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transport of aerosols
and air quality
implications**

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Intercontinental transport of pollution and dust aerosols: implications for regional air quality

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Abstract

We use the global model GOCART to examine the impact of pollution and dust aerosols emitted from their major sources on surface fine particulate matter concentrations at regional and hemispheric scales. Focusing on the North America region in 2001, we use measurements from the IMPROVE network in the United States to evaluate the model-simulated surface concentrations of the “reconstructed fine mass” (RCFM) and its components of ammonium sulfate, black carbon (BC), organic matter (OM), and fine mode dust. We then quantify the RCFM budget in terms of the RCFM composition, type, and origin to find that in the eastern U.S., ammonium sulfate is the dominant RCFM component ($\sim 60\%$) whereas in the western U.S., dust and OM are just as important as sulfate but have considerable seasonal variations, especially in the NW. On an annual average, North America regional pollution accounts for nearly 30–40% of the surface RCFM in the western U.S., and for a much higher proportion of 65–70% in the eastern U.S. By contrast, pollution from outside of North America contributes to just 2–6% ($\sim 0.2 \mu\text{g m}^{-3}$) of the total RCFM over the U.S. on an annual average. In comparison, long-range transport of dust is more efficient than that of pollution, which brings 3 to 4 times more fine particles to the U.S. ($0.5\text{--}0.8 \mu\text{g m}^{-3}$ on an annual average) with a maximum influence in spring and over the NW. Of the major pollution regions, Europe has the largest potential to affect the surface aerosol concentrations in other continents due to its shorter distance from receptor continents and its larger fraction of sulfate-producing precursor gas in the outflow. With the IPCC emission scenario for the year 2000, we find that European emissions increase levels of ammonium sulfate by $1\text{--}5 \mu\text{g m}^{-3}$ over the surface of northern Africa and western Asia, and its contribution to eastern Asia ($\geq 0.2 \mu\text{g m}^{-3}$) is twice as much as the Asian contribution to North America. Asia and North America pollution emissions exert strong impacts on their neighboring oceans, but their influences on other continents are relatively small ($\leq 10\%$) due to the long traveling distance across the oceans and efficient removal during transport. Among the major dust source regions, Asia displays a significant in-

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fluence over large areas in the Northern Hemisphere except over the North Atlantic and the tropics, where African dust dominates. The trans-Pacific transport of Asian dust is much more efficient than that of Asian pollution because of the higher elevation and lower removal rate of dust. We notice that the African dust can travel eastward through a pathway spanning across Asia and North Pacific to settle down over the surface of western North America. African dust transported through such a pathway is difficult to detect because it usually merges and travels together with the Asian dust.

1 Introduction

Aerosol, also known as particulate matter (PM), is one of the major air pollutants determining ambient air quality. Airborne particle sizes vary widely from a few nm (10^{-9} m) to a few hundred μm in diameter; those with diameters smaller than $10\ \mu\text{m}$ (PM_{10}) are of health concern because they can penetrate into the lungs, and those smaller than $2.5\ \mu\text{m}$ ($\text{PM}_{2.5}$) pose the most serious risks to human health, being linked to respiratory or cardiovascular diseases and even deaths (Ostro et al., 1999, 2000; World Health Organization, 2002; Pope, 2002). Aerosol is also known to cause regional haze, which leads to discoloration, loss of texture, and deterioration of visual range in national parks and wilderness areas (Malm et al., 2000). Sources of PM include both direct emissions and chemical transformations of precursor gases emitted from power plants, automobiles, wood burning, forest and agriculture fires, and natural sources (e.g., dust, volcanoes, vegetation).

Airborne PM concentration has been regulated in the United States by the Clean Air Act since 1970. Currently, the indices of “good” air quality for PM_{10} and $\text{PM}_{2.5}$ in the U.S. are $50\ \mu\text{g m}^{-3}$ and $15\ \mu\text{g m}^{-3}$ on an annual average, respectively, as set by the U.S. Environmental Protection Agency. Recognizing the importance of visual air quality, the U.S. Congress included legislation in the 1977 Clean Air Act to remedy existing and prevent future visibility impairment in national parks and wilderness areas. Although pollutant emissions in the U.S. are projected to decrease in the next 100

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years (IPCC, 2001), the long-range transport of aerosols from other regions in the world could increase the surface PM level and may reduce the benefits generated from local emission controls. Recently, much attention has been focused on assessing the long-range transport of aerosols from Asia to North America, because Asia is the fastest economically developing region and has large active deserts, from which the trans-Pacific transport can bring increasingly significant amount of pollution and dust to North America. For example, episodes of elevated sulfate concentrations that were observed in the northwestern U.S. in springtime were attributed to Asian emissions (Jaffe et al., 2003; Bertschi et al., 2004), and satellite images have clearly revealed the trans-Pacific transport of Asian dust during dust storm events, which bring large amounts of dust to western North America and strongly affect the radiation budget and air quality (Husar et al., 2001). Previous global modeling studies have estimated that trans-Pacific Asian pollution could account for 30% of “background” surface sulfate concentrations over the U.S. (i.e., concentration without U.S. anthropogenic emission) (Park et al., 2004), which peaked in the spring season, adding $0.16 \mu\text{g m}^{-3}$ to the northwestern U.S. (Heald et al., 2006). It is estimated that the Southeast Asia is the largest contributor to the black carbon over the Arctic and is also partially responsible to the “Arctic Haze” problem (Koch and Hansen, 2005; Koch et al., 2007).

In the meantime, major economically developed regions such as the U.S. and Europe remain major pollution sources. The impact of the North American export of ozone and its precursors on the surface ozone over Europe has been extensively studied, showing a maximum impact during the summer time (Stohl and Trickl, 1999; Wild et al., 2001; Li et al., 2002, 2005; Guerova et al., 2006). Similarly, European pollution increases the surface ozone concentrations over northern Africa, the Mediterranean, and the Near East to a level that exceeds the European Council’s human health standards (Stohl et al., 2002; Duncan and Bey, 2004). The transport of sulfate aerosol from Europe and North America to the Arctic has been identified as the major cause of the “Arctic Haze” (the phenomenon of a visible reddish-brown haze in the atmosphere at high latitudes in the Arctic due to air pollution) (Shaw, 1995, and references therein). However,

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so far, most of the previous source-receptor studies regarding hemispheric transport have primarily concentrated on ozone and individual aerosol species (e.g., sulfate or carbonaceous) or aerosol radiative forcing, leaving large gaps in a comprehensive assessment of the surface PM levels that are determined from regional emissions and long-range transport.

We present here a global modeling assessment of intercontinental transport of aerosols and its implication for regional air quality with the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model. In this study, we focus on the year 2001, which has been used in our previous analysis of satellite and ground-based sunphotometer measurements of aerosol optical thickness (AOT) and aircraft data from the ACE-Asia field experiments (Chin et al., 2003, 2004). The present study brings our focus to the surface aerosol mass concentrations and is intended to (1) quantify the surface fine aerosol mass concentrations over the U.S. that come from regional pollutant emission, natural sources, and intercontinental transport, and (2) examine the impact of the pollution and dust emitted from one major source region on the surface fine aerosol mass over other areas in the Northern Hemisphere. We will briefly describe the model (Sect. 2), compare the model simulated surface aerosol concentrations with measurements in the U.S. (Sect. 3), provide assessments of the impact of emissions and long-range transport on the U.S. surface air quality (Sect. 4), and examine the hemispheric influence of the intercontinental transport (Sect. 5), followed by a summary and conclusions (Sect. 6).

