°N from Thailand and Laos to southern Japan in the free atmosphere. Regional budgets of tropospheric aerosols showed that total emissions were 105 Tg for dust, 8.3 TgSO₂ for sulfate (73%) from human activities and 27% from volcanic activities), and 3.07 Tg for carbonaceous aerosols. Dry deposition, gravitational settling, and northward outflow of dust accounted for 33%, 27%, and 14% of total emissions, respectively. Wet deposition, eastward outflow, and dry deposition of sulfate accounted for 33%, 27%, and 21%, respectively. Regarding carbonaceous aerosols, the outflow to the east has the highest fraction (49%), followed by dry deposition (16%) and the outflow to the north (14%).

AGU Index code number: 0305: Aerosols and particles, 0322: Constituent sources and sinks, 0345: Pollution--urban and regional, 0365: Troposphere–composition and

chemistry, 0368: Troposphere-constituent transport and chemistry

1. Introduction

Atmospheric aerosols from natural and anthropogenic sources have important effects on the global and regional climate system because they scatter and absorb solar and thermal radiation (direct effect), modify the cloud optical properties acting as cloud condensation nuclei (CCN) (indirect effect), and change atmospheric radiative budgets [e.g., *Twomey*, 1974; *Charlson et al.*, 1992; *Kaufman et al.*, 2002]. Previous aerosol model studies specifically addressed the evaluation of the role of tropospheric aerosols (i.e., mineral dust, sulfate, carbonaceous aerosols, and sea salt) for the global direct radiative forcing. However, the estimated radiative forcing for individual aerosol components differs greatly among the studies. Main reasons for these differences include the inhomogeneous distribution of aerosols caused by their short lifetimes in the atmosphere, wide range of size distributions, and complicated chemical and optical properties. Therefore, it is important to develop a high-resolution three-dimensional aerosol transport model to represent spatial distribution of tropospheric aerosols and to increase our knowledge for various aerosol properties based on a detailed comparison of intensive observation data.

Aerosol transport and studies of its radiative impact also showed high aerosol concentration and radiative impacts in several regions. *Liousse et al.* [1996], *Takemura et al.* [2000] and *Chin et al.* [2002] simulated the essential contribution of carbonaceous aerosols to the aerosol optical thickness (AOT) over biomass-burning regions (e.g., Africa, Brazil, and Southeast Asia) using the global model. Moreover, *Novakov et al.* [1997] reported a large amount of carbonaceous aerosols over industrial regions using results of

aircraft measurement. Carbonaceous aerosols roughly consist of black carbon, which is a primary absorbing aerosol, and organic carbon that mainly scatters solar radiation and is considered to be an important aerosol that controls the climate system through complicated interactions. Along with carbonaceous aerosols, sulfates, which have a negative radiative effect, are another predominant aerosol over industrial regions. Both developing countries (e.g., South and East Asia) and developed countries (e.g., eastern North America and Europe) have reported high anthropogenic sulfate aerosol concentrations because anthropogenic sulfates arise primarily from oxidation of sulfur dioxide generated by fossil fuel combustion. Aside from these anthropogenic sources, natural tropospheric aerosol sources are dispersed worldwide. *Tegen and Fung* [1995] and *Chin et al.* [2002] simulated that a large amount of dust is emitted into the atmosphere from Australia, northern Africa, the middle East, and Asia. However, it is difficult to accurately quantify the emission strength of the aerosol sources complexly dispersed, which consequently leads to the improper aerosol transport simulation. Therefore the aerosol transport study that focus on the regional scale is required.

Under that situation, East Asia includes not only biomass burning and industrial regions, but also desert regions, volcanoes (the main source of natural SO₂) and the ocean (the sea salt aerosol source). Especially, recent economic growth in East Asia has sharply increased the emission strength of anthropogenic aerosols. This region has become an important anthropogenic aerosol source in the world. Although a number of aerosol transport model studies have been performed, there is relatively little observation of physical, chemical and optical properties of aerosol focus on this region, which largely limits our knowledge for aerosol. Therefore, it is important to perform a simulation applying intensive observation over this region, and to perform comprehensive analysis based on observational facts to understand aerosols long-range transport, their transport processes and their radiative impacts.

The Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) was designed to elucidate physical, chemical and optical properties of aerosols and characteristics and mechanisms of aerosols transport, and radiative forcing [*Huebert et al.*, 2003]. Intensive observation was conducted using three aircraft, research vessels, surface stations and numerical models during the spring of 2001. The ACE-Asia intensive observation period corresponded to the period when aerosols are frequently transported from the Asian continent to the Northwest Pacific region. Many physical, chemical and optical properties of aerosols over the ACE-Asia regions such as Japan, Korea, and China were reported. Observations from these experiments improve the model capability and also help us understand more detailed characteristics of aerosol transport. However, it has remained difficult to capture a clear overview of aerosol distribution, its vertical profile, its transport processes and its radiative impact upon East Asia because of spatial and temporal limitations of observation.

This paper reports aerosol simulation results based on the CFORS model [*Uno et al.*, 2003b]. Here, CFORS was developed as a regional chemical transport model coupled with the regional meso-scale meteorological model to overcome above-mentioned difficulties and clearly illustrate aerosol transport processes and radiative impacts based on the hind-cast mode calculation to the ACE-Asia intensive observation period. This study focuses on producing an overview of aerosol transport and the radiative impact of tropospheric aerosols over East Asia. It is important to reproduce the continental outflow

from the Asian continent; therefore observation sites under the large-scale continental outflows are chosen for model validation. Moreover, we provide characteristics of aerosol transport and its radiative impacts for each aerosol type based on comparisons of model results with observations. Finally, our approach achieves a detailed analysis of aerosol transport patterns in East Asia and regional budgets for individual aerosols, as one unique point of this study. This approach addresses prominent features of aerosol transport, outflow pattern, its main transport altitude, and an overview of aerosol transport processes during spring 2001.

2. Outline of Chemical Transport Models

2.1 Regional Meteorological Model

The Regional Atmospheric Modeling System (RAMS) developed at Colorado State University [*Pielke et al.*, 1992] was used to simulate the regional scale meteorological field. The RAMS contains numerous options that make it amenable to use in a wide range of applications. This study used RAMS options identical to that for CFORS [*Uno et al.*, 2003b] (e.g., horizontal and vertical mixing coefficients are evaluated as by *Smagorinsky* [1962] and the level 2.5 turbulent closure of *Mellor and Yamada* [1974, 1982], respectively). RAMS includes Kuo-type cumulus parameterization to represent sub-grid scale convective cumulus and the Kessler-type microphysics model [*Walko et al.*, 1995]. The microphysics module is capable of simulating meso-scale cloud and precipitation. A more detailed general model description of RAMS and its capabilities have been given by *Pielke et al.* [1992].

The simulated domain encompassed East Asia; its rotated polar stereographic mapping

center was set at 25°N and 115°E. The Arakawa C grid system is employed [*Mesinger and Arakawa*, 1976] for the grid structure and all thermodynamic and moisture variables are defined at the center of a grid volume with velocity components staggered 1/2 of a grid space in their normal direction. The horizontal grid consists of 100 by 90 grid points, with resolution of 80 km. The vertical model domain is divided into 23 layers (the top level is 23 km) with the 23 non-uniform grids varying from 150 m thick at the surface to 1800 m thick at the top of the model layer in the terrain-following sigma-z coordinate. The sigma-z coordinate has an advantage over the sigma-p coordinate: the model levels reach the same absolute height above the ground at all times during a simulation. **Figure 1** shows the simulation domain. That domain includes many industrial and rural regions, dramatic variation of the land usage type, and topography, which sometimes alters the transport direction of aerosols.

The RAMS is a regional meteorological model and requires initial and boundary meteorological conditions. For long term simulations, a four-dimensional data analysis (FDDA) option using the nudging technique was included based on RAMS/ISAN (ISentropic ANalysis Package) output. The ISAN package converts the longitude-latitude grid with specified pressure coordinate data to the RAMS rotated polar stereographic terrain following the vertical coordinate system. European Center for Medium-range Weather Forecasts (ECMWF) global analysis data (6 h intervals with 1° resolution) was used as the input for RAMS/ISAN step. The ECMWF global meteorological data set includes wind velocity, temperature, geopotential height, and relative humidity at specified pressure levels of 1000, 925, 850, 700, 500, 400, 300, 250, 200, 150, 100, 70, 50, 30, and 10 hPa. This RAMS/ISAN output was used for model initialization and

continuous nudging calculation.

For the surface boundary condition, we used the dataset of 1-km resolution land cover characteristics produced by the U.S. Geological Survey (USGS) (based on AVHRR data obtained in 1992-1993), topography (10-min resolution) and sea surface temperature (SST) taken from the National Center for Atmospheric Research (NCAR) database (1° resolution).

2.2 Outline of Chemical Transport Model

a) General Descriptions

The chemical transport model was developed using RAMS' additional scalar transport options to simulate transport of major tropospheric aerosols, i.e., sulfate, carbonaceous (black and organic carbons), mineral dust, and sea salt aerosols, over East Asia. Therefore our chemical transport model is coupled with the RAMS so that all meteorological information calculated in RAMS is used directly by the chemical transport model at the same time step. This is a unique point in this study. This chemical transport model includes main aerosol transport processes such as emission, advection, diffusion, and deposition. The equation of aerosol mass conservation is

$$\rho_{air} \left\{ \frac{\partial}{\partial t} q_a + div (q_a \cdot \mathbf{v}) \right\} = F_{emis} + F_{dep} + F_{diff} \quad , \tag{1a}$$

where ρ_{air} is the air density, q_a is the aerosol mass mixing ratio and **v** is the wind velocity; F_{emis} , F_{dep} , and F_{diff} are the emission, deposition, and diffusion fluxes of aerosols, respectively. This study assumes the three types of removal processes of wet deposition, F_{wet} , dry deposition, F_{dry} , and gravitational settling, F_{grav} , that have the following relation:

$$F_{dep} = F_{wet} + F_{dry} + F_{grav}.$$
 (1b)

Wet deposition is divided into two processes: one is below-cloud scavenging whereby raindrops collide with aerosol particles because of different dropping velocities. The other is the in-cloud scavenging, by which aerosol particles in cloud water are removed from the atmosphere by precipitation. We address only below-cloud scavenging in this simulation. Parameterization of that process is based on the scheme of *Westphal et al.* [1988]. The mass flux with below-cloud scavenging is calculated as

$$F_{wet} = -\Lambda_{wash} \cdot q_a \cdot \rho_{air} \tag{2a}$$

$$\Lambda_{wash} = C_1 \cdot Pr^{C_2} , \qquad (2b)$$

where Λ_{wash} is the scavenging rate (1/h), C_1 and C_2 are dimensionless parameters, and Pr is the rainfall rate (mm/h) calculated in RAMS. **Table 1** summarizes deposition parameters for each aerosol and SO₂. C_1 and C_2 for mineral dust and sea salts have equivalent values to the number of bin because of consideration of aerosol size distribution (size bin).

