

# **Evaluating the potential influence of inter-continental transport of sulfate aerosols on air quality**

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## Abstract

In this study, we compare the potential influence of inter-continental transport of sulfate aerosols on the air quality of continental regions. We use a global chemical transport model, Model of Ozone and Related Tracers, version 2 (MOZART-2), to quantify the source-receptor relationships of inter-continental transport of sulfate aerosols among ten regions in 2000. In order to compare the importance of foreign emissions relative to domestic emissions and estimate the effect of future changes in emissions on human exposure, we define an “influence potential” (IP). The IP quantifies the human exposure that occurs in a receptor region as a result of a unit of SO<sub>2</sub> emissions from a source region. We find that due to the non-linear nature of sulfate production, regions with low SO<sub>2</sub> emissions usually have large domestic IP, and *vice versa*. An exception is East Asia (EA), which has both high SO<sub>2</sub> emissions and relatively large domestic IP, mostly caused by the spatial coincidence of emissions and population. We find that intercontinental IPs are usually less than domestic IPs by 1-3 orders of magnitude. SO<sub>2</sub> emissions from the Middle East (ME) and Europe (EU) have the largest potential to influence populations in surrounding regions. By comparing the IP ratios (IPR) between foreign and domestic SO<sub>2</sub> emissions, we find that the IPR values range from 0.00001 to 0.16 and change with season. Therefore, if reducing human exposure to sulfate aerosols is the objective, all regions should first focus on reducing domestic SO<sub>2</sub> emissions. In addition, we find that relatively high IPR values exist among the EU, ME, the former Soviet Union (FSU) and African (AF) regions. Therefore, based on the IP and IPR values, we conclude that a regional agreement among EA countries, and an inter-regional

agreement among EU, ME, FSU and (north) AF regions to control sulfur emissions would benefit public health in these regions.

## 1. Introduction

Sulfur containing pollutants (i.e., SO<sub>2</sub>, sulfate, and acid rain) and their adverse impacts on human health and ecosystems have been of increasing concern over the past few decades [Alcamo, *et al.*, 1995; Akimoto, 2003; Pope, *et al.*, 2004]. As more and more nations industrialize, sulfur pollutants, once considered to be local pollutants, are gradually being recognized to have regional and even hemispheric influence [Andreae, *et al.*, 1988; Husar, *et al.*, 2001; Park, *et al.*, 2004; Bergin, *et al.*, 2005]. SO<sub>2</sub>, the form in which most anthropogenic sulfur is released, is a precursor of sulfate aerosol. Sulfate aerosol is an important component of PM<sub>2.5</sub> (particulate matter, 2.5µm diameter or smaller). Both short-term and long-term exposure to PM<sub>2.5</sub> are associated with elevated human mortality rates [Pope, *et al.*, 2002; Bell, *et al.*, 2004]. In order to improve local air quality, many industrialized nations have not only implemented stringent domestic air pollution control strategies, but have also signed bilateral or multilateral agreements to abate emissions cooperatively [Grennfelt and Hov, 2005]. One of the most successful multilateral treaties to limit trans-boundary transport of air pollution is the Convention on Long-Range Trans-boundary Air Pollution (LRTAP) established in 1979. The LRTAP convention has been ratified by nearly 50 parties including most European countries, the Russian Federation, Canada, and the United States. Eight protocols, focusing on cooperative reduction of the emission of SO<sub>2</sub> and other air pollutants (e.g. nitrogen oxides, volatile organic compounds, persistent organic pollutants, etc.), have each been ratified

by more than 20 countries since 1979. As a result, SO<sub>2</sub> emissions from European countries have been reduced by 70% since 1980 [*Grennfelt and Hov, 2005*].

Recently, international concern about trans-boundary transport of air pollutants has extended beyond Europe to other regions. Due to a rapid increase in energy use (particularly the use of coal), ambient air pollution levels in many developing nations have increased dramatically. For example, in some Chinese cities, the PM<sub>2.5</sub> concentrations are 2 to 10 times higher than the U.S. standard of 35 µg/m<sup>3</sup> over a 24-hour period [*He, et al., 2002*]. SO<sub>2</sub> emissions from China and India increased by approximately 60% and 150%, respectively, between 1980-2000 [*Carmichael, et al., 2002*] and are expected to continue to increase [*Klimont, et al., 2001; Carmichael, et al., 2002*]. As a consequence, trans-Pacific transport of Asian emissions is of increasing concern to downwind countries including Japan and the United States [*Nakada and Ueta, 2004; Park, et al., 2004*].