2 GOCART model simulation of global aerosol distributions

The GOCART model is used to simulate major tropospheric aerosols including sulfate, dust, black carbon (BC), organic carbon (OC), and sea-salt. Detailed descriptions of the model have been presented in our previous publications (Chin et al., 2000a, b, 2002, 2004; Ginoux et al., 2001, 2004) with a few modifications applied to this work. We use assimilated meteorological fields from the NASA Goddard Earth Observing

System version 3 Data Assimilation System (GEOS-3 DAS) for 2001 with a spatial resolution of 2° latitude by 2.5° longitude and 30 vertical layers. Processes in the model include emission, chemistry, convection, advection, boundary layer mixing, dry and wet deposition, and gravitational settling. AOT is calculated from dry mass concentrations and mass extinction coefficients which are determined from the size distributions, refractive indices, and hygroscopic properties of the individual aerosol types as a function of ambient relative humidity (Chin et al., 2002).

Emissions from fuel combustion sources are taken from the Special Report of Emission Scenarios (SRES) for the year 2000 (Nakićenović et al., 2000) for SO₂ and from Cooke et al. (1999) for BC and OC. Biofuel emissions of BC and OC are based on the biofuel use assessment study of Yevich and Logan (2003) with emission factors of 0.6 g BC and 4.0 g OC per kg dry mass consumed. Biomass burning emissions in 2001 are based on the monthly dry mass burned data from the Global Fire Emission Dataset (van der Werf et al., 2003), using emission factors of 8, 1, and 1.1 g kg⁻¹ for OC, BC, and SO₂, respectively. These emission factors have been somewhat modified from our previous work but remain within the range of the values reported in the literature (see Chin et al., 2002, for details). Dust and sea-salt emissions for particle size from 0.1 to 10 μm (dry radius) are calculated as a function of surface properties and wind speed (Ginoux et al., 2001; Chin et al., 2002, 2004). In addition, biogenic and volcanic emissions are also included in the model. The annual emissions of total sulfur (mostly in the form of SO₂ over land and dimethylsulfide, or DMS, over ocean), carbonaceous species (OC and BC), and dust in the Northern Hemisphere for 2001 are listed in Table 1, with the northern hemispheric emission pattern shown in Fig. 1. Major industrial pollution regions in the Northern Hemisphere, identified as North America, Europe, and Asia, account for about 85% of global pollution sources of SO₂, BC, and OC. As for dust, more than 90% is emitted from desert regions of northern Africa, Middle East (including Central Asia), and Asia (including China and Mongolia), with almost 60% from northern Africa (Table 1). The domains of three major anthropogenic source regions are outlined in Figs. 1a and b and three major dust regions in Fig. 1c. The

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impacts of these regional emissions will be analyzed in Sects. 3 and 4.

Our previous analyses of the satellite and ACE-Asia field experiment data for spring 2001 have shown that the observed aerosol distributions and long-range transport were adequately captured by the model (Chin et al., 2003, 2004). Figure 2 illustrates the global fields of AOT, separated by fine mode (red) and coarse mode (green) components, on 13 April and 22 August 2001 observed by the Moderate Resolution Imaging Spectroradiometer (MODIS) instrument on the EOS-Terra satellite and simulated by the GOCART model. The fine mode AOT consists mainly of pollution and biomass burning aerosols while the coarse mode consists mainly of dust and sea-salt aerosols. On 13 April 2001, the cross-Pacific transport of dust and pollution from Asia to North America and the cross-Atlantic transport of dust from Africa to Central America are clearly revealed, while on 22 August large smoke plumes from South America to the southern Atlantic and from southern Africa to the Indian Ocean are evident from both MODIS and the model. The impact of such large-scale transport on the receptor region's air quality depends on the perturbation of the surface aerosol concentrations, which we will discuss in the following sections.

3 Surface aerosol concentrations over the U.S. from IMPROVE and model

3.1 IMPROVE data

Our model results for surface aerosol concentrations are evaluated here with observations over the U.S. from the Interagency Monitoring for Protected Visual Environments (IMPROVE) network. The IMPROVE program was initiated in 1985 by the U.S. federal agencies EPA, National Park Services, Department of Agriculture-Forest Service, and other land management agencies as a part of the EPA Regional Haze program (Malm et al., 1994). IMPROVE has implemented an extensive, long-term monitoring program in order to establish current visibility conditions and track visibility changes in national parks and wilderness areas. There are more than 100 IMPROVE sites in

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the U.S. routinely measuring aerosol chemical compositions as well as fine and coarse mode aerosol mass concentrations (<http://vista.cira.colostate.edu/IMPROVE>). The locations of the IMPROVE sites that provided data in 2001 are shown in Fig. 3. We divide the U.S. into four quadrants, NW, SW, NE, and SE with dividing lines along 40° N and 100° W, to differentiate regional features.

The fine mode aerosol species of sulfate, OC, BC, and dust from IMPROVE are used in this study. While sulfate particles are collected on a nylon filter and analyzed by ion chromatography, OC and BC (aka elemental carbon or EC) are derived based on thermal optical reflectance combustion, a process that involves multiple steps including heating the sample and oxidizing/reducing the carbon to different chemical compounds (e.g., CO₂, CH₄) (see Malm et al., 1994, for details). As such, the reported OC and BC data from the IMPROVE sites would have relatively larger uncertainties than would sulfate. The mass of soil dust is calculated from a linear combination of the measured elements associated predominantly with soil, including Al, Si, Ca, Fe, and Ti (Malm et al., 1994); uncertainties associated with the reported dust values reflect the range and variation of mineral composition from a variety of soil types. Finally, in addition to PM_{2.5} and PM₁₀, IMPROVE also reports the quantity of “reconstructed fine mass” (RCFM), which is the sum of mass concentrations of ammonium sulfate, organic matter (OM), BC, and fine dust particles (diameter less than 2.5 μm). Nitrate and sea-salt are excluded from the RCFM because of the significant loss of nitrate particles during sampling and the negligible amount of sea-salt at most of the sites (Malm et al., 1994). We will therefore use the quantity of RCFM in this study, which is particularly appropriate because the GOCART model does not include nitrate. The IMPROVE measurements showed that concentrations of RCFM are very close to that of PM_{2.5} in most places over the U.S., except in California where nitrate can make up a significant fraction of PM_{2.5} (Malm et al., 1994, 2003). In the reported RCFM, ammonium sulfate mass is converted from the measured sulfate ion by multiplying a factor of 1.375, assuming that sulfate exists in the form of ammonium sulfate (or 100% neutralization); the OM is estimated by applying a correction factor of 1.4 to the OC mass concentrations to

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account for non-carbon mass contained in the organic carbon aerosol (Malm et al., 1994, 2003). The same conversion factors are applied to the model-calculated sulfate and OC to construct the modeled value of RCFM.

3.2 Composition of surface fine aerosol mass over the U.S.

5 Daily variations in surface aerosol species and RCFM concentrations are illustrated in Fig. 4 at four IMPROVE sites, each located in the NW, SW (in Fig. 4a), NE, and SE (in Fig. 4b) quadrants. At the NW site of Sula, Montana (SULA1), the seasonal variation of RCFM exhibits two maxima, one in the spring (April-May) controlled mainly by dust from long-range transport, and the other in the summer (July–August) caused by carbonaceous (OC and BC) from biomass burning. At the SW site of Bandelier National Monument, New Mexico (BAND1), dust is the major aerosol component, most of which is from the nearby Sonora Desert (located at southwestern Arizona and southeastern California) but the spring peak is caused by the trans-Pacific transport from Asia (see Sect. 4). Notably, the model-simulated BC and OC concentrations in the first and last quarter of the year are much lower than the measurements (average 80% lower). This may be explained in part by the fact that we did not consider the seasonal variation of anthropogenic BC and OC emissions which are suggested to be 15 to 50% higher in the winter than in the summer (Park et al., 2003).

10 The two eastern sites (Fig. 4b), Bondville, Illinois (BOND1) in NE and Sipsy, Alabama (SIPS1) in SE show much higher RCFM concentrations than the two western sites in the U.S. (Fig. 4a) mainly because of much stronger industrial pollution emissions in the eastern U.S. (Fig. 1). The model attributes the increase of BC and OC in November to biomass burning, but it misses the high episode in March that is likely also from biomass burning. However, these episodic events cannot be resolved by the monthly averaged fire emission data we used in the model. Fine dust at the two eastern sites shows a spring maximum which is connected with the long-range transport, a feature rather similar to that displayed in Fig. 4a for the western sites.

