An overview of emissions of SO_2 and NO_x and the long-range transport of oxidized sulfur and nitrogen pollutants in East Asia

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ABSTRACT

The long-range transport of oxidized sulfur (sulfur dioxide (SO_2) and sulfate) and oxidized nitrogen (nitrogen oxides (NO_x) and nitrate) in East Asia is an area of increasing scientific interest and political concern. This paper reviews various published papers, including ground- and satellite-based observations and numerical simulations. The aim is to assess the status of the anthropogenic emissions of SO_2 and NO_x and the long-range transport of oxidized S and N pollutants over source and downwind region. China has dominated the emissions of SO_2 and NO_x in East Asia and urgently needs to strengthen the control of their emissions, especially NO_x emissions. Oxidized S and N pollutants emitted from China are transported to Korea and Japan, due to persistent westerly winds, in winter and spring. However, the total contributions of China to S and N pollutants across Korea and Japan were not found to be dominant over longer time scales (*e.g.*, a year). The source–receptor relationships for oxidized S and N pollutants in East Asia varied widely among the different studies. This is because: (1) the nonlinear effects of atmospheric chemistry and deposition processes were not well considered, when calculating the source–receptor relationships; (2) different meteorological and emission data inputs and solution schemes for key physical and chemical processes were used; and (3) different temporal and spatial scales were employed. Therefore, simulations using the same input fields and similar model configurations would be of benefit, to further evaluate the source–receptor relationships of the oxidized S and N pollutants.

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Introduction

East Asia has experienced rapid growth of populations and economies in recent years. Extensive use of fossil fuel has caused a significant increase of emissions of sulfur dioxide (SO_2) and nitrogen oxides (NO_x) (Streets et al., 2003). As a result, sulfur (S) and nitrogen (N) pollutants have damaged public health and degraded regional air quality; examples of these impacts include acid deposition (Cogbill and Likens, 1974; Streets et al., 1999; Hao et al., 2000; Han et al., 2001; Tang

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et al., 2010), photochemical smog (National Research Council, 1991; Ma et al., 2002; Wang and Kwok, 2003), and heavy haze (Quinn and Bates, 2003; Wu et al., 2005; Che et al., 2007; Tie et al., 2009; Liu et al., 2013). Hence, increasing emissions of S and N pollutants in East Asia have become a focus of research.

Since the 1980s, anthropogenic emissions of SO_2 and NO_x have leveled off across North America and Europe, following the implementation of stricter legislation (NADP, 2000; Akimoto, 2003; Richter et al., 2005; Stern, 2006; Monks et al., 2009). In contrast, anthropogenic emissions have increased across East Asia, due to economic growth (Zhang et al., 2009). Cofala et al. (2012) reported that East Asia contributes about 36% SO_2 and 29% NO_x to global emissions, much more than those of the United States and Europe. Oxidized S (SO_2 and sulfate) and oxidized N (NO_x and nitrate) pollutants produced in East Asia can be transported not only to neighboring but also distant countries that are in downwind regions or continents, affecting air quality and climate forcing there. Many observations in the North Pacific have provided extensive evidence for the transport of oxidized S and N pollutants from East Asia (Hoell et al., 1996, 1997; Huebert et al., 2003; Jacob et al., 2003; Singh et al., 2009). Furthermore, oxidized S and N pollution from East Asia has long been observed in the western areas of Canada, the US, and beyond (Jaffe et al., 1999; Biscaye et al., 2000; van Curen and Cahill, 2002; Grousset et al., 2003; Stohl et al., 2007; van Donkelaar et al., 2008). As a result, the long-range transport of oxidized S and N pollutants is an area of increasing scientific interest and political concern (Arndt and Carmichael, 1995; Ichikawa and Fujita, 1995; Huang et al., 1995; Streets et al., 1999).

S and N are emitted into the atmosphere as SO_2 and NO_x and then are transformed into sulfate and nitrate. Oxidized S and N pollutants can be transported away from source areas in association with air flows or dust storms. In this review paper, we summarize the literature from oxidized S and N field observations (ground, aircraft, and satellite), and model simulations. The aim is to characterize the levels and trends of SO_2 and NO_x emissions and depict the features of long-range transport (including transport pattern, transport pathways, seasonal variability, and the quantification of trans-boundary transport), along with providing some suggestions for the mitigation of atmospheric oxidized S and N pollutants.