Since Europe, East Asia and the United States (i.e., the three regions emitting the largest quantities of SO<sub>2</sub> and together contributing more than 50% of global anthropogenic SO<sub>2</sub> emissions) are all located in the northern mid-latitudes, air pollutants emitted from any of these regions may be transported rapidly in the mid-latitude westerlies and influence downwind regions [*Fiore, et al., 2002; Stohl, et al., 2002; Liu, et al., 2005*]. Given the remarkable achievements of the LRTAP protocols and potential effects of trans-boundary transport, the development of new environmental regimes to regulate inter-continental transport of air pollution is of interest to policy makers. Discussions of the possibility of expanding or duplicating the LRTAP regime in other regions or of creating new global or hemispheric regimes to regulate intercontinental

transport are occurring [Wettestad, 1997; Holloway, *et al.*, 2003; 2004; Keating, *et al.*, 2004; Selin, 2004; Brachtel, 2005].

The primary purpose of this study is to establish source-receptor relationships for the inter-continental transport of sulfate aerosols, and use them to explore the potential for inter-regional cooperation to jointly mitigate SO<sub>2</sub> emissions. We believe this is an important first step in determining which regions would benefit most from a multilateral environmental regime to address intercontinental transport. We focus on SO<sub>2</sub> emissions rather than other air pollutants because: 1) SO<sub>2</sub> is a precursor of sulfate aerosol, an important component of PM<sub>2.5</sub> which is harmful to human health [Pope, *et al.*, 2002]; 2) sulfate aerosols may be transported across regions and even continents [Park, *et al.*, 2004]; and 3) the technical and political mechanisms for controlling anthropogenic SO<sub>2</sub> emissions have been successfully implemented in many individual industrialized nations as well as under LRTAP and are potentially transferable to developing countries.

## **2. Methodology**

### **2.1 Influence potential and influence potential ratio**

Protecting human health and welfare are key motivations a nation has for mitigating domestic emissions of fine aerosols and their precursors. Human exposure to domestic and inter-continental transport of fine aerosols is influenced by a series of factors, including magnitude and location of emissions, speed of chemical transformation, physical transport and removal, distance and prevailing wind direction between regions, coincidence of population centers with elevated concentrations, etc. When mitigation of domestic emissions alone is insufficient or too costly to meet environmental goals,

countries could seek to obtain further reductions in ambient concentrations through international cooperation to reduce foreign emissions. In order to compare the effect of sulfate aerosol transport between regions on human exposure (i.e. eliminating the influence from varying emission magnitudes), we define an influence potential (IP). The IP is the population-weighted concentration over a receptor region resulting from a unit change of emissions from a source region (Equation 1):

$$IP(S, R) = \frac{\partial C_{PW}(S, R)}{\partial E(S)} \cong \frac{\Delta C_{PW}(S, R)}{E(S)} = AIP(S, R) \quad (1)$$

The  $IP(S, R)$ , represents the source-receptor (S-R) relationship for the transport of a specific pollutant from a source region  $S$  to a receptor region  $R$ ; it indicates the average exposure of an individual in  $R$  to the pollutant transported from  $S$ .  $C_{PW}(S, R)$  is the population-weighted concentration in  $R$  resulting from emissions in  $S$ .  $E(S)$  is the total annual emission from region  $S$ .  $AIP(S, R)$  represents the average influence potential, namely the emission-normalized population-weighted pollutant concentration in  $R$  resulting from emissions transported from  $S$ .