The second removal process is the dry deposition. Dry deposition is caused by turbulent mixing with a flux F_{dry} given as

$$F_{dry} = -v_{dry} \cdot q_a \cdot \rho_{air} \cdot \Delta z_1, \qquad (3)$$

where v_{dry} is the dry deposition velocity and Δz_1 is the layer depth at the first model layer. The v_{dry} of each aerosol and SO₂ for the land and sea surface are set as shown in Table 1.

Gravitational settling is determined by Stokes' law; the mass flux is determined by gravitational settling calculated using the terminal velocity v_{stk} ,

$$v_{stk} = \frac{2\rho_{aero} \cdot r^2 \cdot g}{9\nu} , \qquad (4)$$

where ρ_{aero} is the particle density, r is the aerosol particle radius, g is the acceleration of

gravity, and *v* is the viscosity of the air. Then, mass flux with gravitational settling is given as

$$F_{grav} = -v_{stk} \cdot q_a \cdot \rho_{air} \cdot \Delta z_k , \qquad (5)$$

where Δz_k is the layer depth at the *k*th vertical layer. This process is considered for mineral dust and sea salts because the gravitational settling process is only an efficient removal process for large particles.

Cumulus convection plays an important role for vertical distribution of biomass burning sources in subtropical regions, such as southern China, Thailand, and Myanmar. Vertical redistribution of tracers by cumulus activities was treated as enhanced vertical turbulent diffusivity from the bottom to top of cumulus cloud layers as identified by the RAMS simplified Kuo-cumulus scheme. Its effect is expected mainly in subtropical zones because of frequent convection over them.

The CFORS simulation was performed with the hind-cast mode from 20 February to 30 April with an output interval of 3 h. This period contains the ACE-Asia intensive observation period; it corresponds to the period when biomass burning in Southeast Asia and dust storms in Chinese desert regions are relatively frequent. Moreover, we conducted sensitivity simulation experiments of (VolcOFF) without volcanic SO₂ emissions to investigate volcanic impact on the atmospheric environment and radiation.

b) Sulfur Dioxide and Sulfate

As mentioned previously in this paper, sulfate aerosols have a negative effect on the

global warming effect; therefore they have an important role in climate forcing. The main source of sulfate aerosols is oxidation of SO_2 . In the CFORS model, emissions of anthropogenic SO_2 and sulfate are calculated using the dataset taken from *Streets et al*. [2003], which includes industry, residential, transportation, power generation, and agriculture.

Figure 2 shows total emissions between 20 February and 30 April 2001 for a) anthropogenic sulfur, b) anthropogenic total carbonaceous aerosols (BC+OC), c) total carbonaceous aerosols (BC+OC) from open biomass burning, d) mineral dust, and e) sea salt, respectively. Anthropogenic sulfur emission is very high in eastern China, especially in major cities in China such as Beijing (39.9°N, 116.4°E) and Xi'an (34.3°N, 108.9°E) (Fig. 2a). For example, total anthropogenic sulfur emissions from the Beijing region and Xi'an region are about 170 Gg-SO₂/year and 140 Gg-SO₂/year, respectively. Volcanic eruptions are another important source of SO₂. Fujita et al. [1992] and Streets et al. [2003] have estimated emissions of volcanic SO₂. Emission of SO₂ from the Miyakejima volcano in Japan (34.08°N, 139.53°E) was renewed based on the latest observation taken from the Japan Meteorological Agency (JMA) because of the huge volcanic eruption since 8 July 2000. Consequently, estimated emission of volcanic SO₂ from the Miyakejima volcano is 10 Mg-SO₂/year. Emission from Miyakejima is highest among the East Asia countries. Its emission intensity is almost equivalent to 10 times the total anthropogenic SO₂ emission in Japan.

All of the anthropogenic SO_2 and sulfate aerosols (assuming 3% direct emission from SO_2) are released into the atmosphere boundary layer. Volcanic SO_2 emission is injected into a layer that corresponds to the elevation of the volcanoes. For oxidation of SO_2 , we

assume a linear conversion of 1%/hr throughout the simulation [*Uno et al.*, 2003b; *Carmichael et al.*, 2003].

c) Carbonaceous Aerosols

Carbonaceous aerosols consist of BC and OC: BC is produced by incomplete combustion of fossil and biomass fuels; OC, the most abundant component of the carbonaceous aerosol material, is introduced into the atmosphere either directly in particulate form or by gas-to-particle conversion of anthropogenic and biogenic precursor gases under a low vapor pressure level in the atmosphere. Emission intensity of BC and OC is also taken from the data set by *Streets et al.* [2003]. It is mainly high in several cities over East Asia (Fig 2b). In spring in East Asia, biomass burning is an especially important source of BC and OC. Their emission rates change day by day as a result of daily variation of biomass burning. Therefore, we assigned the daily averaged emission dataset taken from *Streets et al.* [2003]. The daily emission rates of BC and OC are estimated using the AVHRR satellite fire count and the TOMS AI dataset. Figure 2 shows that biomass emission of carbonaceous aerosols is especially high in southeastern Asia (Fig 2b). Total carbonaceous aerosol emissions in spring in East Asia comprise about 47% from human activities and 53% from biomass burning. In CFORS, we consider only primary OC emission.

d) Mineral Dust

Among the various atmospheric aerosols, mineral dust has the largest emission

intensity [*IPCC*, 1995]. The main sources of mineral dust are the desert and semi desert. Mineral dust is generally emitted into the atmosphere by wind processes. In this model, the mineral dust source region is defined as the desert and semi-desert area from the USGS land coverage data set. By this approach, most parts of the Gobi and Taklimakan Desert are allocated as source regions. In present calculation, the Loess Plateau and Nei Mongol's small desert regions are also defined as source regions [*Uno et al.*, 2003b]. The uplifted dust from these source regions is distributed uniformly within the mixing layer. The emission flux of mineral dust is calculated for a 12 bin mode (from 0.1 to 20 μ m radii; the effective radii are also used for calculation of mineral dust AOT). The dust emission flux *F_{emis}* is given according to the empirical relation as [*Gillette and Passi*, 1988]:

$$F_{emis,p} = C \cdot C_s \cdot C_W \cdot u_*^4 \left(1 - \frac{u_{*th}}{u_*} \right) \ (u_* > u_{*th}), \tag{7}$$

where *C* is a dimensional factor, C_s is the snow coefficient that is determined using the snow-cover data, C_W is the soil wetness coefficient that is determined using the soil wetness information calculated in RAMS, u_* is the frictional velocity and u_{*th} is the threshold frictional velocity. More detailed description of CFORS dust module can be found in *Uno et al.* [2003b]. As shown in Fig. 2d, most of mineral dust is emitted around the Taklimakan and Gobi. It is important to point out that dust emission from Taklimakan is remarkably strong when compared with that of the Gobi desert. Comparisons of lidar and TOMS AI observations with the CFORS results were performed after the ACE-Asia campaign. We found that the Taklimakan emission rate is underestimated. The dust module then, is tuned after several careful sensitivity simulations. The tuned dust module explains successfully why much dust is transported from the Taklimakan. More detailed

description for this will follow in a separate paper.

e) Sea Salt

Sea salt transport is also addressed by CFORS. Sea-salt aerosols are injected mainly into the atmosphere by bubble bursting at the ocean surface. Therefore sea-salt emissions from the ocean are highly dependent on the surface wind speed. We simulated sea-salt emission with a 12 bin mode (from 0.005 to 20.48 µm diameter). The emission flux of sea-salt aerosols, F_{emis} (N m⁻² s⁻¹), is calculated using the empirical relationship from *Gong et al.* [1997] and *Monahan et al.* [1986] as

$$\frac{dF}{dr} = 1.373U_{10}^{3.41}r^{-3}(1+0.057r^{1.05}) \times 10^{1.19e^{-\{B(x)\}^2}},$$
(8)

where $B_{(x)} = (0.380 - \log r)/0.65$, U_{10} m/s is the wind speed at the 10-m height and *r* is the sea salt particle radius. The emission flux of sea-salt aerosols is greatest at the east side of Japan because of the high surface wind speed (Fig. 2e). Once sea salt aerosols are released into the atmosphere, they are subject to transport, dry and wet deposition, and gravitational settling. Sea salt aerosols transport processes are also simulated with a 2 bin mode (fine mode, from 0.005 to 2.56 µm diameter and coarse particle mode, from 2.56 to 20.48 µm diameter) in this model.

2.3 Aerosol optical properties

Aerosol Optical Thickness (AOT) for sulfate, black carbon, organic carbon, mineral dust, sea salt and total value at a wavelength 550 nm are calculated based on the *Takemura et al.* [2000] to illustrate the impact of aerosols on atmospheric radiation. The relationship between AOT and the mixing ratio of mineral dust and sea salt aerosols is given as

$$\tau = \sum_{i} \left(\frac{3}{4} \sum_{k=1}^{k_{\text{max}}} \frac{Q_{ext}(i) \cdot q_a(i,k) \cdot \Delta p_k}{\rho \cdot r_{eff}(i) \cdot g} \right),$$
(9)

where τ is the aerosols optical thickness, Q_{ext} is the extinction efficiency factor as a function of the particle size bin *i*, q_a is the aerosol mixing ratio, Δp_k is the layer thickness by pressure between the *k*th vertical layer and *k*+1th vertical layer in our model, ρ is the particle density, and r_{eff} is the effective radius of the aerosol. **Table 2** lists optical parameters of mineral dust and sea salt used in the simulation. In evaluating AOT for mineral dust and sea salt, the same particle size and effective radius are used as in the transport processes. The radii of the sulfate and organic carbon particles are sensitive to ambient relative humidity, so we take hygroscopic growth of their aerosols into account in the same manner as *Takemura et al.* [2001]. In this method, lognormal size distribution is assumed and the mass extinction coefficient of each particle (σ) is estimated as a function of relative humidity. The relationship between AOT and the mixing ratio of sulfate and carbonaceous aerosols is given as

$$\tau = \sum_{k=1}^{k_{\text{max}}} \frac{\sigma(RH(k))q_a(k)\Delta p_k}{g} .$$
(10)

Table 3 also shows optical parameters of sulfate and carbonaceous aerosols. Each particle size under the relative humidity of 95% is nearly twice the size of that under the dry mass condition. The aerosol extinction coefficient and single scattering albedo are also calculated with the evaluated AOT.