1. SO$_2$ and NOx emissions

1.1. Trends and amounts of SO$_2$ and NOx emissions

The emissions of SO_2 and NO_x from East Asia in the 1990s surpassed those from North America and Europe, and are expected to continue to exceed them for decades (Streets et al., 2000; Akimoto, 2003). China, the world's most populous country, has been undergoing persistent rapid industrialization and urbanization over the last two decades. As a result, SO_2 and NO_x emissions in China have dominated the changes within East Asia (including China, Japan, South Korea, North Korea, and Mongolia) in recent years. Until 2006, SO_2 emissions in China accounted for about 94.6% of the total emissions in East Asia, and NO_x emissions accounted for

about 81.7%. During the same period, SO_2 emissions in Japan and South Korea only accounted for about 2.7% and 1.2% of the total emissions in East Asia, and NO_x emissions in the two countries accounted for about 9.4% and 5.1%, respectively (Zhang et al., 2009). In the light of this situation, SO_2 and NO_x emissions in the past, present, and future are shown to help understand the regional atmospheric environment. Fig. 1 shows the temporal evolution of SO_2 and NO_x emissions between 1980 and 2020 in Asia. The data are from Foell et al. (1995); Arndt et al. (1997); Streets and Waldhoff (2000); Streets et al. (2000, 2001); Hao et al. (2002), the State Environmental Protection Administration, China (SEPA, 2003), Streets et al. (2003); Ohara et al. (2007), and Zhang et al. (2007, 2009).

Due to the Asian economic crisis, after 1995, emissions of SO_2 did decrease for a few years. In contrast, there has been an ongoing increase of NO_x emissions since 1980, with the highest growth having occurred after 2000 (Monks et al., 2009). The annual average growth of NO_x emissions between 1990 and 1997 was about 6.2%, considerably larger than the SO_2 growth rate, which was only 2.2%, per year (Streets et al., 2001).

Similarly, Chinese SO_2 emissions also experienced a highly significant increase between 1980 and 1995. The emissions of SO_2 have been controlled by the Chinese government since the middle of the 1990s (Hao et al., 2000), through a reduction in economic growth, the mining of higher quality coals, enhanced environmental awareness, and a reduction in industrial coal use; as a result, a slowdown in the growth of SO_2 emissions appeared, and a reduction actually occurred in 1997 (Streets et al., 2000). However, the decreasing trend could not last long, and SO_2 emissions started to increase again after 2000, due to the economic boom and growing infrastructure investments in China (Larssen et al., 2006; Ohara et al., 2007; Zhang et al., 2009; Tang et al., 2010). In preparation for the 2008 Olympic Games, SO_2 emissions over East China showed a remarkable decrease after 2007, because of strong controls on pollutant emissions (Zhang et al., 2012). In contrast to the pattern of SO_2 emissions, NO_x emissions in China have continued to grow rapidly since 1980. During 1994–2000, NO_x emissions in China increased by 13% (the transport and chemical evolution over the Pacific (TRACE-P) inventory; Streets et al., 2003), and the increasing trend accelerated further to 55% between 2001 and 2006 (the intercontinental chemical transport experiment-phase B (INTEX-B) inventory; Zhang et al., 2009). These NO_x trends during 1996–2004 were validated by tropospheric satellite observations, which demonstrated a highly significant increase of about 50% in annual growth rates over the industrial areas of China (Richter et al., 2005).

A few studies have estimated the future emissions of SO_2 and NO_x in China in 2020 (van Aardenne et al., 1999; Streets and Waldhoff, 2000; Ohara et al., 2007). The results from the regional air pollution information and simulation (RAINS-Asia) have suggested that SO_2 emissions will increase from 23.0 Tg in 1990 to 30.6 Tg in 2020 (33% growth rate), if considerable advancements in control technologies occur, or by 164% if current practices are not altered. Growth rates of NO_x emissions are expected to be even greater. Emissions of NO_x are projected to increase from 2.9 Tg N in 1990 to somewhere in the range of 9.3 Tg N by 2020 (180% growth rate), or more than 210% if there are no changes in environmental policies and control. Although these studies have indicated that NO_x emissions from China

have increased markedly and will continue to increase further, there are no agreements in place to limit such growth at present.