25 Comparisons of monthly averaged concentrations of RCFM between the measure-

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ments and the model at 142 IMPROVE sites for 2001 are plotted in Fig. 5. The majority of these values agrees to within a factor of 2; the points where the model is more than 2 times higher or lower than the data are mostly from the disagreement in fine dust (higher in the model) and carbonaceous (lower in the model) concentrations. Overall, the model captures the IMPROVE reported RCFM levels and seasonal variations with small bias as indicated by the relatively high correlation coefficient (R), low bias (B), and high skill score (S), but the statistics are different for each RCFM component, as listed in Table 2. The model shows the best skill in simulating sulfate ($R=0.82$, $B=1.15$, $S=0.88$), but its simulated carbonaceous aerosol concentrations (especially OC) are lower and fine dust concentrations are higher than the IMPROVE reported values and with lower correlation coefficients ($R=0.5-0.6$). This discrepancy reflects either deficiencies in the model, such as a lack of seasonal variation of BC and OC emissions from fossil fuel and biofuel consumptions, as discussed earlier, or insufficient removal of dust during transport. On the other hand, uncertainties in the IMPROVE carbonaceous and dust aerosol data are also relatively high because they are either inferred from optical/thermal measurements (for BC and OC) or derived from a combination of multiple mineral elements (for dust) as described in Sect. 3.1. In any case, the discrepancies suggest that the model may underestimate the influence of North American regional sources but overestimate that of long-range transport on the surface RCFM over the U.S., since most carbonaceous aerosols are generated by the North American sources while most dust is transported into the U.S. (see Sect. 4).

The area-averaged seasonal cycle of RCFM and its composition calculated with the model for the entire domain of NW, SW, NE, and SE are shown in Fig. 6. As indicated earlier, sulfate is counted as ammonium sulfate and OC as OM in constructing the quantity of RCFM. The annual averaged RCFM concentrations are highest in the SE ($8.32 \mu\text{g m}^{-3}$) and lowest in the NW ($3.43 \mu\text{g m}^{-3}$), with NE ($5.36 \mu\text{g m}^{-3}$) and SW ($4.91 \mu\text{g m}^{-3}$) in between. The major difference between the western and eastern U.S. is that ammonium sulfate dominates the RCFM in the east by taking up nearly 60% of the total RCFM while it shares the burden with dust and OM in the western U.S.

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The seasonal cycle of OM is significantly different among the four quadrants, which is regulated primarily by biomass burning with profound spatial and temporal variations. BC makes up just 4–6% of RCFM across the U.S., although it is the most important species for light absorption and aerosol atmospheric heating. The model calculated annual averaged concentrations of RCFM and its components for the four quadrants are tabulated in Table 3, together with the results from model experiments aimed at associating the RCFM with the source types and regions. These model experiments will be discussed next.

4 Origins of surface aerosols over the U.S.: regional sources vs. intercontinental transport

Having analyzed the spatial and temporal variations of the surface RCFM over the U.S., we now try to quantify the amount of RCFM from regional pollution sources that may be regulated by the air quality management as opposed to those from natural sources or from long-range transport that is difficult to control. We have conducted a series of model experiments to estimate the RCFM from North American pollution sources vs. “background” (i.e. from natural and long-range transport), and from total regional sources (pollution, natural, and biomass burning) vs. intercontinental transport. The model experiments are designed in such a way that emissions from a selected source type (e.g., fossil fuel, biomass burning, dust) or a source region (e.g., Asia, Europe, Africa) are excluded in each model run, and the difference between the “standard” run with all emissions included and the runs with a particular source type or region excluded is considered as the contribution from that source type or region. We categorize the source types as those tabulated in Table 2: “pollution”, which includes emissions from fossil fuel and biofuel consumptions, “biomass burning”, which includes both natural wild fires and burning from agriculture waste or land management practice, and “natural”, which includes emissions from ocean, vegetation, deserts, and volcanoes.

Figure 7 shows the seasonal cycles of RCFM that are attributed to different source

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categories averaged over the four quadrants in the U.S. In the western half, North American regional pollution (NAM in Fig. 7) accounts for 28% of the surface RCFM in the NW and 38% in the SW, with the rest being “background”. In contrast, regional pollution exhibits a clear dominance in the eastern U.S., accounting for 71% and 65% of the total surface RCFM in the NE and the SE, respectively. The intercontinental transported pollutants (TAP) provide about $0.2 \mu\text{g m}^{-3}$ RCFM to the U.S., which is about 4 to 10 times less in the western and 20 to 30 times less in the eastern U.S. than North America’s own pollution RCFM. On the other hand, long-range transport of dust adds significant amount of fine particles, $0.7\text{--}0.8 \mu\text{g m}^{-3}$ over the western and a little over $0.5 \mu\text{g m}^{-3}$ over the eastern U.S. on an annual average. While North American pollution aerosols have relatively small seasonal variations, background aerosols (BMB, NAT, TDU_f , and TAP) fluctuate considerably by season and region. For example, biomass burning aerosol contribution can change from zero during non-burning seasons to dominant in the peak burning season; likewise, fine dust concentrations during the peak transport season can be hundreds of times higher than those in the “quiet” seasons. Thus, the significance of the background aerosol varies significantly with location and time, especially in the NW.

The total amount of RCFM from the intercontinental transport is the sum of pollution and fine dust originating in regions other than North America ($\text{TAP} + \text{TDU}_f$ in Fig. 7), with a majority being dust (70–80%). On an annual and area average basis, the intercontinental transport raises RCFM in the NW, SW, NE, and SE regions by 1.0, 0.91, 0.72, and $0.75 \mu\text{g m}^{-3}$, respectively, corresponding to 30%, 19%, 14%, and 9% of the total RCFM in those regions. Therefore, the majority of the surface fine mass over the U.S. is from North American regional sources, including pollution, biomass burning, and natural sources.

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5 Hemispheric influence of intercontinental transport

Although our main focus in this paper is to assess the impact of aerosols from different types and regions on surface air quality over the U.S., we further examine the hemispheric influence of pollution and dust aerosols from their major source regions in order to gain a broader perspective on intercontinental transport influences. Because ammonium sulfate is generally the major component of pollution aerosols, we use it as a surrogate for pollution aerosols in the following analysis. The major pollution and dust source regions are shown in Fig. 1 with their emission amounts listed in Table 1. Recall that the contribution from each region is estimated from the difference between a model simulation with all emissions included and one with a particular regional emission excluded. Our previous studies have shown that the model reproduces the observed sulfate and dust concentrations and seasonal variations to within a factor of 2 in the major pollution sources regions (e.g., North America and Europe) and over the areas affected by the transport (e.g., western Pacific and North Atlantic) (Chin et al., 2000b, 2003; Ginoux et al., 2001, 2004), providing a foundation for our assessment here.

5.1 Pollution transport

Figure 8a displays the annually averaged surface ammonium sulfate concentrations in the Northern Hemisphere in 2001 and contributions from pollution sources in Asia, Europe, and North America. Over East Asia, the surface ammonium sulfate concentration is about twice as high as that over the eastern U.S. and Europe (top panel, Fig. 8a), due to higher emission of SO_2 (Table 1) and faster oxidation rate of SO_2 to form sulfate (Chin et al., 2000b). As expected, pollution sources exert the largest impact on their own regions and neighboring oceans (2nd to 4th panels in Fig. 8a) where more than 80% of the surface sulfate in the source regions and 50% over neighboring oceans are from regional pollution.