3 Comparisons With Observations From ACE-Asia Field Experiments

This section will show the model performances during ACE-Asia observation. Lidar observation at Beijing, China and Nagasaki, Japan are used first to examine the simulated

dust field. Particularly the onset timing and its vertical distribution are compared with the model. The interaction between dust and anthropogenic air pollution is also discussed based on the lidar depolarization signal. Secondly, surface observation data of PM_{10} , sulfate and total carbonaceous aerosols (TC) are targeted to show the typical time variation during large-scale continental outflow from the Asian continent. Here, surface observation sites from the remote island are selected to avoid unnecessary contamination from local pollution. Finally, we will compare the model results with TOMS Aerosol Index data. Time variation of TOMS AI and CFORS aerosol AOT will be discussed.

A detailed analysis based on obtained information from these experiments and CFORS results has been presented in ACE-Asia heavy dust storm [*Seinfeld et al.*, 2003], surface black carbon analysis [*Uno et al.*, 2003a; *Matsumoto et al.*, 2003], Mie scattering lidar data analysis [*Sugimoto et al.*, 2002; *Shimizu et al.*, 2003], and *Sugimoto et al.* [2002] for analysis of the dust and anthropogenic aerosol plumes in the Northwest Pacific region.

3.1 Vertical Profiles of Dust and Air Pollution by Lidar and CFORS

The National Institute for Environmental Studies (NIES) has made a continuous observation of atmospheric aerosols at Tsukuba, Nagasaki, and Beijing with a continuous polarization Mie-scattering lidar system during the ACE-Asia 2001 observation. The aerosol extinction coefficient is calculated by the method proposed by *Fernald* [1984] and the boundary condition of the calculation was set at 6 km. The extinction coefficient was split to dust (non-spherical aerosol) and non-dust (spherical aerosols such as air pollution) fractions based on the aerosol depolarization ratio. Sugimoto et al. [2002] and Shimizu et

al. [2003] described details of lidar observation and splitting method details.

Figure 3 shows the observed time-height cross section of aerosol extinction coefficients and simulated ones at Beijing, China from 1 March to 30 April 2001. Beijing is located at 39.9°N and 116.3°E and is very close to the dust source area (see Fig. 1). In addition, Beijing city itself is a huge urban air pollution source and is also surrounded by industrial areas in China. Therefore, high anthropogenic air pollutant concentration is expected and proper discrimination of dust and anthropogenic air pollutants within the model is one study objective.

Lidar observation captured several high dust extinction levels. For example, Fig. 3a shows that dense dust layers were transported almost every 10 days. The model also simulated all high extinction coefficients except for arrivals of dust on day 70, shown in Fig. 3c. Simulated depths of dust layers occasionally reached 2-3 km indicating good agreement with lidar observation. For dust on day 100, the model shows arrival of a vertically huge dust cloud. This huge dust cloud is referred as the 'Perfect Dust Storm (PDS)' during the ACE-Asia observation. It was transported to North America while maintaining its relatively dense dust concentration. Our model also successfully captured this PDS. Details of this PDS have been already reported [*Gong et al.*, 2003; *Liu et al.*, 2003].

For anthropogenic pollution, several high values were also observed on day 70, after 90 days, and before 100, 110, and 120 days. The model explains that sulfate aerosols largely contribute to these peaks. Both regions of high extinction coefficients by lidar and the model are located within the lower boundary layer. These vertical profiles are understandable because the air pollutants are emitted mainly from the surface level.

Careful examination of the interaction between dust and air pollution reveals an interesting relationship between dust and sulfate. Figure 3 shows that high extinction level regions of spherical aerosols intermittently appeared through spring 2001. They correspond to a period in which larger dust extinction coefficients are observed. Model results also capture this tendency. Therefore we infer that dust and sulfate are strongly correlated.

In general, the model explains most dust and sulfate events over Beijing, but we can also see that model simulation cannot reproduce the high extinction profile in some cases. Typical examples are found around 70 days and 90 days in Julian days (see Fig. 3). The TOMS Aerosol Index data did not retrieve even these high concentrations (not shown in the figure). One reason for this is that the lidar signal captured the Beijing area's local air pollutants. Horizontal grid size of our transport model is 80 km, so this grid resolution cannot capture the local air pollutant. More detailed comparison between lidar and CFORS output will be discussed by *Uno et al.* [2003c].

Figure 4 shows observed and simulated two-month-averaged vertical profiles of the total extinction coefficient at Beijing and Nagasaki. The dashed lines indicate the averaged profile from lidar observation; solid lines indicate simulated total extinction coefficients. The fractions from dust and sulfate are shaded for clear illustration. Contributions of sea salts are negligible, so we do not distinguish the contribution of sea salt in this figure. For both Beijing and Nagasaki, the lidar observed extinction coefficient has a sharp peak near the ground. Its vertical profile sharply decreases up to the altitude of about 3 km. Above this level, the profile becomes almost constant. The vertical profile of

the simulated extinction coefficient has a small peak altitude at about 1 km and continuously decreases with altitude after the peak.

This underestimation of model extinction coefficients at the surface region and the difference gradients at the higher levels occur because the observed extinction coefficient is sometimes affected by local air pollution and by uncertain cloud screening. However, the temporal averaged vertical profile of modeled and observed extinction coefficients are closely correlated with coefficients of 0.84 at Beijing and 0.94 at Nagasaki. We can see that the model reproduces characteristics of the observed vertical profile.

Relative contributions to the total extinction coefficient are also estimated from the model results. Beijing was found to be 59% from dust, 28% from sulfate, and 13% from carbonaceous aerosols. For Nagasaki, 32% was from dust, 40% from sulfate, 27% from carbonaceous aerosols, and 1% from sea salt. Dust at Beijing has a stronger impact on the atmospheric radiation than sulfate and carbonaceous aerosols. This is true because Beijing is located near dust source regions, as mentioned above; also, large amounts of dust are often transported during the spring [*Liu et al.*, 2003; *Gong et al.*, 2003; *Shimizu et al.* 2003]. *Shimizu et al.* [2003] also reported that the dust has a strong contribution to observed extinction, which is above 60% averaged between March and May 2001. We can see that the model captures this tendency in general. However, we must note that sulfate and carbonaceous aerosols at Beijing are also important in annual cases because many strong anthropogenic sources are located near Beijing. They may cause larger radiative impact than that of dust through a year. For the Nagasaki case, sulfate has the largest fraction. Nagasaki is located in western Japan and its air quality is frequently affected by the continental outflow. A typical example is the high extinction level on days 78 and 79

[*Uno et al.*, 2003b]. On these days, high-pressure system moved eastward from southeastern China (near Shanghai) to the southern side of Japan. Strong southwest continental outflow at the backside of high pressure-system is a typical feature during spring; it exports an air mass that contains many anthropogenic pollutants to Japan. Therefore we find that the strong contribution caused by sulfate at Nagasaki occurred as a result of these continental outflows.

3.2 Time Variation of Tropospheric Aerosols by Ground Observation and CFORS

Figures 5, 6, and 7 show observed and simulated time variation of surface concentration for particulate matter (PM_{10}), sulfate and total carbon (TC) respectively. Table 4 shows their mean value. In Fig. 6, the dotted line indicates the simulated sulfate concentration without volcanic emission (experiment of **VolcOFF**). It must be noted that all dates are Julian days and all times are Japan Standard Time (JST) in these figures. For PM_{10} , we compare the summation of the simulated aerosol concentrations (PM_{model} , i.e., sum of dust, sulfate, BC, OC, and sea salts). Therein, five remote islands (locations are shown by the triangles in Fig. 1) are selected because the local air pollution sources are almost absent. It is therefore expected that observed surface concentrations remarkably reflect transported aerosol concentration. Comparisons at these islands also yield valuable information indicating whether or not the model simulates proper aerosol transport. Measurements at Rishiri (sulfate and TC), Sado (TC), and Hachijo (sulfate and TC) are taken from VMAP measurement networks [*Matsumoto et al.*, 2003]. Amami (sulfate and TC) observation data are taken from *Ohta et al.* [2003]. Gosan (PM₁₀ and sulfate) and

Jejyu Korea are by the National Institute for Environmental Research, Korea; PM_{10} observation data at Rishiri, Sado, and Amami are taken from the East Asia Acid rain monitoring network (EANET).

Among these five islands, Rishiri, Sado and Hachijo are located almost exactly along the line of 140°E, while Amami and Gosan are near the line of 130°E (Fig. 1). It should be noted that these chosen islands are not realized as land in our model because their areas are much smaller than our model grid resolution, meaning that the surface concentration extracted from simulation results differs from those at the real surface level. Therefore, vertically averaged concentrations (below ca. 400 m) are used for comparison with observations.

a) PM_{10} and PM_{model}

In Fig. 5, PM_{10} concentration (aerosols less than 10 µm in diameter) shows a rapid increase, especially when mineral dusts are transported. A salient feature during spring 2001 is that a huge dust storm (PDS) was observed between 96-104 days over the model region. According to *Uno et al.* [2003b], this dust storm can be divided mainly into two major outbreaks of dust. The first outbreak is from the Gobi Desert and Loess Plateau; this dust was transported at relatively high latitude (about 40°N) and arrived at Rishiri on day 99. On day 98, a second dust storm was generated by the arrival of a low-pressure system over Mongolia. One feature of the second is that it transported at lower latitude than the first one. The second storm, in turn, reached Gosan on day 101, Amami on day 102, and Sado on day 103. We can see their features clearly in Fig. 5a. The overall correlation coefficient between the PM_{model} and PM₁₀ is 0.67, indicating that the model captures characteristics of PM₁₀ time variations at the four sites.