Although the overall trends of SO_2 and NO_x emissions are consistent among the inventories and studies from 1980 to 2020, the magnitudes of the SO_2 and NO_x emissions have varied, due to different estimations of energy consumption. The Chinese SO_2 and NO_x emissions derived from several emission inventories and studies during 1985–2010 together with the deviations based on the regional emission inventory in Asia (REAS) show that the emission estimates are significantly different between both emission inventories and years (Table 1). For SO_2 emissions, the RAINS-Asia values were higher than those of REAS, with the highest deviation of 27.3%. The SEPA values were lower than those of REAS from 1995 to 2003, and the deviation changed from −12.7% to −41.1%. The estimates of Streets et al. (2000) were lower than those of REAS by −3.4% to −10%. The values of TRACE-P and INTEX-B were more than 20% lower than those of REAS. NO_x emissions estimated by RAINS-Asia, Hao et al. (2002), and Streets et al. (2001) were higher than those of REAS, with the deviation ranging from 1.3% to 29.4%. The discrepancy in NO_x emissions between TRACE-P and REAS was −16.2%. The estimates of Zhang et al. (2007) for the period 1995–2003 were slightly lower than those of REAS, with the largest deviation being −8.2%.

1.2. Sector source fractions of SO_2 and NO_x emissions

The SO_2 and NO_x emissions from different sectors in China, in the past (1995), present (2006), and future (2020), show a change in the contribution of each sector over time (Fig. 2). Industry, power plants, and domestic sources were the highest contributors to SO_2 emissions, responsible for about 53%, 27%, and 17%, respectively, in 1995. Power plants (59%), instead of industrial sources (31%), became the highest contributors to SO_2 emissions by 2006. The contribution of power plants to SO_2 emissions is likely to be much smaller in 2020, due to emission control measures (Streets and Waldhoff, 2000). The major sources of NO_x emissions in all time periods were industry, power plants, and transportation. The contribution of power plants increased from 34% in 1995 to 44% in 2006, and that of

transportation doubled from 12% to 24%; this reflects the significant increase of some energy-consuming activities, for example, total thermal-based electricity generation and total vehicle numbers (Zhang et al., 2009). Similar to the SO_2 emissions, the contributions of the industry sector to NO_x emissions are expected to increase from 2006 to 2020.

1.3. Spatial distribution and seasonality in SO2 and NOx emissions

Ohara et al. (2007) and Zhang et al. (2009) presented the geographical distributions of SO_2 and NO_x concentrations in Asia. For countries in East Asia, the SO_2 emissions were mainly concentrated in populated and industrialized areas of China, i.e., the Northeastern Plain, the Beijing–Tianjin–Hebei region, the Yangtze River Delta, Pearl River Delta, and the Sichuan Basin. For NO_x emissions, besides the areas mentioned above, the southern areas of the Korean Peninsula and Japan were also important contributors. Regarding the total amounts over China, SO_2 and NO_x emissions show weak seasonal variations, with ratios of 1.4 and 1.3 between their maxima and minima values, because their main sources are industrial and transportation emissions, which only have small seasonal cycles (Zhang et al., 2009). However, seasonal variations can be characterized from region to region; high pollution levels in winter and low levels in summer have been found in Northwest China, whereas the inverse seasonal variation is found in South China; these differences are attributed to energy demands and meteorological conditions (Zhang et al., 2012).