Most of the Asian pollution export moves toward the east across the Pacific Ocean, increasing the ammonium sulfate at the surface over the western U.S. by 0.1–

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0.2 $\mu\text{g m}^{-3}$ on annual average (2nd panel in Fig. 8a). Some of the pollution is transported westward to add 0.1–0.5 $\mu\text{g m}^{-3}$ ammonium sulfate to eastern Europe and Middle East, and more than 0.1 $\mu\text{g m}^{-3}$ to North Africa. Even so, Asian emission results in no more than a 10% increase of surface sulfate to downwind continents on an annual average except Canada and Greenland (2nd panel in Fig. 8a). Trans-Pacific transport from Asia to North America involves the uplifting of Asian air by convection, followed by a rapid horizontal transport in the prevailing westerlies and subsidence over the western part of North America. The large gradient of Asian pollution sulfate between the east coast of Asia and the west coast of North America (a factor of 50 to 100) implies a combined effect of plume height and efficient removal of sulfate during transport. Most pollution sulfate is located within the boundary layer upon leaving the Asian coast, as observed during the ACE-Asia experiment (e.g., Anderson et al., 2003; Chin et al., 2003), subjecting it to relatively fast removal; only the fraction entrained in the westerly jet can make the long journey across the Pacific. Our results thus suggest that Asian pollution does not have a large potential to affect the surface fine particle mass concentrations on other continents despite its high emission rates.

European pollution, on the other hand, is more widely spread than the Asian pollution (3rd panel in Fig. 8a). It provides about 0.1–0.2 $\mu\text{g m}^{-3}$ of ammonium sulfate to the entire Arctic surface, 0.2–2 $\mu\text{g m}^{-3}$ to northern Asia, and 0.2–0.5 $\mu\text{g m}^{-3}$ to eastern China. Our budget analysis shows that the amount of ammonium sulfate from European pollution on average can be 60% of that from Asian pollution over the U.S., with its strongest influence over the NE. The region which is particularly vulnerable to European pollution is northern Africa, simply because of the short traveling distance between these two continents. As much as 5 $\mu\text{g m}^{-3}$ of ammonium sulfate over the northern African countries adjacent to the Mediterranean Sea is attributed to the European pollution source, which accounts for 50–80% of the surface sulfate budget there. It is also notable that European pollution contributes more to the surface ammonium sulfate increase over East Asia (0.2–0.5 $\mu\text{g m}^{-3}$) than Asian emission contributes to North America (0.1–0.2 $\mu\text{g m}^{-3}$) on an annual average in 2001. The export of Euro-

pean pollution often occurs in the boundary layer (e.g. Wild et al., 2001) and contains a higher fraction of SO₂ than does the outflow from either Asia or North America (Chin and Jacob, 1996; Chin et al., 2000b), a combination that allows continued production of sulfate within the boundary layer during transport and thus produces a broader influence on the surface sulfate concentrations.

Relative to the spatial spreading of Asian and European sulfate, North America pollution is more regionalized (4th panel in Fig. 8a), but its contribution to the surface ammonium sulfate over northern Africa and Saudi Arabia (0.1–0.2 μg m⁻³) is similar to the contribution from Asia to the western U.S. It has been suggested that the North American influence over northern Africa and the Middle East is due to the deep boundary layer mixing over those arid regions that entrains the plume from aloft (Li et al., 2001).

Our results for total and “background” ammonium sulfate surface concentrations and the Asian influences over the U.S. are similar to the equivalent quantities from the GEOS-CHEM model simulations that used the same meteorological fields for the same year (Park et al., 2004), as summarized in Table 4. The major difference between the two studies is the European influence over the eastern U.S.: we have found that the pollution transport from Europe and Asia is of comparable importance, but the budget shown in Park et al. (2004, Table 2) implies negligible influence from European pollution, although the European budget was not explicitly calculated in that study. Differences in emission and budget classification from our different model experiment designs may explain the apparent discrepancy, which is expected to be resolved through the modeling exercise in the current United Nation Task Force assessment of Hemispheric Transport of Atmospheric Pollutants (<http://www.htap.org>), in which common pollution regions of North America, Europe, East Asia, and South Asia are defined by the Task Force for all participating models including GOCART and GEOS-CHEM.

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5.2 Dust transport

The annual surface concentrations of fine dust and contributions from major source regions in Asia, Middle East, and North Africa are shown in Fig. 8b. A clear dust band with fine dust concentrations above $5 \mu\text{g m}^{-3}$ expands from the sub-tropical North Atlantic to northeastern Asia (1st panel in Fig. 8b). Away from the source regions, Asian dust seems to have the largest influence on the surface fine dust over extra-tropical North Pacific and North America, with a contribution of $0.2\text{--}1 \mu\text{g m}^{-3}$. The concentration gradient across the Pacific for dust is a factor of about 20 (e.g., the dust concentration off the coast of China is about 20 times higher than that at the west coast of North America), which is much smaller than the concentration gradient of sulfate that we saw earlier, suggesting a more efficient trans-Pacific transport of dust than of sulfate. This is because the higher elevation of the Asian dust plume (above the boundary layer upon leaving Asia, e.g., Anderson et al., 2003; Chin et al., 2003) and the lower wet scavenging efficiency results in slower loss rate of dust than of sulfate during trans-Pacific transport. Over the Arctic, above 50% (or $>0.1 \mu\text{g m}^{-3}$) of the surface fine dust is from Asia, an amount that is comparable to the European pollution contribution to Arctic sulfate (Fig. 8a). These results have yet to be verified with surface measurements, although the optical measurements in April 2002 at the Barrow Alaska baseline observatory detected a clear dust existence for several days that was traced back to the Gobi desert by a trajectory analysis (Stone et al., 2005).

Dust emitted from the Middle East has the highest impact over India, northeastern Africa, and North Indian Ocean with an increase of $1\text{--}5 \mu\text{g m}^{-3}$ (3rd panel in Fig. 8b). The dust emitted from this region can travel to northern and eastern Asia as well as to eastern Africa. The northward transport delivers dust that accounts for more than 10% of the surface fine dust concentrations in the Arctic.

The vast desert area over Africa is the largest dust source on the globe. Much of this dust is transported westward across the Atlantic under the influence of trade winds to arrive at Central America and equatorial South America, whereas the northward

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transport over the Mediterranean is linked to the presence of cyclones (Prospero, 1996; Moulin et al., 1997) to reach southern Europe. Both pathways bring $1\text{--}5\ \mu\text{g m}^{-3}$ of fine dust to the surface of affected areas (4th panel in Fig. 8b). There is also an eastward transport pathway that takes the African dust to Asia, traveling a long distance in the mid-tropospheric westerlies across the North Pacific to eventually subside in western North America, raising fine dust levels by $0.2\text{--}0.5\ \mu\text{g m}^{-3}$ at the surface. While the trans-Atlantic and cross-Mediterranean transport of African dust is frequently detected by satellite (e.g., MODIS data in Fig. 1), the trans-Pacific transport of African dust is difficult to recognize because it easily loses its distinct characteristics after merging with the Asian dust. A recent study has reported detecting of long-range transport of Saharan dust through a pathway spanning Asia and North Pacific to western North America in the absence of any Asian dust activities (McKendry et al., 2007); this finding is supported by our modeling results.

6 Summary and conclusions

In summary, we have used the GOCART model to estimate the origins of the surface fine aerosol particle concentrations over the U.S. that are generated from North American industrial pollution, biomass burning, and natural sources, as well as from intercontinental transport from other major source regions. Comparisons of model-simulated surface concentrations of sulfate, OC, BC, fine dust, and total RCFM with observations at 142 sites in the IMPROVE network have revealed that the model captures the spatial and temporal variations of the observed sulfate with high correlation and low bias, but predicts higher fine dust and lower non-biomass burning carbonaceous aerosols than the IMPROVE derived data. This discrepancy may be explained in part by model deficiencies and in part by the relatively high uncertainties in the data. The quantity of RCFM from the IMPROVE sites, which is equivalent to $\text{PM}_{2.5}$ in most places in the U.S., agrees with the model in both concentration and spatial variation. The model estimates that ammonium sulfate in the eastern U.S. is the dominant component ($\sim 60\%$)

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of the surface RCFM in all seasons, whereas in the western U.S. dust and OM are just as important as sulfate but have considerable seasonal variations especially in the NW. The annual averaged RCFM concentrations in the NW, SW, NE, and SW quadrants in the U.S. are calculated at 3.43, 4.91, 5.36, 8.32 $\mu\text{g m}^{-3}$, respectively, for 2001.