Comparisons between the sites also provide important information. One important feature is that the observed time variation at Sado is relatively similar to that at Amami, which differs from that at Rishiri. A characteristic of the time variation at Rishiri shows a high PM₁₀ level during the PDS period, while observations at Sado and Amami detect high levels not only during the PDS period, but also on days 96 and 114-115. Gosan also shows a concentration peak at 114-115 days. Figure 1 shows that Rishiri, Sado, and Amami are located in the northern, central, and southern parts of Japan, respectively. These differences among observations suggest that these dusts are transported at low latitude.

More detailed comparisons show several discrepancies between PM_{model} and PM_{10} . For example, although observation shows high PM_{10} values on day 111 at Gosan and on days 95 and 114-115 at the Sado and Amami, PM_{model} does not show them. *Chin et al.* [2003] reported that relatively high Ca^{2+} level are observed over the Yellow Sea and the Japan Sea around these days, and their source is mainly a desert region located just north of Shenyang city (41.8°N and 123.5°E). In addition, the weather report produced by the Japan Weather Agency (JWA) reported that small-scale low-pressure systems passed over that region just before these high PM_{10} values were shown; TOMS AI detected relatively high AI values over its region at that time. These facts indicate that these PM10 values are caused mainly by dust emitted from the desert near the Shenyang city and that the model tends to underpredict dust emission from that region.

It is important to point out that model underpredicts PM_{10} value at Rishiri during a PDS period. At the same time, the model shows the presence of a dense dust layer around the

boundary layer level. Therefore this underestimation may be caused by difference in transport altitudes between the model and observations. Another possible explanation is the underestimation of dust emission strength in several desert regions. As shown in Fig. 2d, main dust sources in this model are the Taklimakan desert, the Gobi desert, and the Loess Plateau, which is a common characteristic with other dust modeling studies [e.g., *Wang et al.*, 2000; *Gong et al.*, 2003; *Xuan et al.*, 2002]. However, we found the emission strength of the Gobi desert and Loess Plateau to be relatively lower than in other models. *Shao et al.* [2001] also reported from numerous experimental results that saltation, which represents the horizontal dust emission flux, is one of the most important processes in dust emission; a large amount of dust is emitted into the atmosphere by its process. These indicate that further improvement of the dust emission scheme is needed.

Table 4 shows a significant difference of PM_{10} concentration among three Japanese sites, which is largest between Amami and Rishiri (about 7 µg/m³). This result indicates that PM_{10} is transported on a much larger scale than Japan, on average. Another important feature is that all the mean PM_{model} values are lower than the mean PM_{10} . The largest difference between the model and observation is seen at Gosan: the mean PM_{10} is about twice as large as the mean PM_{model} , whereas about 10 (µg/m³) differences at Amami and Sado are seen among the Japanese sites. These differences are mainly attributable to underestimation of the background level of PM_{10} and of high PM_{10} level on days 96 and 115 at Amami and Sado, as mentioned above.

b) Sulfate and Carbonaceous Aerosols

Regarding sulfate and carbonaceous aerosols, Figs. 6 and 7 show comparisons between

observations and the model results. These figures show that model results agree well with the observations and depict the absolute concentration level and its time variations. Another important feature is that the observed and simulated sulfate and TC at the Hachijo exhibit similar temporal variation with correlation coefficients of 0.72 for model and 0.80 for observation. This feature is also seen at the Rishiri and Amami (with correlation coefficients of 0.76 and 0.36 for the model and 0.47 and 0.54 for observation, respectively). This is because the sulfur emission distribution is closely related to the TC emission, as shown in the Fig. 2. The sulfur and TC emitted from the same region are consequently carried by the same air mass. Several previous aerosol transport studies have reported that the air mass transported from the East Asia simultaneously includes high concentrations of sulfate and carbonaceous aerosols [e.g., *Kaneyasu et al.*, 2000a, 2000b; *Matumoto et al.*, 2002; *Uno et al.*, 2003a].

Detailed analysis of sulfate and BC study at Rishiri and Hachijo has been presented in *Uno et al.*, [2003a, 2003b]. They suggested that modeled wet deposition process of BC and sulfate may be weak with the simulated the precipitation amount. In addition, the sulfate and TC surface concentrations are consequently overestimated at the Rishiri between the Julian days of 95-96, 98-100, and 101-102 and the Hachijo before day 95.

The results of the VolcOFF experiment are also shown in Table 4 and Fig. 6 (by dotted line) to illustrate the impact of the volcanic and the anthropogenic sulfate. As a whole, the strong contribution caused by anthropogenic sulfate is seen in the Table 4. However, the volcanic sulfate occasionally exerts a large influence. For example, the observation captures the sharp peaks at Gosan, Amami, and Hachijo between Julian days of 100-104. During these periods, the weather report presented by the Japan Weather Agency (JWA)

and the RAMS model show the traveling low-pressure systems that passed over southern Japan. The VolcOFF result shows that a sharp increase of the sulfate concentration level occurred as a result of the large scale westerly transport of the Miyakejima volcanic mass by their cyclonic flows [*Uno et al.*, 2003b; *Zhang et al.*, 2003]. Among these four sites, Gosan and Amami are far from the Miyakejima volcano (ca. 1200 km distant), and this fact demonstrates the importance of the large-scale volcanic sulfate under special synoptic weather conditions. *Zhang et al.* [2003] discussed this case study in more detail.

Table 4 shows that simulated sulfate and TC overpredict the observed level at the Hachijo. Obvious overpredictions are visible between Julian days of 95-97 and after Julian days of 110 as shown in Fig. 6. On the other hand, VolcOFF results clearly capture the observed sulfate level during these periods (Fig. 6). This result suggests that these sulfate overestimations are, together with above mentioned wet deposition process of sulfate, caused by transport of the volcanic sulfate. As shown in *Uno et al.* [2003b], the wind direction is always from north to south just before these overestimations, implying that these overestimations occur when the air masses that include the Miyake volcanic sulfate and the TC emitted from Japan are coming from the northern side of Hachijo. The horizontal distances separating Hachijo - Miyake volcano and Hachijo – Honshu Island, Japan are roughly 120 km and 300 km, respectively. The horizontal grid size of 80 km may be too large to simulate precisely the influence of Miyake volcanic sulfate and Japanese TC on Hachijo. Therefore detailed analysis of the Hachijo site simulation requires finer resolution.

In contrast to the Hachijo, the observed TC at Sado greatly exceeds the simulated TC on days 96-102, 108-110, and 116-120 (Fig. 7), which has already been mentioned in *Uno et*

al. [2003b]. One explanation for this is underestimation of Japanese domestic emission intensity as a result of local biomass burning before rice planting.

Along the 140°E meridian, the distribution of the monthly mean surface concentration both for the observed and simulated TC has a peak at Sado Island, as shown in Table 4. This indicates the presence of a north-south gradient of TC. According to the observation, its gradient increases from the Northern Japan to Central Japan and sharply decreases from Central Japan to Southern Japan. Model results also capture this general tendency; however, the concentration gradient is about 80% smaller than the observation. Presence of the latitudinal gradient for TC concentration also leads us to expect it for sulfate because temporal variations of sulfate and the TC have a close relationship as mentioned above. The monthly mean observed concentration for sulfate shows the highest sulfate level at the Gosan. The model also simulates higher levels at Gosan and Sado than those at Amami and Hachijo (Table 4). Westerly winds are prominent around Japan during the spring, so air masses including anthropogenic air pollutants are often transported to central Japan and the Japan Sea regions. This reflects the stronger contribution of continental outflow in the Japan Sea side (Gosan and Sado) than on the Pacific Ocean side (Amami and Hachijo). Consequently, it indicates the presence of a latitudinal gradient for sulfate surface concentration. We will make a further examination in section 3.3 to more clearly elucidate this latitudinal gradient and explain more detailed characteristics of tropospheric aerosols.

3.3 Horizontal Distribution of Boundary Layer Averaged Aerosol Concentration

Figure 8 shows the two-month (March and April) and vertically (below the 1000 m) averaged concentration field for individual aerosols by the line and VolcOFF by the dashed line, respectively. As a whole, maximum values for each aerosol are located near their source regions. For example, mineral dust shows its highest level at the inland desert area of China. High concentrations of sulfate and carbonaceous aerosol are located from the eastern coast of China to south China. One high TC region is Southeast Asia, reflecting the strong contribution from biomass burning through the spring. These results indicate that serious air pollution originates inside China and affects air quality in Japan and over the Pacific Ocean by long-range transport [*Takemura et al.*, 2002]. Finally, high sea salt aerosol regions are located at high latitude, resulting from high emission rates caused by strong surface winds at these locations.

This figure clearly depicts the presence of the latitudinal gradient of sulfate, which has a ridge extending from southern China to central Japan and mainly dominates the central part of the modeling domain (between 25°N and 45°N) (Fig. 8b). Differences between the VolcOFF and the CNTL (standard) simulations exhibit the impact caused by the volcanic sulfate, which is visible over the northern side of Japan and Westside of Japan Sea. Sensitivity analysis shows that the contribution of volcanic sulfate is about 13% of the total sulfate concentration, implying that sulfate from anthropogenic emissions is dominant within the boundary layer.

The averaged TC concentration field has a large gradient between industrial and remote regions. Its distribution, and sulfate's, are dominant between 30°N and 45°N. This typical distribution is consistent with results reported by *Liousse et al.* [1996]. On the other hand, the dust field is located mainly in the northern part of the modeling domain (between 30°N)

and 45°N); its concentration decreases exponentially proportional to downwind distances as reported by *Tsunogai et al.* [1985].

3.4 Comparison of TOMS Satellite Observation With the CFORS AOT Field

Total Ozone Mapping Spectrometer Aerosol Index (TOMS AI) provides effective information to understand aerosol transport mechanisms and paths. Over both land and ocean surfaces, TOMS AI operates at UV wavelength and detects UV-absorbing aerosols such as mineral dust, volcanic ash, and soot from biomass burning. It must be noted that TOMS AI has the observational limitation of detecting aerosols within 1-2 km above the surface [*Herman et al.*, 1997]. Therefore AI values are not always proportional to the total AOT of the air column.

