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Source-receptor relationships between East Asian sulfur dioxide emissions and Northern Hemisphere sulfate concentrations

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Abstract

We analyze the effect of varying East Asian (EA) sulfur emissions on sulfate concentrations in the Northern Hemisphere, using a global coupled oxidant-aerosol model (MOZART-2). We conduct a base and five sensitivity simulations, in which sulfur emissions from each continent are tagged, to establish the source-receptor (S-R) relationship between EA sulfur emissions and sulfate concentrations over source and downwind regions. We find that from west to east across the North Pacific, EA sulfate contributes approximately 80%–20% of sulfate at the surface, but at least 50% at 500 hPa. In addition, EA SO_2 emissions account for approximately 30%–50% and 10%–20% of North American background sulfate over the western and eastern US, respectively. The contribution of EA sulfate to the western US at the surface is highest in MAM and JJA, but is lowest in DJF. Reducing EA SO_2 emissions will significantly decrease the spatial extent of the EA sulfate influence over the North Pacific both at the surface and at 500 mb in all seasons, but the extent of influence is insensitive to emission increases, particularly in DJF and JJA. We find that EA sulfate concentrations over most downwind regions respond nearly linearly to changes in EA SO_2 emissions, but sulfate concentrations over the EA source region increase more slowly than SO_2 emissions, particularly at the surface and in winter, due to limited availability of oxidants (mostly H_2O_2). We find that similar estimates of the S-R relationship for trans-Pacific transport of EA sulfate would be obtained using either sensitivity or tagging techniques. Our findings suggest that future changes in EA sulfur emissions may cause little change in the sulfate induced health impact over downwind continents but SO_2 emission reductions may significantly reduce the sulfate related climate cooling over the North Pacific and the United States.

1 Introduction

Sulfate aerosol is an important component of fine particulate matter ($PM_{2.5}$, diameter $\leq 2.5\,\mu m$), which is associated with increased risk of adverse health outcomes including premature mortality (Pope et al., 2002; Pope et al., 2004). In addition, sulfate aerosols scatter sunlight, reduce visibility, affect regional climate, and harm ecosystems (Gunther, 1992; Giorgi et al., 2003; Park et al., 2004; Marmer et al., 2007; Koch et al., 2007a). Sulfate aerosols are produced by gas phase oxidation of SO_2 by OH radicals and by aqueous phase oxidation of SO_2 by H_2O_2 (Martin and Damschen, 1981) and O_3 (Feichter et al., 1996). Since most anthropogenic sulfate is produced by oxidation of SO_2, many industrialized nations have stringently regulated SO_2 emissions to protect human health and ecosystems (Dutkiewicz et al., 2000; Moldan et al., 2001). However, the benefits of sulfur emission control partly depend on the linearity of the source-receptor (S-R) relationship between SO_2 emissions and sulfate concentrations (Oppenheimer et al., 1985; Dutkiewicz et al., 2000). Generally, the relationship between SO_2 emissions and sulfate concentrations is linear near minor SO_2 sources, but nonlinear near major SO_2 sources because of limited availability of oxidants (Hilst, 1992; Berglen et al., 2004).

Due to rapid industrialization, anthropogenic SO_2 emissions from East Asia (EA), particularly China, have increased substantially in recent decades and are projected to increase further by nearly 50% between 1990 and 2030 (Klimont et al., 2001). However, China has realized that it is urgent to mitigate SO_2 emissions and has designed the Acid Rain Control Zone and SO_2 Pollution Control Zone Program (i.e. the Two Control Zone Strategy) to efficiently control sulfur pollution (Hao et al., 2001). Therefore, future SO_2 emissions from EA are highly uncertain and significant increases or decreases are possible (Streets, 2007). These changes underscore the need to establish quantitative S-R relationships between EA SO_2 emissions and sulfate concentrations over source and downwind regions.

Typical methods for reducing SO_2 emissions include switching fuels or removing sulfur from fuels (e.g., replacing high-sulfur with low-sulfur coal; removing sulfur from gasoline) and reducing end-of-pipe emissions (e.g., installing scrubbers) (Conrad and Kohn, 1996; NAPAP, 2005; Carlson et al., 2000). These strategies have been widely used in the US to reduce SO_2 emissions (Carlson et al., 2000), and are believed to be the affordable strategies by the Two Control Zone Program to mitigate SO_2 emissions in China (Hao et al., 2001). Unlike improving energy efficiency (which reduces energy use), reducing SO_2 emissions by using low-sulfur coal or scrubbers does not reduce other pollutants, such as NO_x and VOCs (which are the precursors for atmospheric oxidants, such as OH, O_3, H_2O_2) or carbon dioxide (CO_2) the primary greenhouse gas. Therefore, in this study we only change SO_2 emissions and leave the emissions of other chemical species unchanged.

Two techniques are used by the atmospheric modeling community to establish the effect of regional emissions on global concentration distributions, namely tagging emission tracers (Liu and Mauzerall, 2005, 2007; Liu et al., 2005) and conducting sensitivity studies (Chin et al., 2007; Park et al., 2004; Koch et al., 2007b; Heald et al., 2006). The results using these two approaches can differ depending on the linearity of the chemical conversion of SO_2 to sulfate between the source and receptor regions. In this study, we use a coupled tagging-sensitivity approach to quantify the S-R relationship and compare the difference between these two techniques. Our objectives are: (1) to determine the effect of potential future increases or decreases in EA SO_2 emissions on sulfate concentrations over downwind regions (Sect. 3); (2) to quantify the linearity of the S-R relationship between EA SO_2 emissions and sulfate concentrations globally (Sect. 4); and (3) to compare the source-receptor relationships obtained using tagging and sensitivity studies (Sect. 5).