5 By targeting individual source types and source regions in our model experiments, we have been able to place the surface RCFM in the context of regional industrial pollution, biomass burning, natural emissions, and intercontinental transport. We have found that North America regional pollution accounts for nearly 30 to 40% of the surface RCFM in the western U.S. and much more in the eastern U.S. with 65 to 70%. In contrast, pollution transport from outside North America only contributes to 2 to 6% of the total RCFM over the U.S., with the NW being the most affected area.

10 Of the “background” RCFM, namely the RCFM from sources other than North American pollution, fine dust is the most important component in the western U.S., comprising 30–40% of the total surface RCFM in that region. While almost half of the fine dust in the NW is from Asia, more than 60% in the SW is from the local desert areas. In the eastern U.S., however, natural aerosols, mostly OM from terpene oxidation, are the most significant contributor to background RCFM, especially in the summer. Biomass burning aerosols account for about 10–27% of the background aerosol on an annual average, yet they have shown very strong seasonal variations and could well become the predominant RCFM component during the burning season. There is also a large seasonal variation for fine dust over the U.S from long-range transport that peaks in the springtime.

20 The total amount of imported RCFM from outside North America consists of dust and pollution, with the proportion of dust being 2 to 4 times higher than that of pollution. The total import is a factor of about 2 to 10 times smaller than the “native” RCFM which includes pollution, biomass burning, and natural aerosols originating in North America. It is also much smaller than the native pollution RCFM, except in the NW. We should note that large episodic events, such as dust storms in Asia, can create intermittent air quality problems over North America, especially over the west coast, even though

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these events are usually short-lived. As a result of the above analysis, it seems very clear that controlling North America pollution emission is both the most responsible and the most effective way to improve air quality within the region.

We have also demonstrated the extent to which pollution and dust emissions from their major source regions may affect PM surface concentrations on a hemispheric scale. Of the three major industrial pollution regions, Asia, Europe, and North America, we have found that on an annual average, Asian pollution emission contributes to more than 30% of the surface sulfate over an extended area in the North Pacific and North Indian Oceans and to between 10 and 30% over the Arctic, but to no more than 10% everywhere else, including North America. North America pollution exerts a strong impact on the western North Atlantic but generates only a limited influence on other continents, even though North Africa and the Middle East seem to receive more North American pollution than other places do. In comparison, European pollution has the largest potential to affect fine particulate levels over other continents due to the shorter traveling distance to receptor continents and larger fraction of sulfate-producing precursor gas in the predominantly boundary layer outflow. While the European pollution exerts its largest influence over northern Africa, it can increase the surface ammonium sulfate concentration over eastern Asia by $0.2\text{--}0.5\ \mu\text{g m}^{-3}$, an amount which is twice as high as the level of the Asian pollution impact we have found on western North America. Interestingly, the amount of the pollution ammonium sulfate Europe sends to northern Africa is about the same as the amount of fine dust Africa sends to southern Europe ($1\text{--}5\ \mu\text{g m}^{-3}$) on an annual average.

Of the three major dust source regions, eastern Asia displays the greatest influence over North Pacific, North America, and the Arctic, whereas Africa dominates the entire North Atlantic, tropical Pacific, Central America, and tropical South America. Considering nearly 3 times lower emission rate (Table 1), Asian dust therefore has a larger “impact potential” than African dust to affect a wide geographic area. We have also found that the trans-Pacific transport of Asian dust is much more effective than that of Asian sulfate because of the higher plume elevation and lower loss of dust during

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transport. We have noticed that even though the transport of African dust mostly takes place across the North Atlantic and Mediterranean regions, it can go eastward through a pathway spanning Asia and the Pacific to settle down over the surface of western North America. The smallest influential dust region is the Middle East, whose impacts are mostly sustained by its neighbors of the northern Indian Ocean, Eastern Europe, and India, even though it can increase the fine dust over the Arctic surface by about 20%.

The scope of the intercontinental transport is controlled by several factors, the most important of which are emissions from the source regions, convective lifting of the aerosol and its precursors into the free troposphere, advection in both the free troposphere and boundary layer, wet and dry deposition during transport, and subsidence over the receptor regions. The relative importance of these factors varies significantly with time and space, although we have mostly focused on the surface- and time-averaged features in this paper. The time of the greatest impact of the long-range transport is spring, when horizontal transport rapidly spreads pollutants and dust over an extended area in the Northern Hemisphere, including the Arctic.

The pollutant emission we used in this work is based on the “present day emission scenario” used in the IPCC 2001 Third Assessment Report (Penner et al., 2001). As economic growth and environmental regulations in the major industrial regions may have changed since then and will change in the future, pollutant emissions and consequently the extent of their impacts for different time periods are expected to be different from the ones in this study. Dust and biomass burning emissions have considerable year-to-year variations; as such, the year of 2001 may not be a “typical” year to generalize the impact. In addition, our model results need to be further evaluated with observations, especially in Asia and Arctic, but data in those areas are sparse or not easily accessible. Lastly, results from other models may deviate from ours, as several intercomparison studies have shown considerable differences in aerosol composition and vertical distribution (Barrie et al., 2001; Penner et al., 2002; Textor et al., 2006). Nonetheless, our study should provide guidance not only for current regional air quality

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management but also for projecting future intercontinental impacts.

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Table 1. Annual emissions of aerosols and precursors in this study.

Sulfur (TgS yr ⁻¹) ^a Total	90.7
Pollution	64.9
Asia	26.3
Europe	17.3
North America	12.3
Other	9.0
Biomass burning	3.5
Natural	22.3
Ocean	13.6
Volcano	8.7
OC (TgC yr ⁻¹) Total	82.3
Pollution	19.6
Asia	9.4
Europe	6.1
North America	1.0
Other	3.1
Biomass burning	50.0
Natural ^b	12.7
BC (TgC yr ⁻¹) Total	14.1
Pollution	7.8
Asia	3.6
Europe	2.5
North America	0.7
Other	1.0
Biomass burning	6.3
Dust (Tg yr ⁻¹) ^c Total	2773
Asia	566
Middle East	443
Northern Africa	1609
Other	155
Sea-salt (Tg yr ⁻¹) ^c Total	9838

^aSulfur source includes SO₂ (~97%) and sulfate (~3%) from industrial and transportation emissions, dimethyl sulfide (DMS) from ocean, SO₂ from biomass burning and volcanoes.

^bNatural OC is from terpene oxidation.

^cDust and sea-salt emissions include the size range up to 10 μm in radius.

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Table 2. Statistics between the model-calculated and IMPROVE measured monthly mean concentrations of aerosol species at 142 IMPROVE sites over the U.S.

	R	B	S
Sulfate	0.815	1.154	0.879
BC	0.519	0.884	0.680
OC	0.454	0.568	0.721
Fine dust	0.577	1.747	0.687
RCFM	0.742	0.940	0.870

R=correlation coefficient; B=relative bias; S=Skill score computed from standard deviations and correlation coefficient (Taylor, 2001; also see Chin et al., 2003, 2004).

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Table 3. Model-calculated annual averaged concentrations of RCFM and its composition and origins for the four quadrants in the U.S.