Figure 9 depicts the simulated optical thickness of tropospheric aerosols and TOMS AI over the gray-colored regions in Fig. 1. Model results are shown for 1200 JST because TOMS observation times are scheduled at the local solar noon. **Table 5** summarizes relative contributions from individual aerosols for total AOT over the simulated period. In all, four regions are specified: NC (Northeastern China), NJ (Northern Japan), JS (Japan Sea), and SJ (Southern Japan). Model aerosol AOT and TOMS AI are averaged over the regions in Fig. 1.

Overall, Fig. 9 indicates the close relationship between the simulated dust and TC and high AI values. For example, AI values are relatively higher in the northern side; the model captures this tendency. In addition, three large dust storms occurred in Northeastern China (NC), Northern Japan (NJ) and Japan Sea (JS) during spring 2001 (between Julian days of 60-70, 70-85 and 95-115). Both TOMS AI and modeled total AOT show high values on these days.

Figure 9 also shows several interesting features for aerosol transport. The first feature is that dust AOT is high when sulfate has a large AOT fraction at the NC, NJ, and JS. In section 3.1, we mentioned this relationship at Beijing. However, we can also see its relationship at the NJ and JS regions except for Julian days of 85-95. This result provides important evidence of the correlation between dust and sulfate transports. Another feature is that a large amount of dust and carbonaceous aerosols over the NC, NJ and JS, and SJ, respectively, are transported almost daily. In addition, its tendency for dust is stronger as the spring proceeds. Indeed, we investigated the number of dust AOT exceeding 0.1 in the NJ and JS using model results: results were 8 d for March and 15 d for April at NJ, and 9 d for March and 20 d for April at JS. These suggest that dust transport during April is more frequent when compared with that during March.

More detailed analysis of model results also gives some important information concerning aerosol radiative impact. The modeled AOT shows that most of the observed high TOMS AI values for the NC region are associated with dust transport. This value is prominent compared with the dust contribution at other sites. In contrast to dust, carbonaceous aerosols show the strongest contribution in the Southern Japan (SJ) region, whereas the sulfate fraction is highest at the Japan Sea. Therefore we can find that dust AOT has a strong fraction to the total AOT in the northern side of the model domain as well as the boundary layer case. Regarding sulfate, its fraction is almost even throughout the Japanese three regions. It is also important that carbonaceous aerosols have an opposite gradient to the dust explained in Table 5.

3.5 Horizontal Distribution of the Averaged AOT Field

Figure 10 shows the two month-averaged AOT from each aerosol type and the total AOT. The dashed line in Fig. 10b indicates results of VolcOFF. The same contour level as that of total AOT is used in this figure to represent each aerosol fraction to the total AOT. As seen in Fig. 10, the dust AOT has almost identical distribution as that within the boundary layer (Figs. 8a and 10a). This means that dust distribution dominates over the northern part of modeling domain in the free atmosphere as well because AOT is an integrated quantity from surface to the top of the model layer. However, we can see slight differences between their distributions. Compared with boundary layer dust concentration, a ridge of dust AOT is located slightly to the north. Wind speed is stronger and its direction is more even in the free atmosphere because reduced effects of the surface friction. Therefore most of the dust particles that are lifted above the boundary layer level are transported directly to the east by the influence of the prevailing westerly winds. These differences between the two distributions suggest that a large amount of dust is transported in the free atmosphere and that the dust transport directions are slightly shifted to the north with increasing altitude. These phenomena result in a larger latitudinal gradient for dust AOT between the JS and SJ regions. Section 4.2 presents a more detailed structure of transport flux.

Along with dust, sulfate AOT has almost identical distribution to that within the boundary layer (Figs. 8b and 10b). Its main ridge extends from southeastern China in a northeasterly direction. It is an important feature that an industrialized region such as the

eastern coast of China is not included in the highest AOT region. This is because SO_2 is transported to other regions where it transforms into sulfate. Another reason is that calculations of sulfate and carbonaceous AOT are very sensitive to atmospheric relative humidity (Tables 2 and 3); a meso-scale model RAMS, on the average, simulates the high relative humidity in the Southern China. It is noteworthy that sulfate AOT has the highest fraction at the JS region (Table 5). Because the JS region is not only located in the downwind area of the Asian continental outflow, but also located near active volcanoes, this region is influenced daily by the continental outflow and sometimes by volcanic sulfates [*Xiao et al.*, 1997]. As shown in Figs. 10b and 10f, impacts occurring as a result of volcanic sulfate raise the sulfate AOT over Japan. The ratio of volcanic sulfate to the standard AOT is about 16% for the entire model domain, expressing its stronger influence in the free atmosphere than in the boundary layer.

The highest carbonaceous AOT region is located from Southern China (as a result of high relative humidity) to Southeast Asia (as a result of intensive biomass emission). It results in the same region as the boundary layers (Figs. 8c and 10c), but its distribution differs greatly from that within the boundary layer. It is predominant around the line of 30°N, in contrast to dust and sulfate. This fact indicates that the carbonaceous transport direction is mainly eastward in the free atmosphere. *Kaneyasu et al.* [2000b] reported that a great increase in the black carbon level was observed routinely around the 30°N in research cruises of 1993 to 1996. This observational fact is consistent with our results.

For sea salt aerosols, high AOT regions are the South China Sea and the east side of Japan as a result of high surface winds. However, compared to other aerosols, model

results show that sea salt AOT is relatively low because the mean surface concentration of sea salt aerosols is lower than that of mineral dust; also, sea salt aerosols do not attenuate sunlight as effectively as sulfate and carbonaceous aerosols (Tables 2 and 3).

Figure 10e shows that the total AOT has a very complex distribution. However, the model analyses presented above indicated that this complex distribution is composed of different meteorological conditions, aerosol source regions, emission strengths, and transport processes. Consequently, dust is the most predominant aerosol over the northern part of the modeling domain; sulfate also dominates from the industrialized regions in China to the eastern side of Japan. Carbonaceous aerosols caused by intensive biomass burning events are the most important aerosol type over Southeast Asia. They are dominant from that region to the southern side of Japan. These characteristics are confirmed by latitudinal variations of averaged AOT for each component at 130°E shown in **Fig. 11**. According to this, AOT for dust, sulfate, and carbonaceous aerosols have peaks at 41°N, 38°N, and 27°N, respectively. Carbonaceous AOT is higher in the southern part of the modeling domain, whereas the mineral dust fraction is higher in the northern domain. Sulfate's high AOT region is located mainly between 25°N and 45°N.

Finally, mean contributions of each component to total AOT were evaluated. Contributions to total AOT were 36% for carbonaceous aerosols, 25% for sulfate, 24% for dust, and 15% for sea salt aerosols. Importantly, carbonaceous aerosols and sulfate of which the emissions are almost dominated by the human activity contribute largely to total AOT. Consequently, they exert a strong impact on the atmospheric environment and its radiation field.

4. Asian Scale Transport and Regional Budgets

The preceding data illustrated the idea that each tropospheric aerosol has a characteristic distribution within the boundary layer and in the free atmosphere. In addition, their distributions are closely related to different meteorological conditions, aerosol source regions, emission strengths, and transport processes. Especially, variation of the surrounding wind pattern plays an important role in transport from the Asian continent to downwind countries in East Asia. These distributions further demonstrate their average transport. Our approach ultimately yielded an overview of tropospheric aerosol transport in East Asia during spring 2001.

4.1. Wind Fields During Spring 2001

Spring in East Asia corresponds to the period of transition between the winter and summer monsoons. Main features of the winter monsoon in East Asia are that very strong high-pressure and low-pressure systems form over the Mongolia and Siberia (Siberian High) and the northern Pacific Ocean (Aleutian Low), respectively. Thereby, a strong northerly wind is produced and cold air masses from the north are brought to the area around Japan. As the spring comes, the strength of the Siberian High decays while a high-pressure system is established over the central Pacific Ocean, bringing warmer tropical air from the south. Figure 12 shows the simulated two-month averaged sea level pressure (shown as a line) and wind fields (shown as vectors). In this figure, wind fields are also vertically averaged from the surface to the boundary layer level (z_* = 1000 m).

During March and April, the Pacific anti-cyclone increasingly develops and frequency of the incursion of the warm air mass to the north increases. Therefore, this anti-cyclogenesis greatly alters wind fields around Japan. Presence of the divergence zone over the southern side of China is also visible (marked as D in Fig. 12). This results mainly from the contact of air masses driven by the winter monsoon with the marine tropical air masses from Pacific. It becomes more marked as the winter monsoon weakens and the summer monsoon builds [*Bey et al.*, 2001].

Meteorological conditions in spring in East Asia are also characterized by large traveling high- and low-pressure systems. They appear cyclically over the Asian continent, slowly moving eastward to pass over the Japan's main island. Such passages of pressure systems frequently cause the strong continental outflow containing air pollutants. In addition, low-pressure systems sometimes bring bad weather, and entrainment of dust occurs behind the cold front. Entrained dust is carried from source regions to the remote region together with the eastward movement of the low-pressure systems. These traveling pressure systems play an important role in long-range transport of aerosol. Thereby, the distinction of aerosol transport over the East Asia was complicated further. Numerous reports have detailed the importance of such pressure systems and their impact on air quality [*Xiao et al.*, 1997; *Bey et al.*, 2001; *Uno et al.*, 2001; *Gong et al.*, 2003; *Liu et al.*, 2003].

4.2. Aerosol Transport Patterns

Figure 13 shows the two-month averaged aerosol transport flux fields. The left column

and middle column in Fig. 13 show the vertically averaged horizontal flux (by vectors) of dust, sulfate and carbonaceous aerosols and their magnitudes (by color) for the boundary layer and for the total column, respectively. Here the horizontal transport flux (*HFLX_x* and *HFLX_y*) are calculated as

$$HFLX_{x} = \frac{1}{T} \int^{T} u \times C_{aero} dt , \ HFLX_{y} = \frac{1}{T} \int^{T} v \times C_{aero} dt , \qquad (11)$$

where u and v are the wind speed in the x, y directions, C_{aero} is the aerosol concentration, and T is the averaging period. Thus, the transport flux (*HFLX*) magnitude is given as

$$HFLX = \sqrt{\left(HFLX_{x}\right)^{2} + \left(HFLX_{y}\right)^{2}} .$$
(12)

The mean cross section of the longitudinal transport from the west to east across longitude 130°E is also shown in Fig. 13 in the right column.