2 Methods

We use the three-dimensional global chemical oxidant-aerosol (fully coupled) transport model MOZART-2 (Model of Ozone and Related Tracers, version 2) (Horowitz et al.,

ACPD
8, 5537–5561, 2008

2003) driven with NCEP/NCAR reanalysis meteorology to simulate inter-continental transport of sulfate aerosols. The model is configured with a T62 (1.9°×1.9°) horizontal resolution and 28 hybrid vertical levels from the surface to 2.7 mb. Standard MOZART-2 emission inventories are used which represent global emissions in the early 1990s (Olivier, 1996; Horowitz et al., 2003). The sulfur emissions from EA account for approximately 21% of global total sulfur emissions. Detailed descriptions of the model and model evaluation are provided by Horowitz et al. (2003; 2006), Tie et al. (2005) and Ginoux et al. (2006).

MOZART-2 simulates sulfate production including both gas phase oxidation of SO_2 by OH radicals and aqueous phase oxidation of SO_2 by H_2O_2 (Martin and Damschen, 1981) and O_3 (Feichter et al., 1996). In addition, naturally produced dimethyl sulfide (DMS) is oxidized to SO_2 by gas-phase reactions with OH and NO_3 radicals. Aqueous oxidation of SO_2 to sulfate depends on cloud water content and acidity, temperature, and abundance of oxidizing agents (namely H_2O_2 and O_3). When clouds are present, MOZART-2 first predicts the pH values based on the mixing ratios of SO_2, CO_2, HNO_3, sulfate, and NH_3. It then uses the predicted pH and temperature to calculate the temperature dependent effective Henry's Law coefficients and aqueous reaction rate coefficients (Tie et al., 2005). When the cloud pH is below 5, the reaction rate between SO_2 and H_2O_2 is much faster than that between SO_2 and O_3 (Brasseur et al., 1999; Seinfeld and Pandis, 1998). Removal processes for sulfur species include both dry deposition and wet scavenging. The dry deposition velocities for SO_2 are from Feichter et al. (1996) and are much faster over ocean (0.8 cm/s) and land (0.6 cm/s) than over snow (0.2 cm/s). The dry deposition velocities for sulfate are 0.2 cm/s (Feichter et al., 1996) over all surfaces. Wet deposition includes both in-cloud rainout and below cloud washout (Horowitz et al., 2003). In MOZART-2, the wet deposition rate for SO_2 is set equal to that of H_2O_2. For sulfate, the wet deposition rates are set to 20% of that for the highly soluble gas HNO_3 (Horowitz, 2006).

In this study, we quantify S-R relationships using a coupled approach including tagged tracers and sensitivity simulations. We first conduct a baseline simulation with standard sulfur emissions. We then conduct five sensitivity simulations with anthropogenic sulfur emissions from EA reduced by 80%, 50%, 20%, and increased by 20% and 50% relative to the base simulation. In all cases, we tag the anthropogenic emissions of SO_2 from the ten continental regions including North America (NA), South America (SA), Europe (EU), the Former Soviet Union (FSU, excluding part of Russia in the European domain), Africa (AF), Indian Subcontinent (IN), East Asia (EA), Southeast Asia (SE), Australia (AU), and the Middle East (ME) as shown in Fig. 1 and track their conversion to sulfate (SO_4^{2-}). Each simulation covers the 2-yr period from 1990 to 1991 with the first year used for initialization.

Since this study uses meteorological inputs from the NCEP reanalysis (rather than MACCM-3 as in Horowitz et al., 2003), we then evaluate simulated aerosol concentrations by comparing the model results with various observations for sulfate, including the data collected by the Rosenstiel School of Marine and Atmospheric Science (RSMAS) at the University of Miami (Prospero, 1996) and by regional observation networks, namely the Interagency Monitoring of Protected Visual Environments (IMPROVE) in the United States (http://vista.cira.colostate.edu/IMPROVE/), the Cooperative Program for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP: http://www.emep.int), and the East Asian Monitoring Network (EANET: http://www.eanet.cc). We average observations over 1980s–1990s for RSMAS data, over 1990 to 1992 for IMPROVE and EMEP data, and over 2000 to 2004 for EANET (since many EANET observation stations did not begin to report data until 2003). Figure 2 compares the MOZART-2 simulated sulfate concentrations with global observation networks. The MOZART-2 results are generally in good agreement with observations, with annual mean sulfate concentrations within a factor of 2 of observations at 80% of the global stations considered.

3 Global contribution of sulfate aerosol from EA

The simulated surface concentrations of EA sulfate (SO_4^{2-}) range from more than 10 μg m^{-3} over EA to less than 0.5 μg m^{-3} over the western US (Fig. 3). Seasons associated with relatively high EA sulfate concentrations at the surface over the southwestern US (>0.1 μg m^{-3}) are MAM (the northern hemispheric spring) and JJA (the northern hemispheric summer). This differs from our previous findings, using idealized tracers, that trans-Pacific transport is strongest in winter-spring and weakest in summer (Liu et al., 2005; Liu and Mauzerall, 2005) . Comparing Fig. 3a and b, in summer the transport of EA sulfate to the central Pacific is weak at the surface but very strong at 500 mPa where strong westerlies prevail. The summer high EA sulfate concentration over the western US is the net result of a series of processes, including stronger convective transport over EA, faster sulfate production, faster wet removal, slower surface transport, and stronger subsidence (within summer highs) over the western US in summer than in winter. Most recent field work examining trans-Pacific transport (e.g. the 2002 Intercontinental Transport and Chemical Transformation campaign) and modeling research (e.g. Heald et al., 2006) has focused on spring. Our finding suggests that additional investigation of EA influence on sulfate concentrations over the western US in summer may be worthwhile.