In order to derive the relationship between the IP and emissions of a particular species, a series of sensitivity studies which examine the relationship between emissions from source regions and concentrations in receptor regions are needed. These simulations are computationally expensive. However, when concentrations are linearly related to emissions, the IP is equal to the emission-normalized population-weighted concentrations, or the average influence potential (AIP), expressed in units of  $\mu\text{g} \cdot \text{m}^{-3}/(\mu\text{g} \cdot \text{yr}^{-1})$ . For sulfate aerosols, the S-R relationship is non-linear over source regions but close to linear following inter-continental transport [*Liu, et al., 2007b*]. Therefore,

the AIP is equivalent to the IP except near the emission source (where  $AIP > IP$ ). The relative importance of emissions from different foreign regions on a single receptor region can be evaluated by comparing the magnitudes of foreign IP or AIP values. These IP or AIP values also permit the policy-relevant analysis of how human exposure on downwind continents may change as a result of changes in emissions from any particular upwind continental region. The IP and AIP values thus allow the impact of emission changes to be estimated.

In order to compare the importance of foreign emissions relative to domestic emissions, we derive influence potential ratios ( $IPR = AIP_F / AIP_D$ ) of foreign ( $AIP_F$ ) to domestic ( $AIP_D$ ) average influence potentials for a receptor region. The IPR compares the average health damages caused by a unit of foreign emissions to a unit of domestic emissions. When the IPR is large, the influence of foreign emissions on the domestic population is large relative to the influence of domestic emissions. In this case, assisting in the mitigation of foreign emissions could be a viable policy option to improve domestic health and welfare. Conversely, when the IPR is small, the influence of one region on the other is relatively weak, and policies which control domestic emissions would be relatively more effective.

## **2.2 Model configuration**

We use the global chemical transport model, MOZART-2 (Model of Ozone and Related Chemical Tracers, version 2), to simulate the physical transport and chemical evolution of the sulfur pollutants used to calculate the AIP. Meteorological inputs are from the NCEP/NCAR reanalysis data at a horizontal resolution of 1.9 latitude x 1.9

longitude with 28 hybrid vertical levels from the surface to 2.7hPa. Anthropogenic emissions for 2000 are from *Dentener, et al* [2005] and *Stevenson, et al.* [2006]. Biomass burning emissions are from *van der Werf, et al.* [2003; 2004]. Detailed model descriptions and evaluations of gas-phase and aerosol species are given by [*Tie, et al.*, 2001; *Horowitz, et al.*, 2003; *Tie, et al.*, 2005; *Liu, et al.*, 2007a]. Using emissions representative of 2000, we conduct an 8-year simulation using meteorological fields from 1996-2003 in order to capture meteorological variability around the year 2000. Results for 1996 are used for model initialization and are discarded.

To develop source-receptor relationships, we tag and track the sulfur species from 10 continental regions (i.e., North America (NA), South America (SA), Europe (EU), the former Soviet Union (FSU, excluding part of Russia in the European domain), Africa (AF), the Indian subcontinent (IN), East Asia (EA), Southeast Asia (SE), Australia (AU), and the Middle East (ME)) as shown in Figure 1. Annual total anthropogenic SO<sub>2</sub> emissions from each source region are shown in Figure 2. In addition, we define 10 receptor regions which are identical to the 10 source regions. Evaluation of the tagged sulfur species concentrations and the associated linearity of sulfate production are given by *Liu, et al.* [2007a; 2007b]. We calculate population-weighted concentrations of sulfate in the surface layer above each receptor region using population data from the Gridded Population of the World in 2000 [*CIESIN*, 2000].

## **3 Results**

### **3.1 Average Influence potential (AIP)**

Figure 3 and 4 show the AIP resulting from domestic and inter-continental transport of sulfate aerosols. These IP values are based on the average surface sulfate concentrations from the 1997-2003 MOZART-2 simulation (i.e., using year 2000 emissions and meteorology from 1997-2003). Comparing Figure 3 to 2, the domestic AIP values are relatively high in SA, IN, SE, and AU where local SO<sub>2</sub> emissions are small, and are relatively low in NA and EU where local SO<sub>2</sub> emissions are large. This is consistent with the finding that increases in sulfate concentrations over source regions are proportionally less than the increase in SO<sub>2</sub> emissions due to a lack of oxidants [Liu, *et al.*, 2007b]. Although SO<sub>2</sub> emissions from the ME and IN are similar, the domestic AIP of sulfate in the ME is smaller than that of IN because the low liquid water content in the ME depresses the heterogeneous production of sulfate. The relatively low domestic AIP in the FSU and AF is largely due to the low overlap between surface sulfate concentrations and population. In contrast, EA has both larger SO<sub>2</sub> emissions and a relatively higher domestic AIP than NA or EU because of the coincidence of emissions and high population centers in EA.