2. Long-range transport of oxidized S and N pollutants

2.1. Trans-boundary transport of oxidized S and N from China to Japan and South Korea

2.1.1. Field experiments
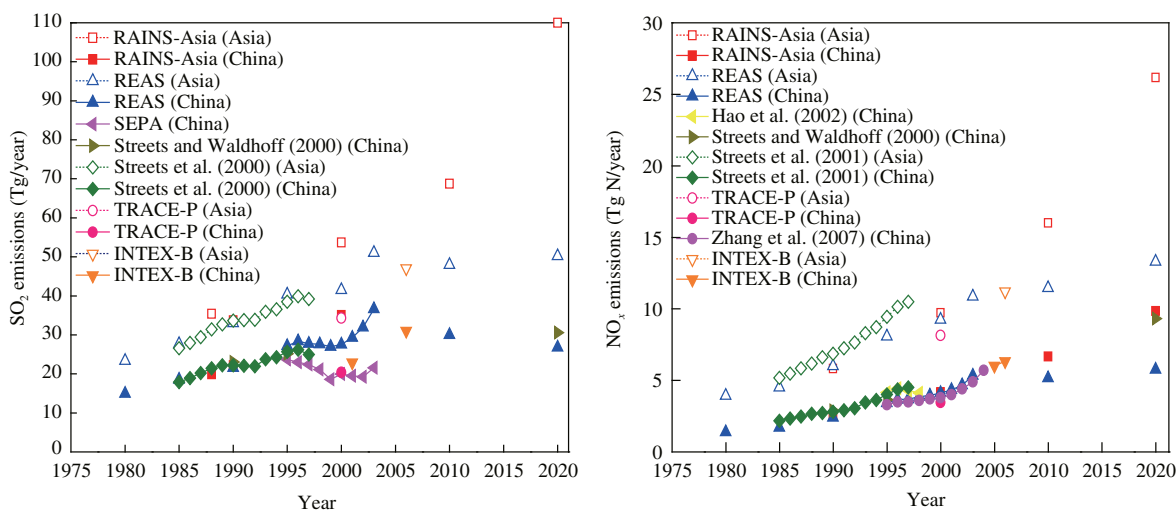Long-range transport of oxidized S and N pollutants from China to Japan and South Korea has been a central issue in

Fig. 1 – SO_2 and NO_x emissions in Asia between 1980 and 2020.

recent years. Under the influence of westerly flows, air pollutants can be transported eastwards across continental Asia to the Korean peninsula and Japan. Increases in the concentrations of anthropogenic oxidized S and N pollutants have often been observed in recent years (Takami et al., 2005; Hatakeyama et al., 2011; Kang et al., 2013). Analyses of SO_2 emissions and deposition in Japan have concluded that Japan receives more sulfur deposition than that which can be attributed to its own emissions (Fujita, 1996; Arndt et al., 1997), due to the long-range transport of pollutants from continental Asia. Chiwa et al. (2013) and Takashima et al. (2011) showed that Japan has been receiving increasing levels of atmospheric nitrogenous pollutants from eastern regions of continental Asia over the last few decades. By analyzing the acid deposition monitoring network in East Asia (EANET) data, Lu et al. (2010) showed that a 1% increase in SO_2 emissions in China corresponds to increases by approximately 0.71% and 1.15% in Japan and South Korea, respectively, of both SO_2 and SO_4^{2-} concentrations. Aikawa et al. (2010) found a clear longitudinal gradient in the measured SO_4^{2-} concentrations over the East Asian Pacific Rim region. The longitudinal gradient peaked in winter and was lower in summer; the winter peak was explained by the highly efficient transport of SO_4^{2-} from the Asian continent during this period.

2.1.2. Quantified transport

Chemical transport models have become critical tools in quantifying oxidized S and N trans-boundary transport. In the early to mid-1990s, long-range transport models started being applied in Asia (Arndt and Carmichael., 1995; Ichikawa and Fujita, 1995; Huang et al., 1995). Since this time, several regional transport models have been used in Asia (Table 2). In contrast to field observations, models can reflect chemical and physical processes during transport, provide more generality (compared to individual long-range transport case studies) based on long-term (monthly, seasonal, and annual) simulations, and present a large-scale (regional or global) view of the distribution of concentrations and deposition of pollutants. Thus, many researchers have used models to quantitatively evaluate the trans-boundary transport of oxidized S and N pollutants from source to receptor regions, by controlling the emissions from a source region and thereby clarifying the source–receptor relationships.

Some researchers have considered that the primary source of oxidized S and N deposition within each country in East Asia was the respective country's own anthropogenic emissions. For example, Huang et al. (1995) found that about 93.7% of the S deposited within Japan and 90.0% within South Korea were from internal sources, while China accounted for only 3.5% of Japan's total S deposition and 7.9% of South Korea's. Similarly, Ge et al. (2014) concluded that the emissions from China contribute 13% of S and 7% of N wet deposition in Japan. Holloway et al. (2002) considered that China contributed 18% of nitrate deposition in Japan and 26% in South Korea. Lin et al. (2008) calculated that long-range transport from eastern China contributed > 20% of anthropogenic reactive nitrogen as well as sulfur deposition in East Asia.