Figure 4 shows the fractional contribution of sulfate concentrations from EA in the base simulation each season. From west to east across the North Pacific, EA sulfate contributes from 80% to 20% of total sulfate at the surface, but contributes at least 50% at 500 hPa. This indicates that EA sulfate is the dominant source of sulfate over the Pacific Ocean, particularly in the free troposphere. Sulfate aerosols directly scatter solar radiation and increase the albedo of clouds. They thus cool the Pacific air mass which could influence regional climate over the western US Investigation into linkages between changing EA sulfate concentrations over the North Pacific and their impact on US climate would be valuable. Over the surface of the US, EA sulfate contributes more than 10% of total sulfate over the western US in MAM and JJA, but its influence is

ACPD

8, 5537–5561, 2008

Source-receptor relationships for East Asian sulfate

J. Liu et al.

negligible over the eastern (<1%) due to the dominance of domestic sources (Fig. 4).

Figure 5 shows the fractional contribution of the North American background sulfate (i.e., total sulfate concentrations minus NA sulfate concentrations) from EA. At the surface, EA sulfate accounts for 30%–50% and 10%–20% of background sulfate over the western and eastern US, respectively. Due to the differences in model use, emission inventories and meteorological input, as well as the definition of "background sulfate" and "East Asian sources", our result differs slightly from that of Park et al. (2004), who find that total East Asian pollution accounts for 30% of background sulfate over both western and eastern US At 500 mPa, EA sulfate accounts for more than 50% of NA background sulfate over the US (Fig. 5a).

Figure 3 shows areas over the western US at both the surface and 500 hPa where for standard EA SO_2 emissions EA sulfate concentrations are at least 0.1 $\mu g\, m^{-3}$ ($A_{EA0.1}$), particularly in MAM and JJA. Figure 6 illustrates how the $A_{EA0.1}$ changes with increases/decreases in EA SO_2 emissions. At the surface, the spatial extent of $A_{EA0.1}$ is constrained to the North Pacific and is sensitive to EA emissions between 20% and 80% of standard EA emissions. Therefore, a decrease of EA SO_2 emissions will significantly decrease the spatial extent of EA sulfate over the surface of the North Pacific, but an increase in emissions will not significantly increase the horizontal extent of EA sulfate at the surface. The eastern boundary of $A_{EA0.1}$ reaches the western US at the surface only when EA SO_2 emissions are larger than half of the standard emissions (especially in MAM and JJA). Since most EA sulfate at low altitudes is efficiently removed by wet and dry deposition during transport over the North Pacific, further increases in EA sulfur emissions will not significantly increase sulfate concentrations at the surface over the US These results are supported by the measurements reported in Jaffe et al. (2005) and Prospero et al. (2003) which indicate that samples from Midway Island in the North Pacific are significantly more influenced by Asian industrial sources of sulfur than measurements at Crater Lake in Oregon. This implies that a substantial loss of sulfate from Asian sources occurs over the Pacific Ocean due to precipitation scavenging. At 500 hPa (Fig. 6a), where sulfate removal is slower, increasing EA emissions cause the eastern boundary of $A_{EA0.1}$ to expand from the western Pacific (20% EA emissions) to the eastern US (150% EA emissions), particularly in MAM and SON. In addition, the spatial extent of $A_{EA0.1}$ at 500 hPa is approximately twice as large as that at the surface in JJA. These findings suggest that future changes in EA sulfur emissions may cause little change in sulfate induced health effects over the US but may cause a significant change in the sulfate related climate impact over the North Pacific and the western US

4 Linearity of the S-R relationship

The linearity between SO_2 emissions and sulfate concentrations is of importance to both policymakers interested in reducing the impacts of sulfate exposure and scientists eager to understand the oxidation rate of SO_2 which depends on the concentrations of H_2O_2, OH, and O_3. Here we quantitatively investigate the linearity of the response of sulfate concentrations to changes in SO_2 emissions. As SO_2 emissions increase, non-linearity may arise as oxidants are consumed increasingly quickly hence reducing the production efficiency of sulfate (Berglen et al., 2004; Koch et al., 2007b)

As shown in Fig. 7, when the S-R relationship is linear, sulfate concentrations increase proportionally with the increase in SO_2 emissions (following line OA). However, if the increase in SO_2 emissions decreases the availability of oxidants, the S-R relationship will follow the OECF curve, which is non-linear. The change of slope along OECF indicates the change of atmospheric oxidation power. To quantify the linearity of the oxidation process, we define a cumulative linearity index (L) which indicates the percentage departure from linearity:

$$L = \frac{S_{OEC} + S_{CFA}}{S_{OAB}} \times 100\% \quad (1)$$

S_{OAB} is the area of the triangle OAB associated with a linear S-R relationship (Fig. 7); S_{OEC} ($S_{OECD} - S_{OCD}$) and S_{CFA} ($S_{CABD} - S_{CFBD}$) are the shaded areas in Fig. 7, indicating departure from linearity. When $L=0$, the S-R relationship is perfectly linear and the atmospheric oxidation power for SO_2 is unchanged. When $L\neq0$, the S-R relationship is non-linear. A larger $|L|$ value implies lower linearity and lower oxidant availability.