Foreign AIPs from transported sulfate are smaller than domestic AIPs by 1-3 orders of magnitude due to the long distance foreign emissions must travel to effect populations in receptor regions. As shown in Figure 4, sulfate aerosols from the ME have a relatively high AIP in EU, FSU, AF, and IN. This is primarily due to the ME being close to these regions, but is also due to the prevailing high pressure system and lack of precipitation in the ME which makes pollution export to the surrounding regions efficient. In addition, sulfate aerosols from EU and the FSU have high AIPs in downwind regions. In contrast, aerosols from EA are mostly transported over the North Pacific, so the AIP of

EA sulfate aerosol is very small in most regions except SE. Similarly, aerosols from NA, SA and AU generally have very small inter-continental AIPs due to the long distance to other continents.

### **3.2 Influence potential ratio (IPR)**

Based on the domestic and intercontinental AIP values, we calculate influence potential ratios (IPR) between foreign and domestic SO<sub>2</sub> emissions in each receptor region. Figure 5 uses IPR values to illustrate the influence patterns of inter-continental transport of sulfate aerosols. Of these ten regions, the EU, FSU, ME and AF regions have higher IPRs than other regions due to both proximity and prevailing wind directions. Therefore, joint implementation of SO<sub>2</sub> controls among these four regions would have larger inter-continental health benefits than that of other regions. As shown in Figure 5, the ME shares strong influence relationships with surrounding regions, consistent with the findings in section 3.1. As a receptor, the ME is influenced by SO<sub>2</sub> emissions from EU while the SO<sub>2</sub> emissions from the ME have a relatively high influence on the FSU and AF. Among the other six regions, the IPRs for the transport of FSU sulfate to EA and ME sulfate to IN are relatively large. Due to long distances, AU, NA and SA usually have low IPRs for inter-continental transport of sulfate.

An important question is whether these inter-continental influence patterns persist throughout the year or vary with season. Figure 6 compares the inter-continental IPR in DJF, MAM, JJA and SON (namely, winter, spring, summer, and fall in the northern hemisphere). The inter-regional IPR values among EU, FSU, ME and AF are large throughout the year. However, the direction of the influence changes with season. IPRs indicate the relative influence of ME sulfate on the EU and FSU is largest in DJF, but

smallest in JJA due to seasonally alternating wind direction associated with the alternation of high and low pressure systems over Europe [Duncan and Bey, 2004]. Among other regions, the IPRs indicate that transport of ME sulfate to IN and FSU sulfate to EA are largest in DJF and MAM. In addition, the trans-Pacific transport of EA sulfate to NA is strongest in DJF and MAM and therefore has the largest IPR in these two seasons, similar to the findings in Liu, *et al.* [2005] and others. However, compared with IPR values between other regions, the effect of EA sulfate on NA is very small.

### **3.3 Uncertainties**

We recognize that uncertainties exist in all components of our study: emissions, physical and chemical transport and exposure estimates. A detailed discussion of these uncertainties is given in Liu, *et al.* [2007a]. Generally, the simulated sulfate concentrations are within a factor of 2 of the global surface observations [Ginoux, *et al.*, 2006; Liu, *et al.*, 2007a]. In addition, we use an AIP to represent the actual IP. This will underestimate intercontinental IPR values, particularly for receptors with large SO<sub>2</sub> emissions (e.g., EU, NA and EA), because AIP values are usually larger than IP values for domestic emissions. Despite these limitations, we believe this analysis provides a clear indication of which regions have the largest influence on which others. These relationships should be of use to policymakers when determining in which regions mitigation of sulfur dioxide emissions will be most effective in reducing domestic human exposure to sulfate aerosols.