In contrast, other researchers have found that China is a major, and significant, source of oxidized S and N pollutants across the whole region. For example, Ichikawa and Fujita (1995) and Ichikawa et al. (1998) estimated that about 40% of the total sulfur deposition in Japan was from sources in continental Asia, and of these, China was a major source, accounting for one-half of the anthropogenic wet sulfate deposition in Japan. Arndt et al. (1998) found that China and South Korea play a major role in the sulfur deposition in southern and western Japan. Kajino et al. (2013) considered that the contribution of three regions in China that produce high levels of emissions accounted for approximately 50%–60% of the total deposition of nitrate in South Korea and Japan. Aikawa et al. (2010) suggested that the proportional contribution of Chinese SO_4^{2-} to the total concentrations in Japan throughout the year was approximately 50%–70% or 40%–60%, depending on different emissions of SO_2 in China.

In order to investigate the differences in estimates, of the contribution of oxidized S and N pollutants from China to the concentration or deposition of these pollutants in Japan and South Korea, the search periods, emission data, and model parameters of relevant studies have been compared (Table 3). The large disagreement in the various source-receptor estimates could be explained by three possible reasons. First, the quantitative source–receptor relationships considered by different studies were based on different periods and spatial

Table 1 – Comparison of the difference between the emissions of SO_2 and NO_x, calculated by different emission inventories and studies in China.

	Study	1985	1990	1995	1996	1997	1998	1999	2000	2001	2002	2003	2010
SO_2	RAINS-Asia		2.9%						27.3%				
	SEPA			−12.7%	−19.0%	−19.3%	−23.7%	−31.2%	−27.6%	−33.5%	−39.7%	−41.1%	
	Streets et al. (2000)	−3.4%	3.1%	−5.3%	−7.7%	−10.0%							
	TRACE-P								−26.0%				
	INTEX-B									−21.8%			
NO_x	RAINS-Asia		4.3%						1.3%				29.4%
	Hao et al. (2002)			21.4%	21.0%	16.6%	6.9%						
	Streets et al. (2001)	28.0%	16.5%	17.4%	19.6%	22.3%							
	TRACE-P								−16.2%				
	Zhang et al. (2007)			−3.7%	−4.5%	−5.0%	−6.6%	−6.5%	−7.8%	−7.7%	−5.9%	−8.2%	

All the data show the deviation based on REAS, calculated by the formula: $\frac{Emission-REAS}{REAS} \times 100\%$.
RAINS-Asia: the regional air pollution information and simulation; TRACE-P: the transport and chemical evolution over the Pacific; INTEX-B: the intercontinental chemical transport experiment-phase B; SEPA: State Environmental Protection Administration; REAS: the regional emission inventory in Asia.
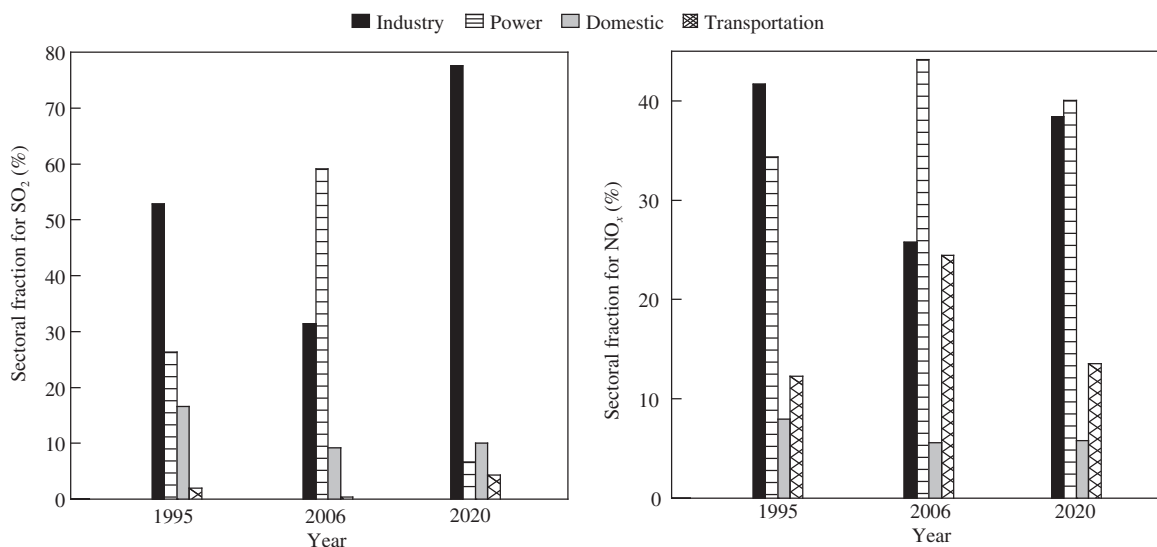
Fig. 2 – Sources of SO$_2$ and NO$_x$ emissions by sector in China, in 1995, 2006, and 2020. Data sources are from Streets and Waldhoff (2000) and Zhang et al. (2009).