Figure 8 shows the distribution of $L_{0-1.5}$ (i.e., the linearity index obtained by varying EA emissions from 0 to 1.5 times the standard EA sulfur emissions) for the S-R relationship between EA SO_2 emissions and EA sulfate concentrations in the Northern Hemisphere. In regions where SO_2 emissions are large, oxidant limitation results in incomplete conversion to sulfate, causing sulfate concentrations to increase more slowly than SO_2 emissions; this results in the convex curve seen in Fig. 7 and a positive value for L. Over EA, high $L_{0-1.5}$ values (>10% and more than 20% over southeastern China) are found at the surface in all seasons, indicating a persistent non-linear S-R relationship over EA and significant variations of atmospheric oxidation power for SO_2. For example, in this study a 50% change (either increase or decrease) of EA sulfur emissions is associated with 5–10% (1–2%), 2–5% (0.5–2), and 1–2% (0.5–1%) change in surface H_2O_2, OH, and O_3 concentrations over EA (eastern Pacific) (not shown). As shown in Fig. 8b, over the western Pacific the $L_{0-1.5}$ values in JJA are low (<5%), indicating a higher atmospheric oxidation power for SO_2 than in other seasons. In addition, over the eastern Pacific and North America, the $L_{0-1.5}$ values are low (<5%) in most seasons except winter (at high latitudes where oxidant levels are low). At 500 hPa (Fig. 8a), the $L_{0-1.5}$ values are relatively low even over the EA source region. Therefore, the S-R relationships between EA SO_2 emissions and EA sulfate concentrations are close to linear everywhere except at the surface over EA.

5 Direct and indirect effects of the change in EA SO_2 emissions

Changing SO_2 emissions in EA leads to a direct change in EA sulfate and an indirect change in sulfate from other sources, which cannot be distinguished using sensitivity studies alone. Usually sensitivity studies represent the total change in sulfate concentrations between a perturbation run in which EA SO_2 emissions are changed and the base run. Using our tagged regional sulfur tracers we identify a direct change in EA sulfate concentrations (due to production and transport of sulfate from SO_2 emitted in EA). We also identify an indirect effect: the response of non-EA sulfate (i.e., the sulfate produced and transported from SO_2 emitted in regions other than EA) to the change in EA SO_2 emissions.

Figure 9 shows the direct and indirect changes in surface sulfate concentrations that result from increasing EA SO_2 emissions by 50%. Raising EA SO_2 emissions has the direct effect of increasing EA sulfate concentrations across the Pacific Ocean (Fig. 6a) and the indirect effect of decreasing non-EA sulfate concentrations over East Asia (Fig. 6b). The negative indirect effect results from a reduction in the concentration of oxidants (particularly H_2O_2) which in turn slows down the oxidation of non-EA SO_2. As shown in Fig. 9, the direct and indirect effects are both largest over the source region and diminish downwind. However, the direct effect exceeds the indirect effect by more than a factor of 10 over the source region and North Pacific (particularly between 20° N–50° N). Therefore, very similar trans-Pacific EA sulfate S-R relationships are obtained using tagging and sensitivity techniques. At 500 hPa, the magnitudes of the direct effect are 0.1–0.5 $\mu g\, m^{-3}$, 0.05–0.1 $\mu g\, m^{-3}$, and less than 0.05 $\mu g\, m^{-3}$ over the western Pacific, eastern Pacific and the United States (not shown). The indirect effect is negligible (<0.01 $\mu g\, m^{-3}$).

6 Conclusions

We analyze the source-receptor relationships between sulfur emissions from East Asia and the resulting sulfate concentrations over both source and downwind regions, using the global oxidant-aerosol model (MOZART-2). We conduct a base simulation and five sensitivity simulations in which EA sulfur emissions are varied. In each simulation, we tag sulfur species from EA and other continental regions.

We find that the contribution of EA sulfate to the western US at the surface (via trans-Pacific transport) is highest in both MAM and JJA (the magnitude of seasonal mean

EA sulfate concentrations over the western US is larger than 0.1 μg m^{-3}), but is lowest in DJF (<0.05 μg m^{-3}). This summertime high EA sulfate concentration is caused by the efficient production of sulfate aerosols over the EA source in conjunction with the convective elevation over the source region and relatively rapid trans-Pacific transport in the free troposphere. Since sulfate aerosol is an important component of $PM_{2.5}$ which is harmful to human health, our findings indicate that, from the health perspective, the summertime trans-Pacific transport of East Asian sulfate to the western US is as important as that in spring.

In terms of relative contribution, we find that present-day EA SO_2 emissions account for at least 50% (20%) of total sulfate concentrations over the whole North Pacific at 500 hPa (at the surface). Over the North American continent, if the NA sulfate is subtracted from the total sulfate (giving the NA background sulfate), the EA SO_2 emissions account for approximately 30–50% and 10–20% of NA background sulfate at the surface over the western and eastern US, respectively. At 500 hPa, the EA SO_2 emissions account for at least 50% of NA background sulfate over even the eastern US.

We compare the areas over which EA sulfate concentrations are at least 0.1 μg m^{-3} ($A_{EA0.1}$) in different sensitivity simulations. We find that reducing EA SO_2 emissions will significantly decrease the spatial extent of $A_{EA0.1}$ over the North Pacific at both the surface and 500 mPa, but the extent is insensitive to emission increases except for spring and fall at 500 hPa. In addition, the spatial extent of $A_{EA0.1}$ at 500 hPa is approximately twice as broad at the surface in JJA, indicating the efficient trans-Pacific transport of EA sulfate in the free troposphere in summer. These findings suggest that future change in EA sulfur emission may cause little change in the EA sulfate induced health impact over the downwind continents but might significantly influence the sulfate related climate change over the North Pacific and the western US.