## **4 Policy implications**

IPs and IPRs between regions provide a valuable tool for policy makers evaluating the potential effectiveness of emission mitigation efforts in foreign countries. An influence potential indicates the potential exposure reduction that would result from a unit decrease in domestic or foreign emissions. As Figures 3-6 imply, abatement of domestic SO<sub>2</sub> emissions results in larger reductions in exposure to sulfate aerosols in source regions than abatement of foreign emissions. Therefore, if reducing human exposure to sulfate aerosols is the objective, all regions should first focus on reducing domestic SO<sub>2</sub> emissions. The advantage of domestic emission reductions persists until the ratio of marginal costs for emission abatement exceeds the ratio of influence potentials between two regions. At that point, regional or hemispheric agreements on international transport of sulfate aerosols become increasingly attractive and potentially beneficial. Figures 5-6 show relatively high IPR values exist among EU, FSU, ME, and (north) AF. Thus, health benefits among these four regions that are sufficient to warrant an examination of the feasibility of inter-regional agreements on sulfur emission reductions are projected to occur if any of these four regions reduce their emissions. Although other aerosol species also have negative effects on human health, agreements to cooperatively reduce emissions of SO<sub>2</sub> may be particularly attractive due to the variation in abatement costs between developed and developing countries as well as the fact that the technical and political mechanisms for controlling anthropogenic sulfur emissions have been successfully implemented in many industrialized nations.

Although inter-continental AIPs and IPRs for other regions are small, the rapid industrialization in the developing countries of South and East Asia is significantly increasing energy demand. This demand is largely met through the combustion of coal

which results in large and increasing anthropogenic SO<sub>2</sub> emissions. Due to both large population size and high domestic IPs, each unit increase in SO<sub>2</sub> emissions within these regions would cause a larger regional health impact than a unit increase in emissions from other regions. Given the relatively large intra-region IP but small inter-continental IP, regional agreements on sulfur abatement and clean development in South and East Asian countries would be particularly attractive and could significantly benefit public health in these regions.

## **5 Conclusions**

In this paper we develop a methodology which couples a global atmospheric model with demographic information. Our purpose is to analyze the influence of emissions from one continental region on another and the benefits in reduced human exposure to sulfate aerosols of cooperative reductions in SO<sub>2</sub> emissions. Using the global chemical transport model MOZART-2, we conduct a simulation of inter-continental transport of sulfate aerosols by tagging regional sulfur emissions over ten continental regions. We define two indicators, namely an average influence potential (AIP, the emission-normalized population-weighted air pollution concentrations transported from a source region to a receptor region) and an influence potential ratio (IPR, the ratio of AIP values of foreign and domestic emissions on a domestic population), which we use to evaluate the potential for bilateral and multilateral cooperation between nations.

Based on the calculated AIP values between each pair of source-receptor regions, we find that over each source region, regions with low SO<sub>2</sub> emissions (such as SA, IN, SE and AU) usually have high domestic AIPs. In contrast, regions with large SO<sub>2</sub> emissions (e.g., NA and EU) usually have relatively low AIPs. This is due to the non-

linear relationship between SO<sub>2</sub> emissions and sulfate production (i.e., the increase in sulfate concentrations over the source region is proportionally less than the increase in SO<sub>2</sub> emissions) [Liu, *et al.*, 2007b]. However, although the total SO<sub>2</sub> emissions in EA are larger than those of EU or NA, EA has a larger AIP than either EU or NA. This is due to the coincidence of SO<sub>2</sub> emissions and large population centers in EA.

In order to compare the importance of foreign emissions to domestic emissions, we calculate the IPR between foreign and domestic emissions. We find that the mean IPR values range from approximately 0.16 to 0.00001. This indicates that if reducing human exposure to sulfate aerosols is the objective, all regions should first focus on reducing domestic SO<sub>2</sub> emissions. The advantage of domestic emission reductions persist until the ratio of marginal costs for emission abatement exceeds the ratio of influence potentials between two regions. Due to both proximity and prevailing winds, the EU, FSU, ME and AF regions are associated with the largest inter-regional IPR. In addition, this high influence pattern is robust throughout the year although the influence directions change by season

Therefore, based on these AIP and IPR relationships, we find that intra-regional agreements among South and East Asian countries, and an inter-regional agreement among EU, ME, FSU, and (north) AF regions to control SO<sub>2</sub> emissions would benefit public health in these regions.

Further research that investigates the marginal abatement costs (MAC) for SO<sub>2</sub> emissions in different countries and evaluates the health impacts due to sulfate exposure would permit a cost-benefit analysis of various cooperative mitigation strategies. Such an analysis would permit a comparison of the MAC and IPR values between countries.