scales. Compared with short-term simulations (a month, Huang et al., 1995), long-term simulations (at least one year covering four seasons) can contain seasonal meteorological variations, which are significant for the estimates of source–receptor relationships. Secondly, the estimates vary because of the highly nonlinear relationship between both the atmospheric chemistry and deposition processes as well as changes in the emissions from each source region, when employing methods such as the zero-out or brute-force methods to derive the source–receptor relationships. The third reason for the disagreement in source-receptor estimates between studies is that there are differences between the input meteorological and emission data and solution schemes for key physical and chemical processes used in the models. These differences are especially dominant in meteorological fields, emissions, chemical conversion rates, and removal rates (Carmichael et al., 2002; Wang et al., 2008; Park et al., 2014). The variation in the concentrations of primary pollutants (SO$_2$ and NO$_x$) can be significantly influenced by changes in their emissions and meteorological conditions (McGregor and Bamzelis, 1995; Chang and Lee, 2007). For example, weaker winds can lead to the accumulation of pollution and stronger winds can quickly disperse air pollutants (Lo et al., 2006; Gietl and Klemm, 2009). Furthermore, the differences of oxidized S and N emissions among inventories and studies (Fig. 1, Table 1) can easily lead to an overestimation or underestimation of SO$_2$ and NO$_x$ concentrations. The relative change of SO$_2$ and NO$_x$ emissions in source and receptor countries in different research periods has important impacts on the source-receptor estimates. SO$_2$ emissions in China have experienced a relatively rapid increase, whereas those in Japan showed a slight decrease from 1980 to 2010; for NO$_x$ emissions both China and South Korea have experienced a sharp increase within the 30 years (Table S1). The assimilated meteorological fields, using field observations and improved oxidized S and N emissions data from satellite measurements, would be a good way to reduce the large differences in the input data. In previous

studies (Huang et al., 1995; Ichikawa and Fujita, 1995; Arndt et al., 1998; Holloway et al., 2002), the oxidation rates of SO$_2$ and NO$_2$ were obtained from a look-up table corresponding to the varying conditions, or were assumed to be proportional to the concentrations of SO$_2$ and NO$_2$. Actually, the formation of SO$_4^{2-}$ and NO$_3^-$ significantly depends on the concentrations of OH (SO$_2$+2OH→H$_2$SO$_4$, NO$_2$+OH→HNO$_3$), which therefore plays a critical role in the chemical conversions (Seinfeld, 2004), especially near the source areas (*e.g.*, within few hundred kilometers). Concentrations of OH radical were taken into account in calculating the concentrations of SO$_4^{2-}$ and NO$_3^-$ in more recent studies (Lin et al., 2008; Aikawa et al., 2010; Kajino et al., 2013; Ge et al., 2014). However, the deviation of OH concentrations remains large, due to uncertainties in the emissions of volatile organic compounds (VOCs) (Wang et al., 2014), which are the main sources of OH. There were also large variations in the way dry and wet deposition of oxidized S and N pollutants were dealt with. In the studies by Huang et al. (1995); Ichikawa and Fujita (1995); Arndt et al. (1998); Holloway et al. (2002), and Kajino et al. (2013), simplified removal rates (*e.g.*, a function of a precipitation rate or a pollutant concentration) were adopted. Arndt et al. (1998) performed a simple test to show the differences, by assuming a precipitation rate of 1 mm/hr and a SO$_2$ concentration of 10 μg/m^3 and concluded that removal rates differed by one or two orders of magnitude. Low removal rates result in large transport rates of pollutants away from source locations. The scavenging coefficient is more complicated and associated with many factors, so Lin et al. (2008); Aikawa et al. (2010), and Ge et al. (2014) used a cloud module to calculate in-cloud and precipitation scavenging and wet deposition amounts.