We quantify the degree of linearity in the S-R relationship between EA SO_2 emissions and EA sulfate concentrations in the Northern Hemisphere by defining a linearity index. We find that EA sulfate concentrations respond nearly linearly (within 5%) to changes in EA SO_2 emissions everywhere except over the EA source region (where non-linearities exceed 10%, particularly at the surface during winter). Sulfate concentrations over EA increase proportionately less than the EA sulfur emissions because the conversion of SO_2 to sulfate reduces the availability of atmospheric oxidants (particularly H_2O_2), which in turn slows down the production rate for sulfate aerosol. However, even under low-oxidant conditions, conversion to sulfate occurs more quickly than transport to other continental regions.

We compare the direct effect (i.e., the change in sulfate produced from EA SO_2 emissions) and indirect effect (i.e., response of non-EA sulfate) of changing EA SO_2 emissions. We find that raising EA SO_2 emissions leads to a positive direct effect and a negative indirect effect on sulfate concentrations, particularly over the source region. However, the magnitude of the direct effect is more than 10 times larger than the indirect effect over the mid-latitude Pacific. We therefore conclude that the tagging and sensitivity techniques will produce nearly identical estimates of the source-receptor relationship of trans-Pacific transport of sulfate aerosols.

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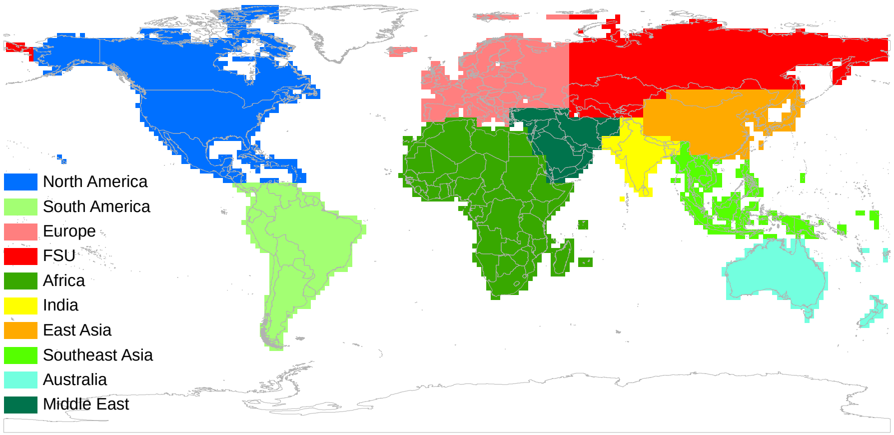


Fig. 1. The tagged ten continental source and receptor regions.

ACPD

8, 5537–5561, 2008

Title Page
Abstract
Introduction
Conclusions
References
Tables
Figures
Back
Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

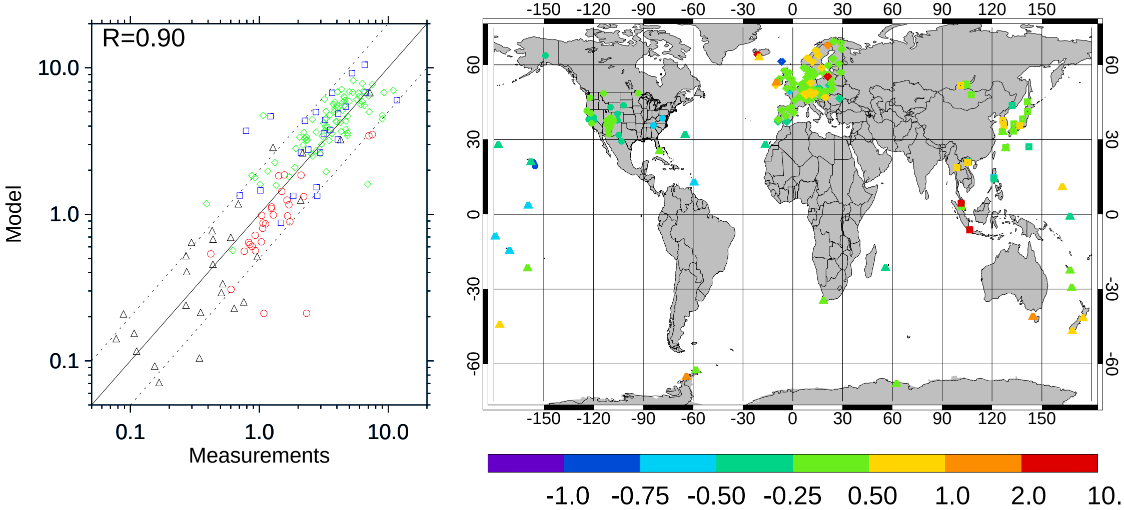


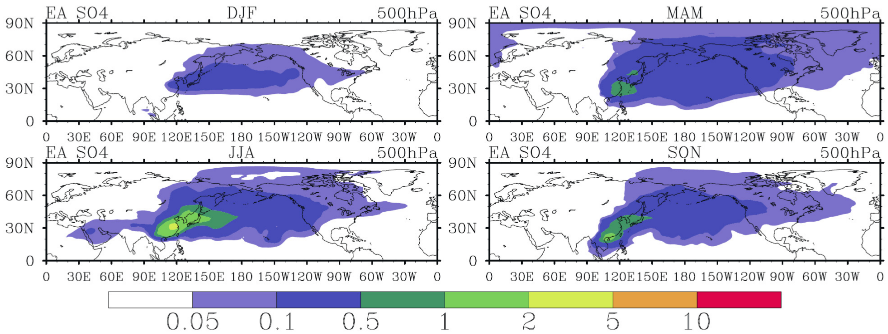
Fig. 2. Scatter plot (left panel) and the relative difference (i.e., (model-obs)/obs, right panel) between the simulated (MOZART2, early 1990s) and observed annual mean sulfate concentrations (SO_4^{2-}; unit: ug·m^{-3}). Observations are from RSMAS (1980s–1990s average, University of Miami, triangles), IMPROVE (1990–1992 average, circles), EMEP (1990–2002 average, diamonds), and EANET (2000–2004 average, squares).