Conc. ($\mu\text{g m}^{-3}$)	NW		SW		NE		SE	
RCFM	3.43		4.91		5.36		8.32	
Composition:								
(NH ₄) ₂ SO ₄	1.03	(29.9%)	1.70	(34.7%)	3.31	(61.7%)	4.98	(59.9%)
OM	1.27	(37.1%)	1.05	(21.4%)	1.21	(22.5%)	2.33	(28.0%)
BC	0.15	(4.3%)	0.24	(4.8%)	0.32	(5.9%)	0.39	(4.7%)
DU _f	0.99	(28.8%)	1.93	(39.2%)	0.53	(9.9%)	0.62	(7.4%)
Origin:								
NAM	0.95	(27.7%)	1.87	(38.1%)	3.79	(70.7%)	5.43	(65.2%)
BMB	0.61	(17.8%)	0.32	(6.5%)	0.21	(3.9%)	0.78	(9.4%)
NAT	0.66	(19.3%)	0.60	(12.3%)	0.61	(11.4%)	1.31	(15.7%)
LDU _f	0.17	(4.9%)	1.22	(24.8%)	0.03	(0.5%)	0.05	(0.6%)
TDU _f	0.82	(23.9%)	0.72	(14.6%)	0.50	(9.4%)	0.56	(6.7%)
TAP	0.22	(6.3%)	0.20	(4.0%)	0.22	(4.1%)	0.19	(2.3%)

NAM=North America pollution; BMB=Biomass burning; NAT=Natural sources (biogenic and volcanic); DU_f=fine dust ($d < 2.5 \mu\text{m}$); LDU_f=fine dust from local source; TDU_f=fine dust from intercontinental transport; and TAP=pollution aerosol from intercontinental transport. Total “background” is the amount of aerosols not from North America pollution source (RCFM–NAM), total “transport” aerosol is the sum of TDU_f and TAP, total pollution aerosol is the sum of NAM and TAP, and total “native” is the aerosol from the North American sources (NAM+BMB+NAT).

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Table 4. Comparison of model-calculated annual-average ammonium sulfate concentration ($\mu\text{g m}^{-3}$) over the western and eastern U.S. from this study and Park et al. (2004).

Concentration ($\mu\text{g m}^{-3}$)	Park et al. (2004)		This study	
	Western U.S.	Eastern U.S.	NW/SW U.S.	NE/SE U.S.
Total Concentration	1.52	4.11	1.03/1.70	3.31/4.98
Background ¹	0.28	0.24	0.29/0.30	0.25/0.26
From Asia	0.13	0.12	0.12/0.11	0.07/0.08
From Europe ²	≤ 0.04	≤ 0.01	0.06/0.05	0.08/0.06

¹The background value in this study is defined as the amount of ammonium sulfate from non-North American pollution sources, which is equivalent to the values of “background from non-U.S. anthropogenic emission” minus the transboundary transport from Canada and Mexico to the U.S. in Park et al. (2004) (western: $0.43-0.15=0.28 \mu\text{g m}^{-3}$, eastern: $0.38-0.14=0.24 \mu\text{g m}^{-3}$).

²Although the amount from Europe is not explicitly listed in Park et al. (2004), the difference between “Transboundary transport” and “Natural” in Table 2 of Park et al. (2004) implies the maximum amount of ammonium sulfate transported from Europe (R. Park, personal communication).

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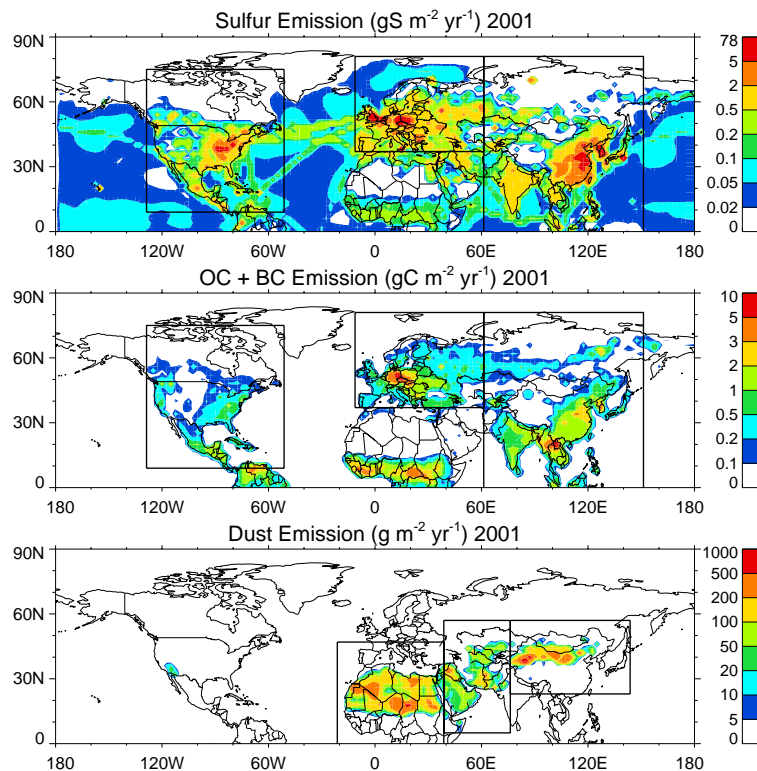


Fig. 1. Annual average emissions of sulfur (SO_2 , sulfate, and DMS), carbonaceous (BC and OC), and dust in 2001 used in the GOCART model. Domains of three major industrial pollution regions of Asia, Europe, and North America are shown in the 1st and 2nd panels and three major dust regions of Asia, Middle East/Central Asia, and northern Africa are shown in the 3rd panel.

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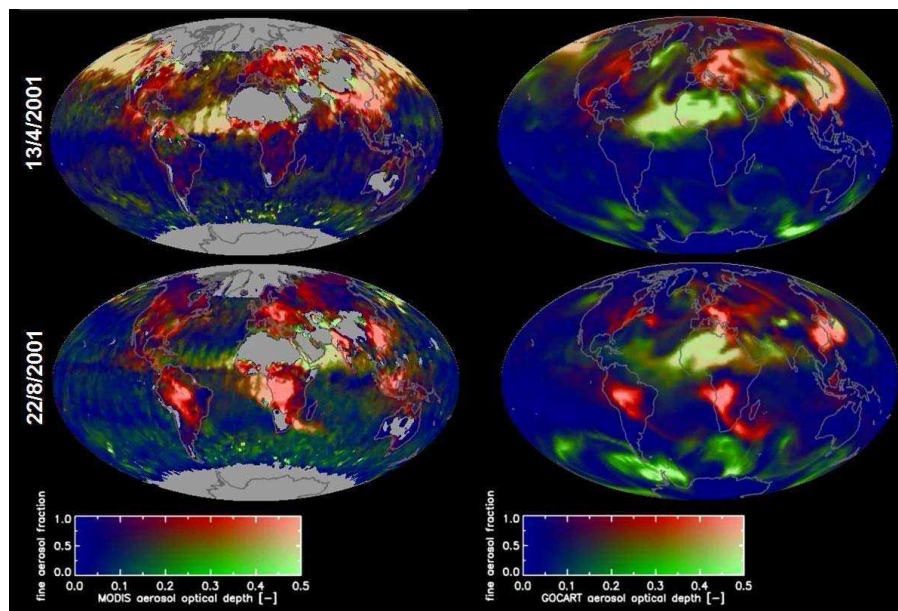


Fig. 2. Aerosol optical depth observed by the MODIS instrument on EOS-Terra (left column) and simulated by the global model GOCART (right column) for 13 April (top row) and 22 August (bottom row), 2001. Red color indicates fine mode aerosols (e.g., pollution and smoke) and green color coarse mode aerosols (e.g., dust and sea-salt). Brightness of the color is proportional to the aerosol optical depth. On 13 April 2001, there are heavy dust and pollutions transported from Asia to the Pacific and dust transported from Africa to Atlantic; while on 22 August large smoke plumes from South America and Southern Africa are evident. Figure credit: Y. Kaufman and R. Stöckli.