Dust horizontal fluxes are highest in the Chinese desert regions because dusts are emitted into the atmosphere by high surface winds. The main export pathway for dust is to the Pacific along the 45°N parallel in westerly flow (middle column in Fig. 13). The latitudinal cross section of the horizontal flux shows that the region of highest dust flux ranges from 2 to 4 km and is centered at 43°N, indicating that the active height for dust transport is the lower free atmosphere. *Iwasaka et al.* [1988] reported that the altitudinal range of mineral dust transport over Japan has been observed to be from 2 to 6 km in most cases. This fact concurs with our results. Another feature for dust transport is that surface level dust shows two main transport directions from their source regions: eastward transport and southeastward transport. In the latitudinal cross section at 130°E, this characteristic is also confirmed, as shown by R1 and R2. This occurs because the mean wind direction from the Yellow Sea to the Japan Sea shifts northward by enhancement of

southerly inflows of air as a result of the Pacific anti-cyclogenesis during the simulation period (Fig. 12). Thus, these two ridges indicate representative transport directions for March and April: southeastward during March and eastward during April. However, dust transport is unified by the strong westerly above the boundary layer.

Surface horizontal fluxes for sulfate and carbonaceous aerosols show a clockwise flow pattern over central China. This outflow is closely related to the divergence zone in central China, as shown in Fig. 12. The north edge of its divergence zone is located in northeastern China and its south edge is located around southern China and Southeast Asia. The north edge location also corresponds to the high anthropogenic emission area, producing a strong westerly outflow pattern associated with anthropogenic pollutants (left column in Fig. 13). We can clearly confirm the presence of its outflow for the latitudinal cross section at 130°E. We also find that its outflow is mainly dominant within the boundary layer. Kim et al. [2001] found that sulfur transport to the Yellow Sea is confined mainly in the boundary layer, which concurs with our analysis. The divergence zone southern edge also plays an important role in aerosol transport. It constrains the Asian continental outflow because this edge comprises easterly flows from the Pacific region. Therefore we conclude that the divergent wind pattern in central China is an important factor for aerosol transport within the boundary layer, driving a strong outflow containing anthropogenic pollutants at the northeastern China, while driving an inflow from the Pacific at more southerly latitudes.

For the total column averaged horizontal carbonaceous flux field, a prominent westerly flow pattern is shown at the southern latitude in the modeling domain. This contrasts sharply with the boundary layer case. Its outflow corresponds to the southern peak in the latitudinal cross section in Fig. 13. Its magnitude is comparable to or larger than that within the boundary layer. The active convection over the subtropical region produces an upward flux that lifts the pollution into lower free atmosphere where it is caught by the strong westerly, resulting in such a prominent flow pattern. In addition, *Uno et al.* [2003b] reported that biomass burning emission in the Southeast Asia also plays an important role for formation of this flow pattern; its contribution in this outflow exceeds 80%. We infer that carbonaceous aerosols have two main transport paths bounded by the boundary layer. One in the boundary layer between 35°N - 45°N is associated with anthropogenic aerosols.

4.3. Regional Budgets

Model results can be integrated to provide further insight into the transport of dust, sulfur, and carbonaceous aerosols during spring 2001. Figure 14 illustrates the regional budgets for each component schematically. It is notable that the outflow directions are considered in the RAMS/CFORS polar- stereographic projection. Total emission of mineral dust is estimated to be 105 Tg, which is equivalent to 3.5-10.5% of the global annual production of windblown dust reported by the *IPCC* [1995]. Anthropogenic emissions of sulfur and carbonaceous aerosols constitute more than half of their total emission. Dust and sulfur total deposition processes also play a major role in their transport processes, whereas carbonaceous deposition processes comprise a small fraction

of the total emission in this simulation because of a set low deposition rate and of the largely transport in the free atmosphere. Overall, eastward and northward outflows are relatively prominent for all components when compared to the westward and southward, which reflects their transport direction. Particularly, the eastward outflow for carbonaceous aerosols has a larger fraction because of the prevailing westerly flows. This result indicates that a large amount of carbonaceous aerosols are exported into northern Pacific regions and cause a larger radiative impact.

5. Conclusions

The transport of the main tropospheric aerosols (mineral dust, sulfate, black carbon, and organic carbon) in East Asia during spring 2001 was studied using the chemical transport model coupled with a regional-scale meteorological model (RAMS/CFORS). To understand the impact of aerosols on atmospheric radiation, AOTs for each component are also calculated. This simulation period contains an ACE-Asia intensive observation period and also corresponds to the period in which aerosols are frequently transported over East Asia. Sensitivity simulation without volcanic emission was performed during the same period to clarify impacts resulting from volcanic and anthropogenic sulfates on the atmospheric environment.

Numerous comparisons of model results with observations clarified model performance and aerosol transport characteristics. Further analyses for aerosol transport with its concentration, AOT, horizontal flux, and budget analysis were performed to produce an overview of aerosol transport and its radiative impact in the springtime 2001. Our main findings are the following:

1) Comparisons of lidar observation with simulation results indicated that the model captured characteristics of dust and sulfate transported to Beijing. Most of the high extinction regions for sulfate are limited to relatively lower altitude levels than that for dust. It was also shown that dust and sulfate transports have a strong correlation. The model reproduced episodic dust transport, on the average, contributes to about 60% of the total extinction coefficient, indicating a strong impact of dust transport on atmospheric radiation at Beijing.

2) Further comparisons with observed surface concentration of PM_{10} , sulfate, and TC show that the simulation has general agreement with them and reproduces many prominent features of aerosol transport over East Asia. In that comparison, temporally averaged values for both model and observation results during April imply the presence of a latitudinal gradient of aerosol concentration along the 140°E meridian, except for dust. This result indicates that the dust transport scale is, on the average, larger than Japan scale.

3) Analysis of the two-month averaged concentration field below 1000 m shows the latitudinal gradients more clearly. The main dust concentration field is located in more northern latitudes (between $30^{\circ}N - 45^{\circ}N$), while sulfate and carbonaceous field are mainly dominant from their main sources in central China and Southeast Asia to northern Japan (between $25^{\circ}N - 45^{\circ}N$). The volcanic

sulfate fraction is also evaluated and contributed about 13% of the total sulfate concentration, indicating that the anthropogenic sulfate mainly dominates over its concentration field within the boundary layer.

4) The modeled total AOT were compared with the TOMS AI over both land and ocean: close relationships between two temporal variations were then shown at all regions (Northeastern China, Northern Japan, Japan Sea, and Southern Japan). Model analyses showed that dust is a major constituent of total AOT at the northern region and the sulfate contribution is dominant from central China to the Kurile Islands. Carbonaceous AOT has the strong contribution from Southeast Asia to Southern Japan along the 30°N parallel. These results indicate that total AOT field in the East Asia is complicatedly composed by the different aerosol source regions, its emission strength and transport processes. Finally, mean relative contributions to the total AOT are estimated to be 36% for carbonaceous aerosols, 25% for sulfate, 24% for dust, and 15% for sea salt aerosols. We conclude that carbonaceous aerosols and sulfate, which mainly emitted by the human activity, largely contribute to the total AOT.

5) Mean sea level pressures and wind fields are closely related to aerosol transport within the boundary layer. Dust has two main transport directions from its source region (northeastward and eastward) within the boundary layer; this transport direction shifts to a more northerly latitude as the strength of the Pacific High increases. On the whole, the main transport path of dust is eastward along

the 45°N parallel. In addition we found that its main transport altitude corresponds to the 2-6 km from latitudinal variation of the eastward horizontal flux. Transport patterns in the boundary layer show a clockwise flow pattern for sulfate and carbonaceous aerosols, with the flow center located over southern China. Those patterns produce the strong outflow associated with anthropogenic emissions at northern latitudes (they are the main transport pathway for sulfate and carbonaceous aerosols) and constrain the continental outflow at the southern latitude. Further analysis shows that carbonaceous aerosols have another transport pathway at an upper level (2-6 km), which is along about 30°N from Thailand and Laos. That flow is dominated by biomass burning aerosols.

6) We performed a regional budget analysis for dust, sulfur, and carbonaceous aerosols. Total emissions during the simulation period are 105 Tg for mineral dust, 8.3 TgSO₂ for sulfate, and 3.07 Tg for carbonaceous aerosols. Total emission of mineral dust is one order larger than that of the other aerosols and is comparable to 3.5-10.5% of the global production of windblown dust [*IPCC*, 1995]. Dry deposition, gravitational settling, and northward outflow of mineral dust occupy 33%, 27%, and 13% of the total emission, respectively. Wet deposition, eastward outflow, and dry deposition of sulfate occupy 32%, 27%, and 21%, respectively. In the case of carbonaceous aerosols, the outflow to the outflow to the north (14%). The most predominant removal processes for each aerosol type are dry deposition for dust, wet deposition for sulfate, and eastward

outflow for carbonaceous aerosols. Overall, eastward and northward outflows are relatively prominent for all components, which reflects their main transport direction. Particularly, the eastward outflow for carbonaceous aerosols has a larger fraction of emissions. A large amount of carbonaceous aerosols are exported into the northern Pacific regions. This result implies a larger radiative impact.

Acknowledgments

This work was partly supported by Research and Development Applying Advanced Computational Science and Technology (ACT-JST) and Core Research for Evolution Science and Technology (CREST) of Japan Science and Technology Corporation (JST). This work was also supported in part by grants from the NSF Atmospheric Chemistry Program, NASA ACMAP and GTE programs, and the NOAA Global Change program. EANET PM₁₀ observation data were provided from the Acid Deposition and Oxidant Research Center, Niigata, Japan. The authors want to express their thanks to Dr. J.-S. Han of NIER, Korea for their measurement data for Gosan, Korea. This research is a contribution to the International Global Atmospheric Chemistry (IGAC) Core Project of the International Geosphere Biosphere Program (IGBP) and is part of the IGAC Aerosol Characterization Experiments (ACE).

References

Bey, I., D. J. Jacob, J. A. Logan, and R. M. Yantosca, Asian chemical outflow to the

Pacific in spring: origins, pathway, and budgets, J. Geophys. Res., 106, 23079-23113, 2001.