ACPD

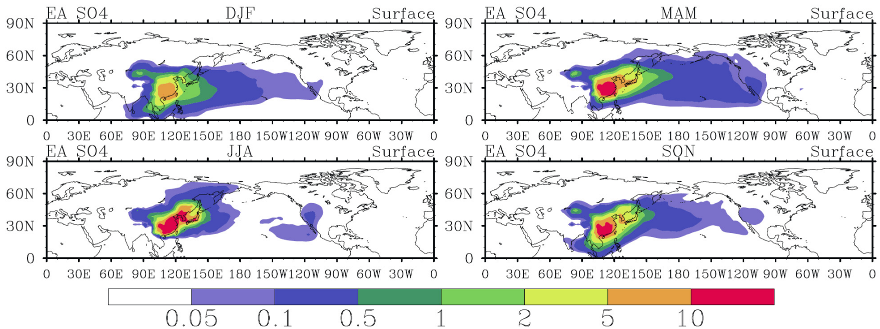
8, 5537–5561, 2008

Source-receptor relationships for East Asian sulfate

J. Liu et al.

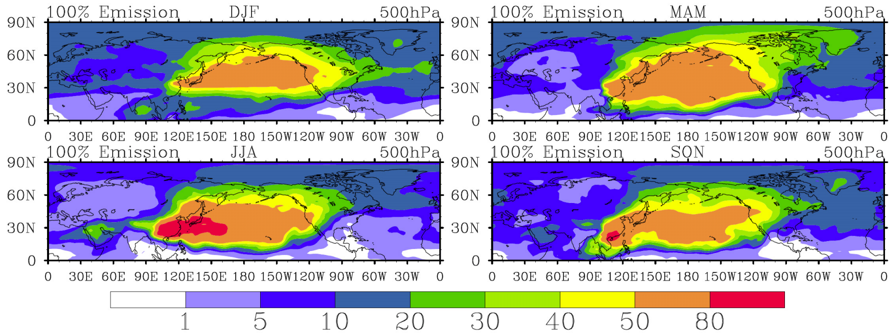


(a)

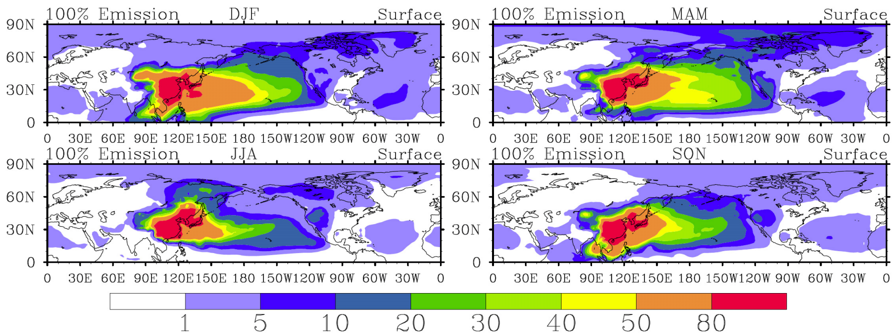


(b)

Fig. 3. Horizontal distribution of EA sulfate concentrations (SO_4; unit: $\mu g\,m^{-3}$) at **(a)** 500 hPa and **(b)** the surface in DJF, MAM, JJA, and SON (letters correspond to the 12 months of the year).

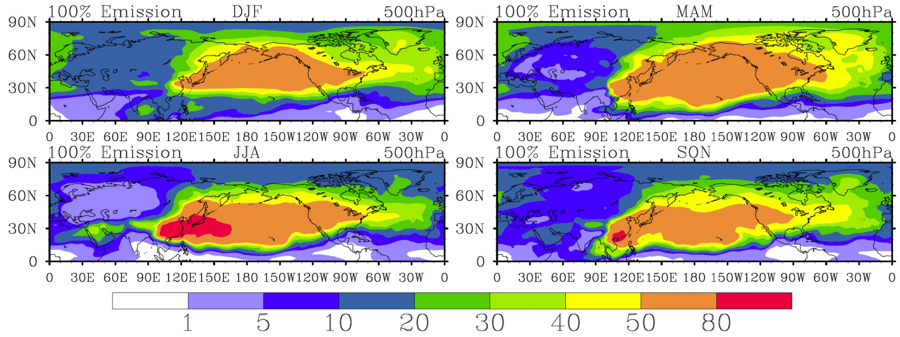


(a)