This would allow the evaluation of the economic motivation of industrialized and developing countries to jointly mitigate SO<sub>2</sub> emissions to protect public health.

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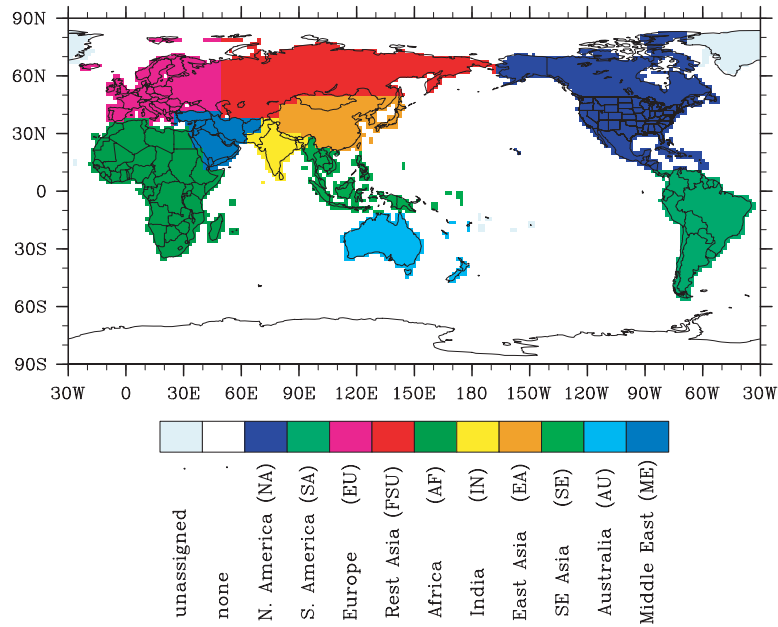
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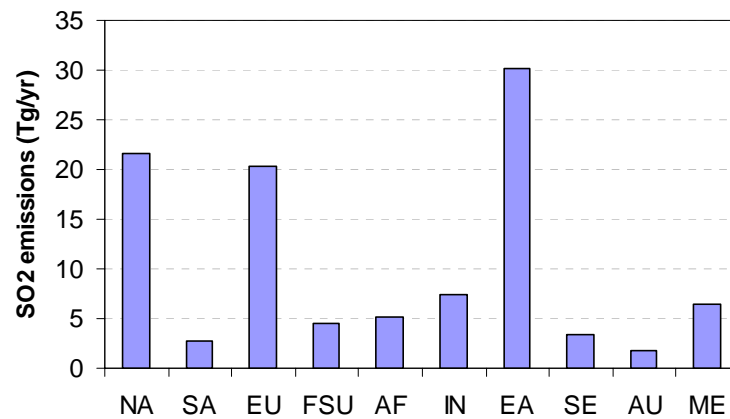
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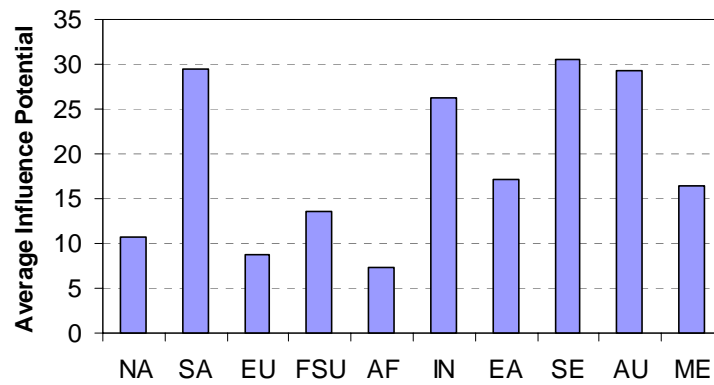
## Figures