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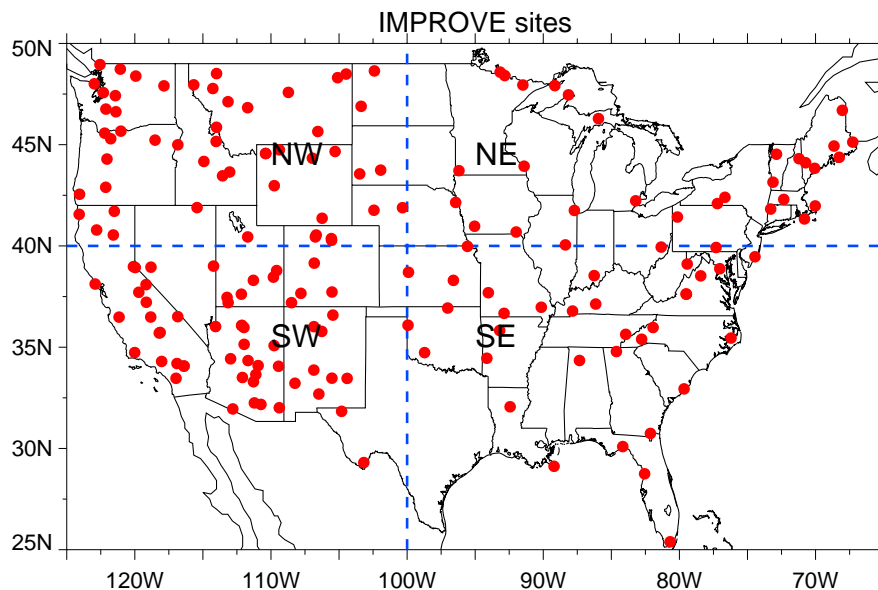


Fig. 3. Location of IMPROVE sites in 2001 and dividing lines of the NW, SW, NE, SE quadrants.

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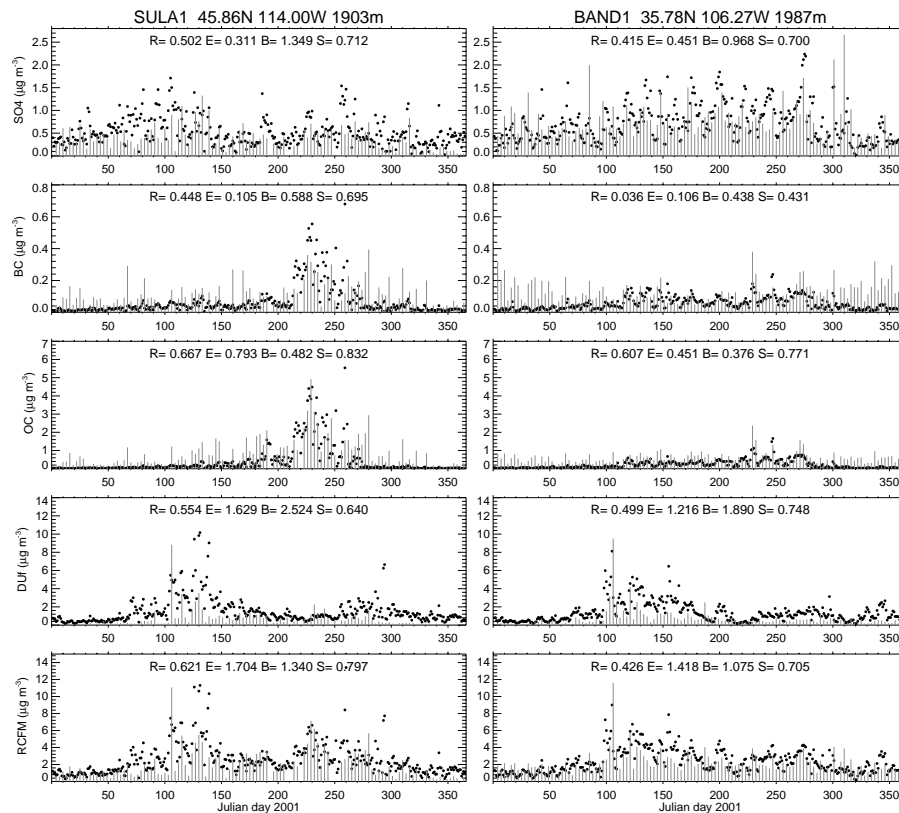


Fig. 4a. Daily concentrations of sulfate, BC, OC, fine-mode dust, and reconstructed fine mass (RCFM) at IMPROVE sites of Sula, Montana (SULA1) in the NW sector (see Fig. 1) and Bandelier National Monument, New Mexico (BAND1) in the SW sector in 2001. Vertical grey line: IMPROVE data; black dots: GOCART model simulations. Showing on each panel: R=correlation coefficient, E=standard error, B=relative bias, and S=skill score which is computed from standard deviations and correlation coefficient (Taylor, 2001; also see Chin et al., 2003, 2004).

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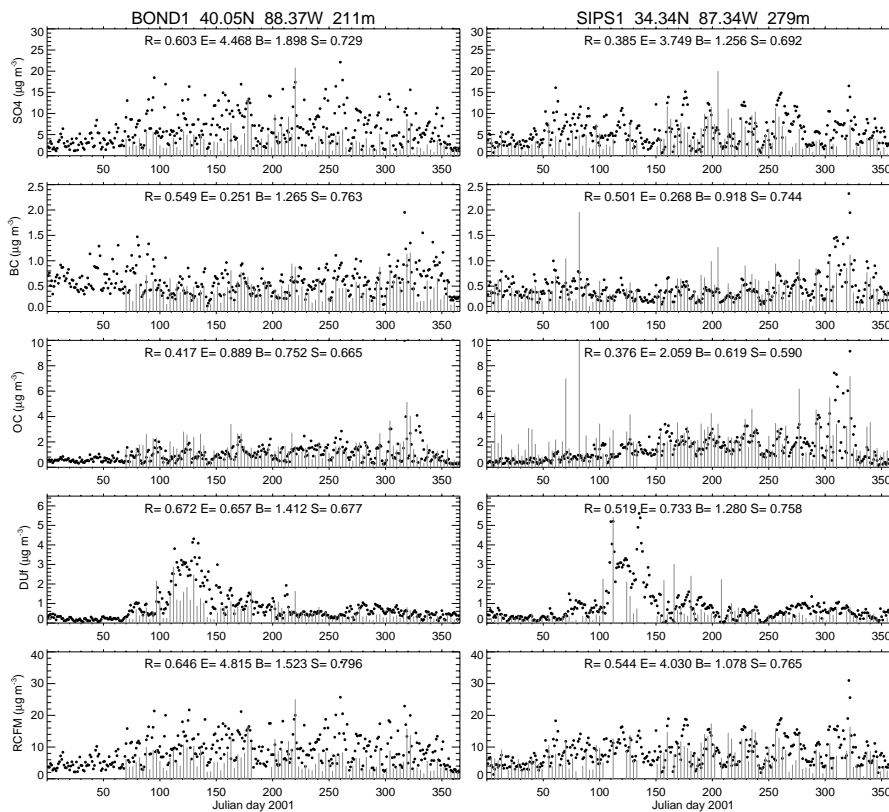


Fig. 4b. Same as in Fig. 4a but at Bondville, Illinois (BOND1) in the NE sector and Sipsy Wilderness, Alabama (SIPS1) in the SE sector.

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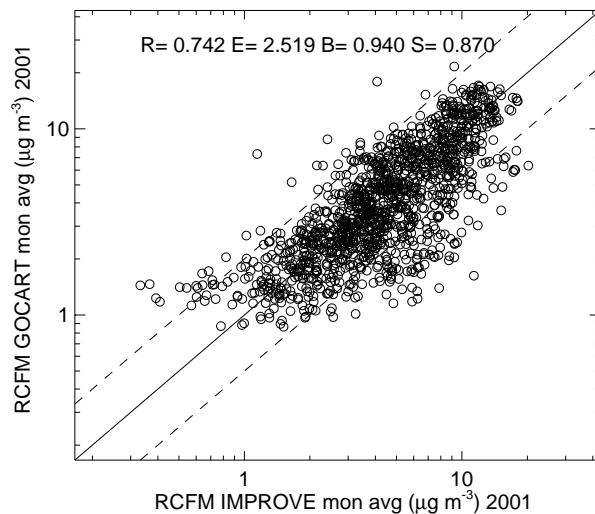


Fig. 5. Scatter plot of monthly averaged surface RCFM concentrations over the IMPROVE sites (location in Fig. 3).