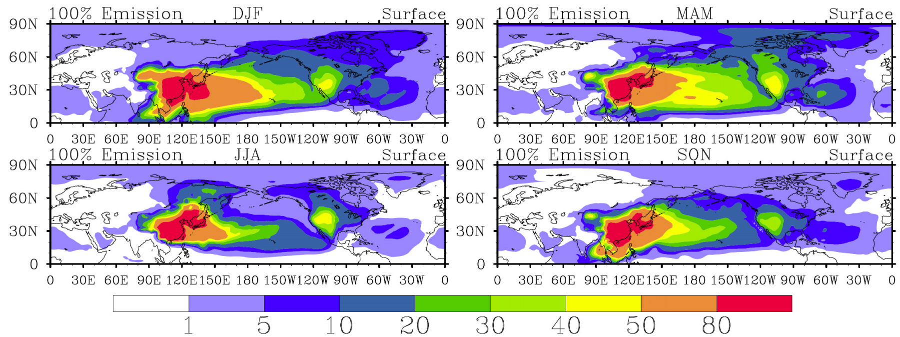


(b)

Fig. 4. Percent contribution of EA sulfate to total sulfate concentrations (with standard emissions) at **(a)** 500 hPa and **(b)** the surface in the same seasons as in Fig. 3.



(a)



(b)

Fig. 5. Same as Fig. 4, but percent contribution of EA sulfate to North American (NA) background sulfate (Note: NA background sulfate is the difference between total sulfate concentrations and the NA sulfate concentrations).

ACPD

8, 5537–5561, 2008

Source-receptor relationships for East Asian sulfate

J. Liu et al.

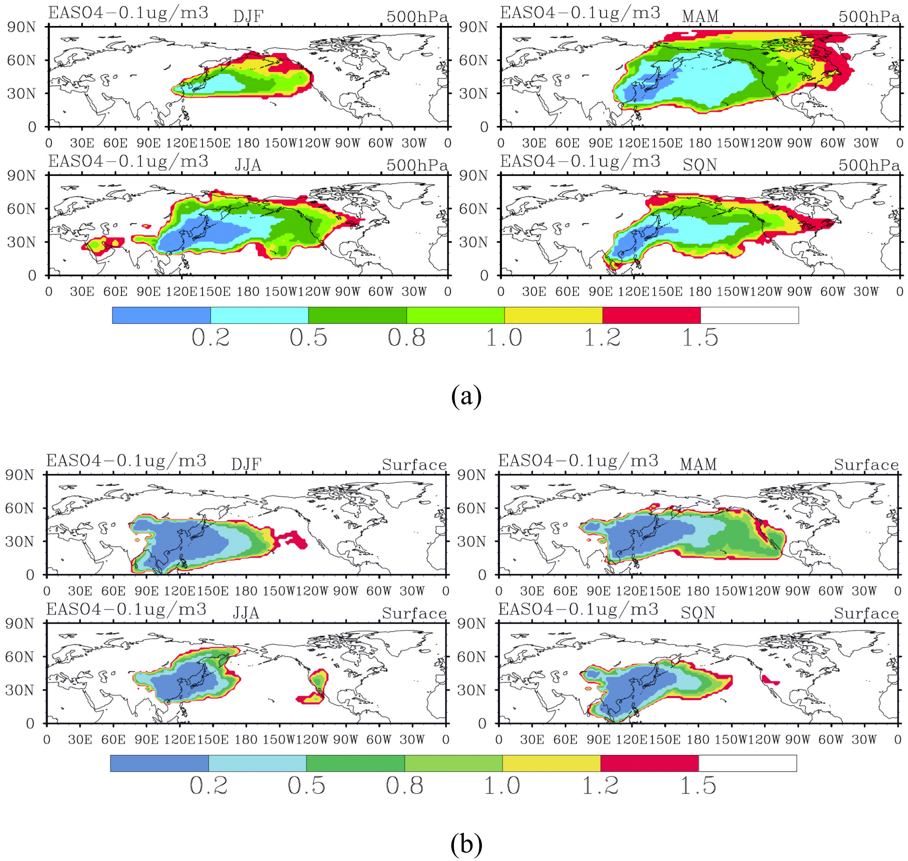


Fig. 6. Areas where at least 0.1 $\mu g\,m^{-3}$ of sulfate is contributed from EA ($A_{EA0.1}$) at **(a)** 500 hPa, **(b)** the surface over the same seasons as in Fig. 3 when EA sulfur emissions are 0.2, 0.5, 0.8, 1.0, 1.2 and 1.5 times the standard EA emissions (indicated by colors).