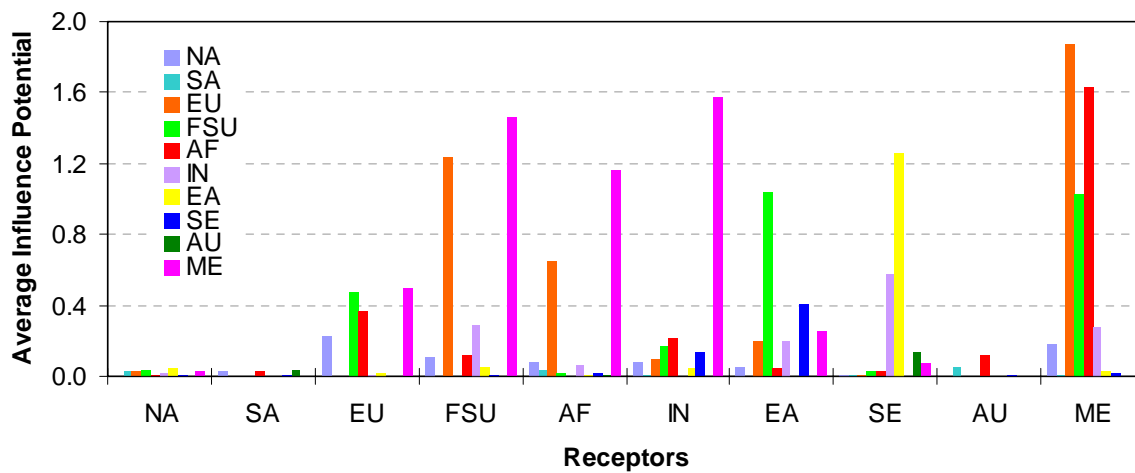
**Figure 1** The ten continental regions tagged in our MOZART-2 simulations.



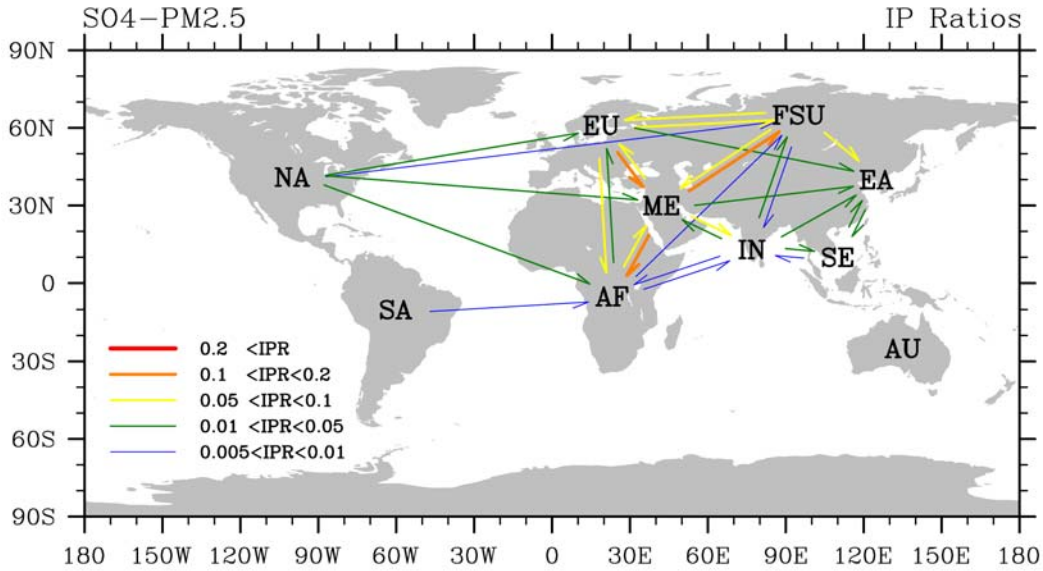
**Figure 2** Annual anthropogenic SO<sub>2</sub> emissions (Tg/year) in 2000 from ten continental regions (based on the RAINS CLE-2000 emission inventory).