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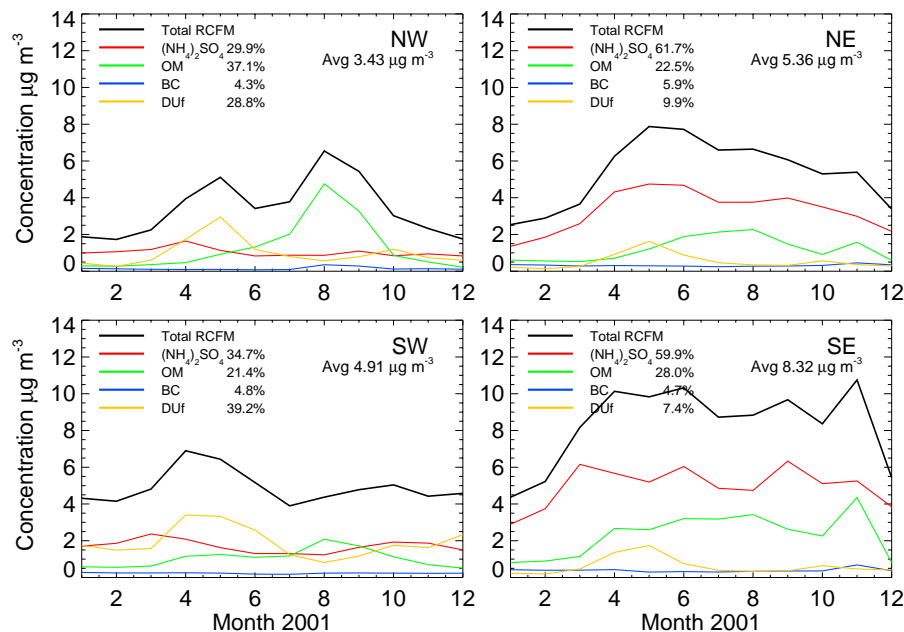


Fig. 6. Surface RCFM annual cycle and composition in NW, SW, NE, and SW of the United States (see Fig. 3 for the domain).

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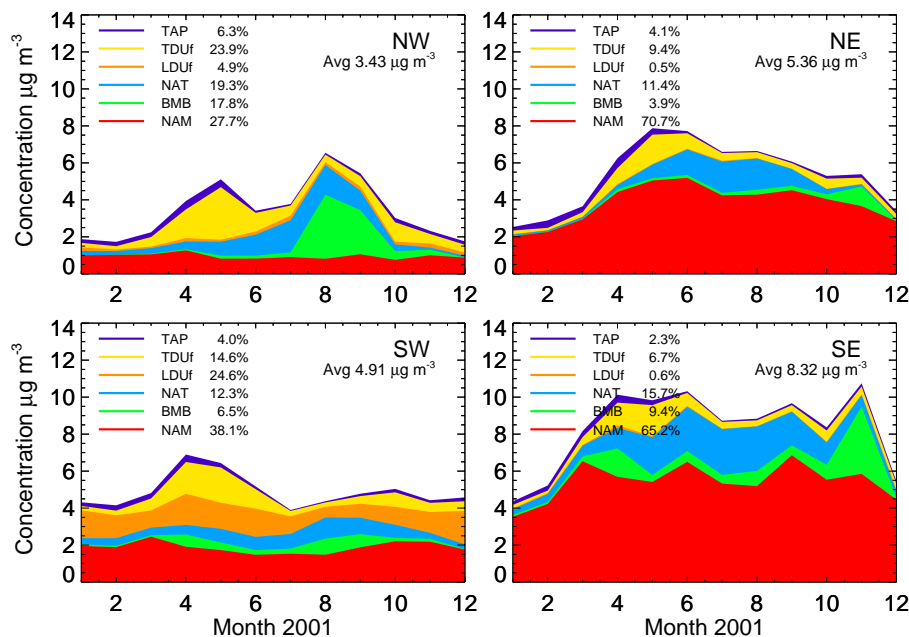


Fig. 7. Monthly and regional average of surface aerosol fine mass in NW, SW, NE, and SW (domain in Fig. 3) in the U.S. from different origins: TAP, transported air pollutants from outside North America; TDU_f, imported fine-dust from outside North America; LDU_f, local fine-dust; NAT, biogenic (organic matter and sulfate); BMB, biomass burning; NAM, North American pollution.

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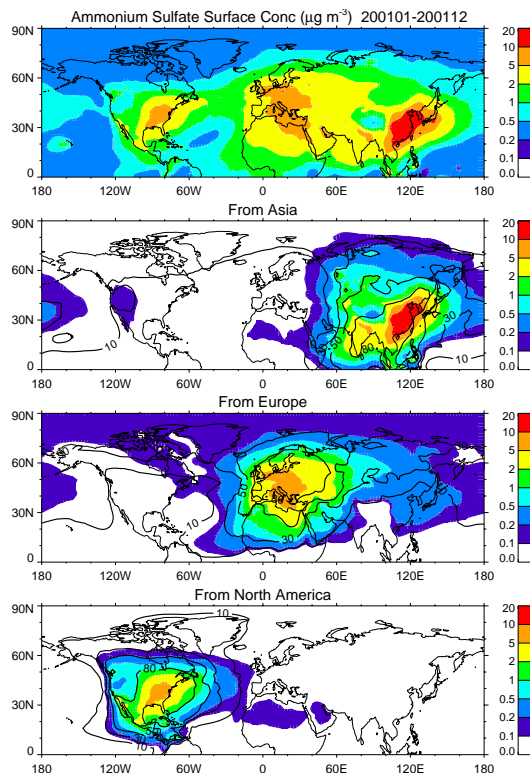


Fig. 8a. Annual average surface ammonium sulfate concentrations in the Northern Hemisphere in 2001 from the GOCART model (top panel) and the amount from major source regions of Asia (2nd panel), Europe (3rd panel), and North America (last panel). Regional domains are shown in Fig. 3. Color scales are concentrations in $\mu\text{g m}^{-3}$, and the contour lines are the percentage contributions to the total ammonium sulfate in 10, 30, 50, 80% intervals.

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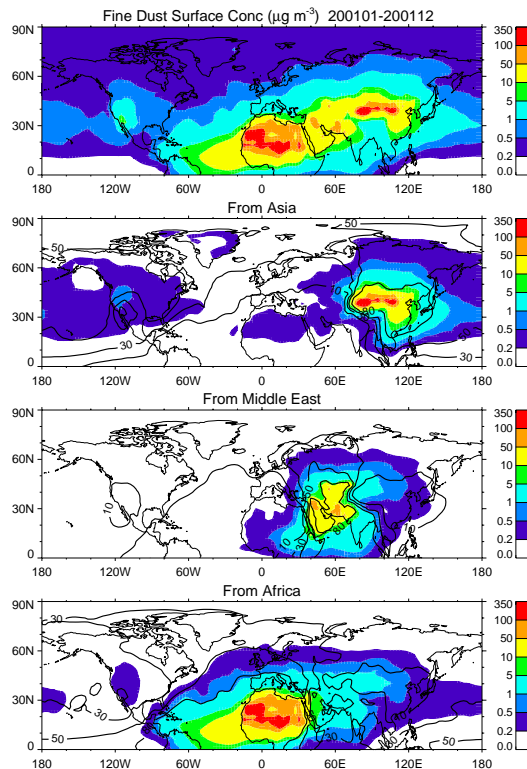


Fig. 8b. Annual average fine dust concentrations in the Northern Hemisphere in 2001 from the GOCART model (top panel) and the amount from major source regions of Asia (2nd panel), Middle East/Central Asia (3rd panel), and northern Africa (last panel). Regional domains are shown in Fig. 3. Color scales are concentrations in $\mu\text{g m}^{-3}$, and the contour lines are the percentage contributions to the total ammonium sulfate in 10, 30, 50, 80% intervals.

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