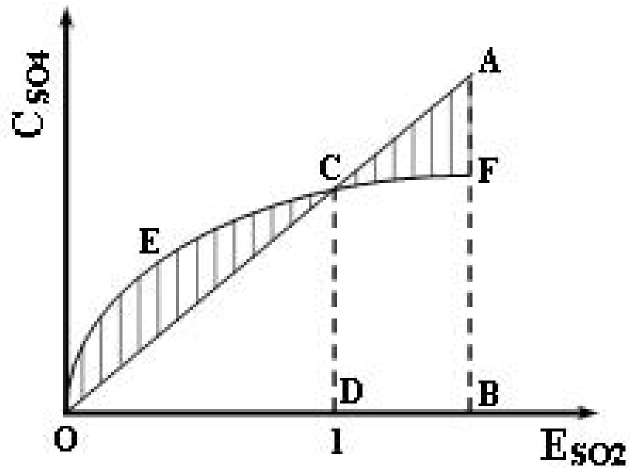
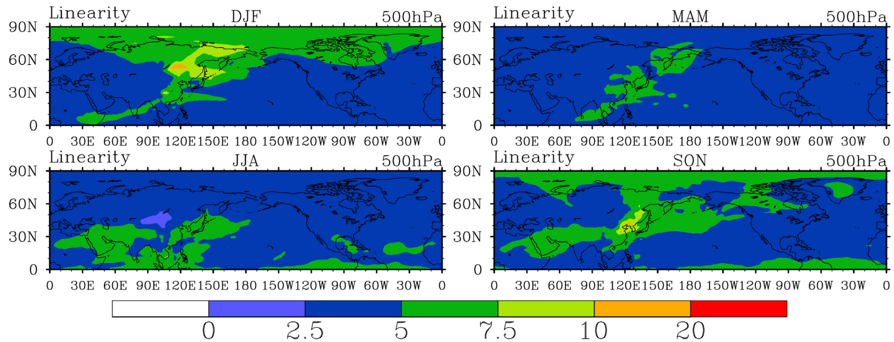
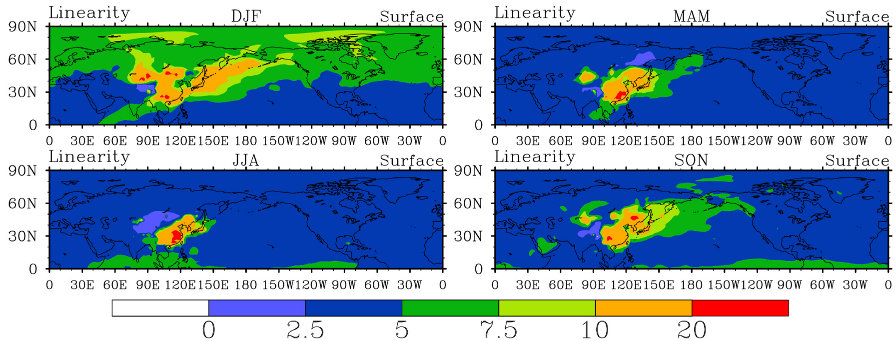


Fig. 7. Sketch of linearity of S-R relationships between SO_2 emissions (E_{SO_2}) and sulfate concentrations (C_{SO_4}). The straight line OA here shows the perfect linear dependence of C_{SO_4} on E_{SO_2}, when E_{SO_2} is varied between O and *B*. While the convex curve OECF shows the actual dependence of C_{SO_4} on E_{SO_2} (see Eq. (1) in Sect. 4 for the definition of linearity index. Note: "1" here indicates standard emissions, and "O" indicates the origin point).



(a)



(b)

Fig. 8. Distribution of the Linearity Index for the S-R relationship between EA SO_2 emissions and EA sulfate concentrations over the Northern Hemisphere (based on Eq. (1) where low numbers indicate approximate linearity; EA sulfur emissions range from 0 to 1.5 times the standard emissions) at **(a)** 500 hPa and **(b)** the surface. Seasons are the same as in Fig. 3.

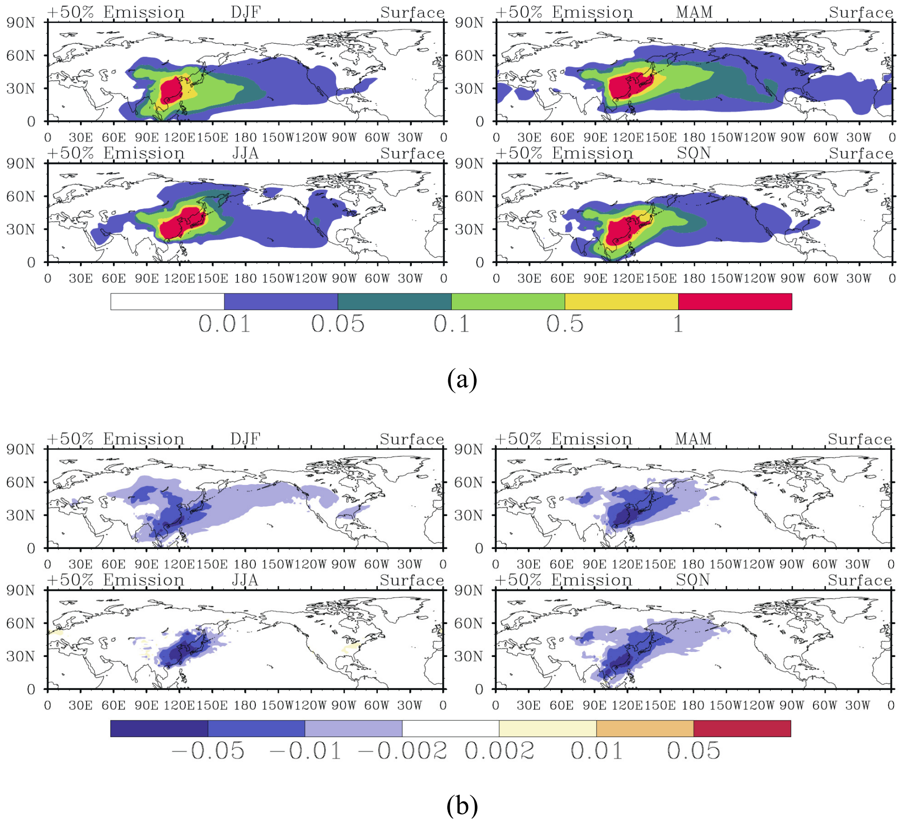


Fig. 9. Direct **(a)** and indirect **(b)** effects on surface sulfate concentrations (unit: $\mu g\, m^{-3}$) from 50% increase of EA SO_2 emissions (note: different color scales). Direct effects refer to changes in sulfate produced from EA emissions, while indirect effects refer to changes in sulfate resulting from emissions in other regions.

ACPD

8, 5537–5561, 2008

Source-receptor relationships for East Asian sulfate

J. Liu et al.