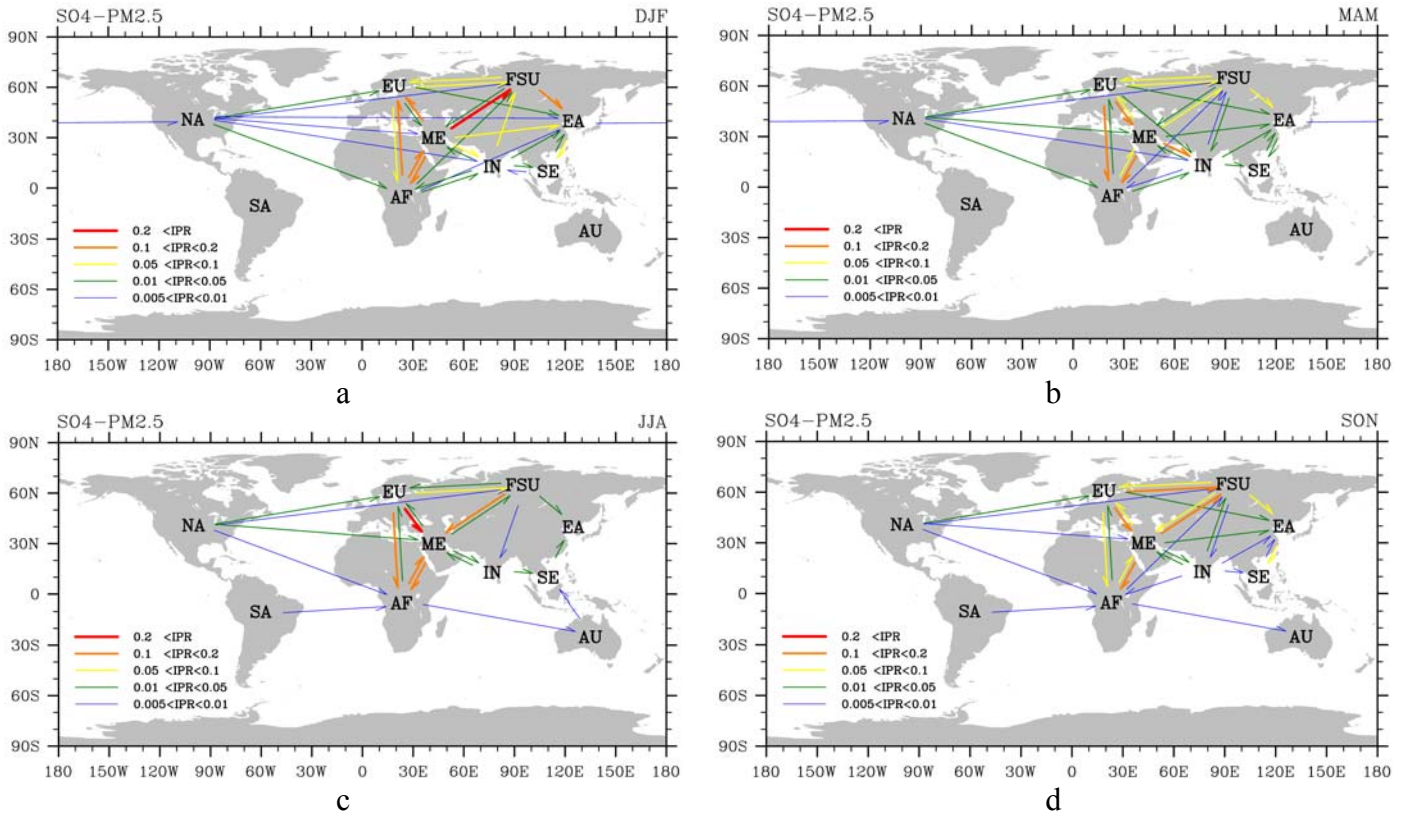
**Figure 3** Average influence potential of sulfate (PM<sub>2.5</sub>) derived from domestic SO<sub>2</sub> emissions over each region. Unit:  $10^{-20} \cdot (\mu\text{g} \cdot \text{m}^{-3}) / (\mu\text{g} \cdot \text{yr}^{-1})$ .



**Figure 4** Same as figure 3, but for the average influence potential from inter-continental transport of tagged sulfate derived from emissions of SO<sub>2</sub> from ten source regions (represented by colors). (Please note the different scales in Figure 3 and Figure 4.)



**Figure 5** Influence potential ratios (IPR) of inter-continental transport of fine (PM<sub>2.5</sub>) sulfate aerosols. Arrows indicate the influence direction from a source to a receptor region. Colors indicate the magnitudes of IPR ranging from red (strong influence) to blue (weak influence). Arrows with IPR less than 0.005 are not shown.



**Figure 6** Same as figure 5, but seasonal IPR values for (a) DJF, (b) MAM, (c) JJA, and (d) SON.