Carmichael, G. R., et al., Regional-Scale chemical transport modeling in support of the analysis of observations obtained during the TRACE-P Experiment, *J. Geophys. Res.*, 2003 (in press).

Charlson, R. J., S. E. Schwartz, J. M. Hales, R. D. Cess, J. A. Coakley Jr., J. E. Hansen, and D. J. Hofmann, Climate forcing by anthropogenic aerosols, *Science*, *255*, 423-430, 1992.

Chin M., et al., Tropospheric aerosol optical thickness from the GORCART model and comparisons with satellite and sun photometer measurements, *J. Atmos. Sci.*, *59*, 461-483, 2002.

Fernald, F. G., Analysis of atmospheric lidar observations: Some comments, *Appl. Opt.,* 23, 652-653, 1984.

Fujita, S., Y. Tonooka, and K. Ohta, Annual contribution of volcano sulfur dioxide emissions to the atmosphere in Japan, *J. Japan Soc. Air Pollution*, *27*, 33-1343, 1992.

Gillette D. and R. Passi: Modeling dust emission caused by wind erosion, *J. Geophys. Res.*, *93*, 14233-14242, 1988.

Gong. S. L., L. A. Barrie, and J.-P. Blanchet, Modeling sea-salt aerosols in the atmosphere: 1. Model development, *J. Geophys. Res.*, *102*, 3805-3818, 1997.

Gong. S. L., X. Y. Zhang, T. L. Zhao, I. G. McKendry, D. A. Jaffe, and Z. J. Zhou, Characterization of soil dust aerosol in China and its transport/distribution during 2001 ACE-Asia, 2. Model simulation and validation, *J. Geophys. Res.*, 2003 (in press).

Herman, J. R., P. K. Bhartia, O. Torres, C. Hsu, C. Seftor, and E. Celarier, Global distribution of UV-absorbing aerosols from Nimbus-7/TOMS data, *J. Geophys. Res.*, *102*, 16,911-16,923, 1997.

Huebert, B., T. Bates, P. Russell, G. Shi, Y. J. Kim, and K. Kawamura, An overview of ACE-Asia: strategies for quantifying the relationships between Asian aerosols and their climatic impacts, *J. Geophys. Res.*, 2003 (in press).

Intergovernmental Panel on Climate Change (IPCC), *Climate Change 1995: The Science of Climate Change*, J. T. Houghton, L. G. Meira. Filho, B. A. Callander, N. Harris, A. Kattenberg, and K. Maskell (Eds.), 572 pp., Cambridge Univ. Press, New York, 1996.

Iwasaka, Y., H. Minoura, and K. Nagaya, The transport and special scale of Asian dust-storm clouds: a case study of the dust-storm event of April 1979, *Tellus*, *35B*, 185-196, 1983.

Kaneyasu, N., K. Takeuchi, M. Hayashi, S. Fujita, I. Uno, and H. Sasaki, Outflow patterns of pollutants from East Asia to the North Pacific in the winter monsoon, *J. Geophys. Res., 105*, 17361-17377, 2000a.

Kaneyasu, N. and S. Murayama, High concentrations of black carbon over middle latitudes in the North Pacific Ocean, *J. Geophys. Res., 105*, 19811-119890, 2000b.

Kaufman, Y. J., D. Tanré, and O. Boucher, A satellite view of aerosols in the climate system, *Nature, 419,* 215-223, 2002.

Kim, B.-G., J.-S. Han, and S.-U. Park, Transport of SO₂ and aerosol over Yellow sea, *Atmos. Environ.*, *35*, 727-737, 2001.

Liousse, C., J. E. Penner, C. Chuang, J. J. Walton, H. Eddleman, and H. Cachier, A global three-dimensional model study of carbonaceous aerosols, *J. Geophys. Res.*, 101, 19411-19432, 1996.

Liu, M., L. Douglas, Westphal, D. L., S. Wang, A. Shimizu, N. Sugimoto, J. Zhou, and Y. Chen, A high-resolution numerical study of the Asian dust storms of April 2001, *J. Geophys. Res.*, 2003 (in press).

Matsumoto, K., M. Uematsu, T. Hayano, K. Yoshioka, H. Tanimoto, and T. Iida,

Simultaneous measurements of particulate elemental carbon on the ground observation network over the western North Pacific during the ACE-Asia campaign, *J. Geophys. Res.*, 2002JD002744, 2003 (in press).

Mellor, G. L. and T. Yamada, A hierarchy of turbulence closure models for planetary boundary layers, *J. Atmos. Sci.*, *31*, 1791-1806, 1974.

Mellor, G. L. and T. Yamada, Development of a turbulence closure model for geophysical fluid problems, *Rev. Geophys. Space Phys.*, *20*, 851-875, 1982.

Mesinger, F. and A. Arakawa, Numerical methods used in atmospheric models. GARP Publication Series, No. 14, WMO/ICSU Joint Organizing Committee, 64 pp., 1976.

Monahan, E. C., D. E. Spiel, and K. L. Davidson, A model of marine aerosol generation via whitecaps and wave disruption, in *Oceanic Whitecaps*, edited by E. C. Monahan and G. Mac Niocaill, pp. 167-174, D. Reidel, Norwell, Mass., 1986.

Novakov, T., D. A. Hegg, and P. V. Hobbs, Airborne measurements of carbonaceous aerosols on the east coast of the United States, *J. Geophys. Res.*, *102*, 30023-30030, 1997.

Ohta, S., R. Kato, N. Murano, and S. Yamagata, Measurements of optical and chemical properties of atmospheric aerosols at Fukue and Amami-ohshima island in APEX-E1 and APEX-E2, submitted to *J. Geophys. Res.*, 2003.

Pielke, R. A., W. R. Cotton, R. L. Walko, C. J. Tremback, W. A. Lyons, L. D. Grasso, M. E. Nicholls, M. D. Moran, D. A. Wesley, T. J. Lee, and J. H. Copeland, A comprehensive meteorological modeling system-RAMS, *Meteorol. Atmos. Phys.*, *49*, 69-91, 1992.

Ramanathan, V., P. J. Crutzen, J. T. Kiehl, and D. Rosenfeld, Aerosols, climate, and the hydrological cycle, *Science*, *294*, 2119-2124, 2001.

Seinfeld, J. H., et al., Regional climatic and atmospheric chemical effects of Asian dust and pollution, *Bull. Am. Met. Soc.*, 2003 (in press).

Shao, Y., A model for mineral dust emission, J. Geophys. Res., 106, 20239-20254, 2001.

Shimizu, A., N. Sugimoto, I. Matsui, K. Arao, T. Murayama, N. Kagawa, and I. Uno, Continuous observations of Asian dust and other aerosols by dual-polarization lidars in China and Japan during ACE-Asia., submitted to *J. Geophys. Res.*, 2003.

Streets, D. G., T.C. Bond, G. R. Carmichael, S. D. Fernandes, Q. Fu, D. He, Z. Klimont, S.
M. Nelson, N. Y. Tsai, M. Q. Wang, J.-H. Woo, and K. F. Yarber, An inventory of gaseous and primary aerosol emission in Asia in the year 2000, *J. Geophys. Res., 108*(D21), 8809 doi:10.1029/2002JD003093, in press, 2003.

Sugimoto, N., I. Matsui, A. Shimizu, I. Uno, K. Asai, T. Endoh, and T. Nakajima,

Observation of dust and sulfate aerosols plumes in the Northwest Pacific with a two-wavelength polarization lidar on board the research vessel Mirai, *Geophys. Res., Lett., 29 (19)*, doi:10.1029/2002GL015112, 2002.

Takemura, T., H. Okamoto, Y. Maruyama, A. Numaguti, A. Higurashi, and T. Nakajima, Global three-dimensional simulation of aerosol optical thickness distribution of various origins, *J. Geophys. Res.*, *105*, 17853-17873, 2000.

Takemura, T., A study on aerosol distributions and optical properties with a global climate model. *Ph.D. dissertation, Center for Climate System Research. Report No. 16,* Tokyo University, 2001

Takemura, T., I. Uno, T. Nakajima, A. Higurashi, and I. Sano, Modeling study of long-range transport of Asian dust and anthropogenic aerosols from East Asia, *Geophys. Res., Lett., 29*(24), 10.1029/2002GL016251, 2002.

Tegen, I. and I. Fung, Contribution to the atmospheric mineral aerosol load from land surface modification, *J. Geophys. Res.*, *100*, 18707-18726, 1995.

Twomey, S., Pollution and the planetary albedo, Atmos. Environ., 8, 1251-1256, 1974.

Uematsu M., Z. Wang, and I. Uno, Atmospheric input of mineral dust to the Western North Pacific region based of direct measurements and a regional chemical transport model, Geophys. Res., Lett., 30 (6), doi:10.1029/2002GL016645, 2003.

Uno, I., H. Amano, S. Emori, K. Kinoshita, I. Matsui, and N. Sugimoto, Trans-Pacific yellow sand transport observed in April 1998: A numerical simulation, *J. Geophys. Res.*, *106*, 18331-18344, 2001.

Uno, I., G. R. Carmichael, D. G. Streets, S. Satake, T. Takemura, J. Woo, M. Uematsu, and S. Ohta, Analysis of surface black carbon distributions during ACE-Asia using a regional scale aerosol model, *J. Geophys. Res.*, 2003a (in press).

Uno, I., et al., Regional chemical weather forecasting system CFORS: Model descriptions and analysis of surface observations at Japanese island stations during ACE-Asia experiment, *J. Geophys. Res.*, 2003b (in press).

Uno, I., S. Satake, G. R. Carmichael, Y. Tang, T. Cahill, N. Sugimoto, and M. Uematsu, Sources, transport and distribution of Asian dust aerosols during springtime 2001 simulated with the CFORS model, *J. Geophys. Res.*, 2003c (writing).

Walko, R. L., W. R. Cotton, M. P. Meyers, and J. Y. Harrington, New RAMS cloud microphysics parameterization, part I: the single-moment scheme, *Atmospheric Research*, 28, 29-62, 1995.

Wang, Z., H. Ueda, and M. Y. Huang, A deflation module for use in modeling long-range

transport of yellow sand over east Asia, J. Geophys. Res., 105, 26,947-26,959, 2000.

Westphal, D. L., O. B. Toon, and T. N. Carlson, A case study of mobilization and transport of Sahara dust, *J. Atmos. Sci.*, *45*, 2145-2175, 1988.

Xiao, H., G. R. Carmichael, J. Durchenwald, D. Thornton, and A. Bandy, Long-range transport of Sox and dust in East Asia during the PEM B experiment, *J. Geophys. Res.*, *102*, 28,589-28,612, 1997.

Xuan, J. and N. Sokolik, Characterization of sources and emission rates of mineral dust in Northern China, *Atmos. Environ.*, *36*, 4863-4876, 2002.

Zhang, M., I. Uno, Y. Yoshida, Z. Wang, and H. Akimoto, Transport and Transformation of Sulfur compound over East Asia during the TRACE-P and ACE-Asia Campaigns, *J. Geophys. Res.*, 2003 (in press).

Species	Dry depositio	n velocity [m/s]	Wet scavenging [s ⁻¹]
species	Land	Sea	(<i>Pr</i> in mm/hour)
SO_2	3.0×10 ⁻³	3.0×10 ⁻³	$2.0 \times 10^{-5} \times Pr$
SO_4	1.0×10^{-3}	2.0×10^{-3}	$5.0 \times 10^{-5} \times Pr^{0.88}$
Black Carbon	1.0×10^{-4}	1.0×10^{-3}	0
Organic Carbon	2.0×10^{-4}	2.0×10 ⁻³	$1.0 \times 10^{-6} \times Pr$
Sea salt (fine)	1.0×10^{-3}	1.0×10^{-3}	$5.0 \times 10^{-5} \times Pr$
Sea salt (coarse)	5.0×10^{-3}	5.0×10 ⁻³	$6.0 \times 10^{-5} \times Pr^{0.83}$
Mineral dust			
r < 1[µm]	5.0×10^{-4}	5.0×10^{-4}	$1.0 \times 10^{-5} \times Pr$
$1[\mu m] < r < 2.5[\mu m]$	3.0×10^{-3}	3.0×10^{-3}	$6.0 \times 10^{-5} \times Pr^{0.83}$
2.5 [μm] < r < 10 [μm]	1.0×10^{-2}	1.0×10^{-2}	$6.0 \times 10^{-5} \times Pr^{0.83}$
10 [µm] < r	1.0×10^{-1}	1.0×10^{-1}	$6.0 \times 10^{-5} \times Pr^{0.83}$

 Table 1. Deposition parameters of each aerosol type

Table2. Modeled effective radii, extinction efficiency factors

	0		5									
Mineral dust												
Mode radius [µm]	0.13	0.20	0.33	0.52	0.82	1.27	2.02	3.20	5.06	8.02	12.7	20.13
Extinction efficiency factor	0.86	2.38	4.10	2.59	2.73	2.28	2.34	2.11	2.17	2.09	2.07	2.05
Scattering efficiency factor	0.82	2.30	3.95	2.35	2.36	1.84	1.79	1.41	1.36	1.20	1.20	1.20
Sea salt												
Mode radius [µm]	0.52	5.62										
Extinction efficiency factor	3.81	2.27										
Scattering efficiency factor	3.81	2.27										

and scattering efficiency factors of dust and sea salt in the model.

and scattering coefficients of sulfate and carbonaceous aerosols.								
Relative humidity [%]	0	50	70	80	90	95	98	99
Sulfate								
Mode radius [µm]	0.07	0.085	0.095	0.103	0.122	0.157	0.195	0.231
Extinction coefficient [m ² /g]	4.28	7.21	10.12	12.60	20.09	35.04	44.95	47.38
Scattering coefficient [m ² /g]	4.28	7.21	10.12	12.60	20.09	35.04	44.95	47.38
Organic carbon								
Mode radius [µm]	0.1	0.108	0.11	0.144	0.169	0.196	0.274	0.312
Extinction coefficient [m ² /g]	3.83	5.05	5.38	12.83	20.62	30.05	57.88	60.76
Scattering coefficient [m ² /g]	3.75	4.96	5.30	12.75	20.53	30.41	57.78	60.67
Black carbon								
Mode radius [µm]	0.0118							
Extinction coefficient [m ² /g]	8.339							
Scattering coefficient [m ² /g]	1.796							

Table3. Relative humidity and modeled mode radii and extinction

Table 4. Monthly mean surface concentration for April ($\mu g/m^3$)

		Carbonaceous (BC+OC)	Sulfate (anthropogenic)	$PM_{10} \text{ and } PM_{model}$
Rishiri	OBS	1.69	3.52	48.50
(N45.1°, E141.1°)	Model	1.74	3.97 (86%)	46.77
Sado	OBS	2.58		52.73
(N38.1°, E138.2°)	Model	1.86	6.68 (60%)	41.25
Gosan	OBS		5.00	131.17
(N33.3°, E126.2°)	Model	2.51	5.50 (71%)	66.67
Hachijo	OBS	0.77	3.67	
(N33.2°, E139.8°)	Model	1.59	4.48 (64%)	45.73
	OBS	2.01	4.51	55.46
	Model	2.67	3.55 (80%)	46.41

	a) North-eastern	b) Northern	c) Japan Sea	d) Southern part
	part of China	part of Japan		of Japan
	(NC)	(NJ)	(JS)	(SJ)
Sea salt	0.4	12.4	5.8	9.9
Sulfate	23.6	40.0	41.8	37.0
Dust	64.2	32.6	36.6	12.3
Carbonaceous	11.8	15.0	15.8	40.8
Total AOT	0.35	0.34	0.37	0.32

Table 5. Relative contribution of each aerosol to the total AOT (%) and the mean total AOT (unit less)

Figure captions

- Figure 1 RAMS/CFORS model domain. Triangles indicate observation sites (A: Amami, B: Beijing, G: Gosan, H: Hachijo, R: Rishiri, and S: Sado). The four gray regions are used to compare simulation results with TOMS AI in section 3.4 (see Fig. 9).
- Figure 2 Total emissions of a) anthropogenic sulfur $(g-SO_2/m^2)$, b) anthropogenic BC+OC (g/m^2) , c) biomass BC+OC (g/m^2) , d) mineral dust (g/m^2) , and e) sea salt (g/m^2) .
- Figure 3 Time-height cross sections of a) mineral dust extinction coefficient observed by lidar (1/km), b) air pollution extinction coefficient observed by lidar (1/km), c) simulated mineral dust extinction coefficient (1/km), and d) simulated sulfate extinction coefficient (1/km) at Beijing.
- Figure 4 Two-month averaged vertical profile of the modeled (solid line) (bottom) and observed (dashed line) (top) total extinction coefficients (1/km) at Beijing. Shaded parts inside the solid line indicate dust extinction for the dark gray, sulfate extinction for gray, and carbonaceous extinction for white. Impacts of spherical aerosols by lidar are also shown as the difference between extinction coefficients of total aerosols and dust.

- Figure 5 Time variation for simulated total aerosol concentrations (PM_{model}) (by solid line) ($\mu g/m^3$) and for observed PM_{10} concentrations ($\mu g/m^3$) (open circle and dot line) for a) Gosan, b) Rishiri, c) Sado, and d) Amami.
- **Figure 6** As Fig. 5, but for the sulfate concentration $(\mu g/m^3)$ for a) Gosan, b) Rishiri, c) Hachijo, and d) Amami. The bold dotted line indicates VolcOFF results.
- Figure 7 Same as Fig. 6, but for carbonaceous aerosols concentration (μ g/m³) for a) Rishiri, b) Sado, c) Hachijo, and d) Amami.
- Figure 8 Horizontal distribution of boundary layer (below 1000 m) averaged aerosol concentration field (μ g/m³) during spring 2001 for a) mineral dust, b) sulfate (solid line) and VolcOFF sulfate (dot line), c) carbonaceous aerosols, and d) sea salt.
- Figure 9 Time variation of the model AOT and TOMS AI (unitless) at a) northeastern China (NC), b) northern Japan (NJ), c) Japan Sea (JS), and d) southern Japan (SJ). Model aerosol AOT and TOMS AI are averaged over the gray regions in Fig. 1.
- Figure 10 Horizontal distribution of the averaged model AOT field (unitless) during spring 2001 for a) mineral dust, b) sulfate (solid line) and VolcOFF sulfate (dot line), c) carbonaceous aerosols, d) sea salt, and e) total aerosols AOT.

- Figure 11 Latitudinal variation of the averaged model AOT for main tropospheric aerosols at 130°E.
- Figure 12 Two monthly mean sea level pressures by line (hPa) and wind fields by vector (m/s) during spring 2001. Wind fields are averages for the surface to z*= 1000 m.
- Figure 13 The boundary layer (below 1000m) averaged horizontal mass flux (left column), the column averaged horizontal mass flux (middle column) and the latitudinal cross section of the eastward horizontal mass flux at 130°E (right column) during spring 2001 for mineral dust, sulfate and carbonaceous aerosols.
- Figure 14 Regional budgets for a) mineral dust, b) sulfate, and c) carbonaceous aerosols. Emission, deposition, and outflow are shown by arrows in Tg (Tg-SO₂ for sulfur cycle). Numbers in parentheses under each deposition and outflow indicate the ratio to emission.



Figure 1.









d) Mineral dust emission (g/m²)





Figure 2.



Height (km)

Height (km)



Figure 4.



Figure 5.





Figure 7.









Figure 8.



a) Mineral dust







e) Total aerosols (dust, sulfate, BC+OC, and sea salt)



Lev = 0.03, 0.05, 0.1, 0.15, 0.2, 0.3, 0.4, 0.5

Figure 10.



Figure 11.







Figure 12.



Figure 13.

a) Mineral dust

	Mass load	ling in the atn	nosphere (6.7 Tg (6%)
	Dry	Wet G	Fravitation	al
	deposition	deposition	settling	
	(33%)	9.4 1g (9%)	26.4 1g (27%)	outflow from simulation domain
			_	East 9.02 Tg (9%)
Emis	ssion		_	West 3.30 Tg (2%)
105	Tg		_	South 0.93 Tg (1%)
	L L	↓ ↓	• -	North 15.2 Tg (13%)

b) Sulfate and Sulfur dioxide



c) BC + OC (TC)



Figure 14.