2.2. Trans-Pacific transport of oxidized S and N from East Asia

2.2.1. Field experiments
The regional trends of oxidized S and N pollutants have been of particular concern in Europe and North America since the 1970s.

Table 2 – Chemical transport models used in Asia and their main characteristics.

Name	Type	Organization/country	Main focus	References
CRIEPI trajectory model	Lagrangian 1-layer	CRIEPI, Japan	Long-term evaluation	Ichikawa and Fujita (1995)
ACDEP ASIA	Lagrangian 1-layer	CRIEPI, Japan and National Environmental Research Institute of Denmark	Long-term evaluation	Carmichael et al. (2001)
YU-SADM	3D Eulerian	Yonsei University, Korea	Episodic and long-term estimation of S sources and contributions	Carmichael et al. (2001)
ATMOS-2	Lagrangian multi-layer	University of Iowa	Long-term evaluation	Arndt and Carmichael (1995)
RIAM/RAMS	3D Eulerian	Kyushu University, Japan	Episodic and long-term chemical climate studies	Hayami and Ichikawa (2001)
OPU-Model	3D Eulerian	Osaka Prefecture University, Japan	Long-term deposition	Carmichael et al. (2001)
STEM	3D Eulerian	CGRER, University of Iowa	Episodic and long-term studies	Carmichael et al. (1991)
MATCH	3D Eulerian	Swedish Meteorological and Hydrological Institute	Long-term concentrations and depositions of O_3 and acid substances	Robertson et al. (1999)
CFORS	3D Eulerian	Japan	Transport and optical thickness of major tropospheric aerosols	Uno et al. (2003) Satake et al. (2004)
RADM	3D Eulerian	The United States	Episodic and long-term studies	Chang et al. (1987)
MSSP	3D Eulerian	Kyoto University, Japan	Episodic and long-term studies	Kajino et al. (2004)
Polair3D	3D Eulerian	France	Episodic and long-term studies	Boutahar et al. (2004)
PATH	3D Eulerian	Hong Kong Environmental Protection Agency	Episodic and long-term studies	Carmichael and Ueda (2008)
CADM	3D Eulerian	Korea	Episodic and long-term studies	Lee et al. (1998)
RAQM	3D Eulerian	The Institute of Atmospheric Physics, Chinese Academy of Sciences	Episodic and long-term studies	An et al. (2001) Han et al. (2006)
NAQPMS	3D Eulerian	The Institute of Atmospheric Physics, Chinese Academy of Sciences	Episodic and long-term studies	Wang et al. (2001)
CMAQ	3D Eulerian	EPA, the United States	Episodic and long-term studies	Byun and Ching (1999)
CAMx	3D Eulerian	ENVIRON, the United States	Episodic and long-term studies	ENVIRON (2011)
WRF-chem	3D Eulerian	NCAR, the United States	Episodic and long-term studies	Grell et al. (2005)

However, scientific data that could quantify the budget of oxidized S and N pollutants in East Asia were not available until the 1990s (Table 4). A series of experiments were conducted across East Asia, the North Pacific and western North America, to study Asian pollution outflow, chemical processes and long-range transport over the northwestern Pacific Ocean. For example, the National Aeronautics and Space Administration (NASA) funded three experiments: the Pacific Exploratory Mission-West A and B (PEM-West A and PEM-West B), the TRACE-P, and the INTEX-B. In addition, the Asian Pacific regional aerosol characterization experiment (ACE-Asia) was funded by the International Global Atmospheric Chemistry Program (IGAC). The EANET, in which 10 countries in Asia participate, also provides data to determine trends of acid deposition fluxes. Table 4 lists detailed information, including the observation period, region, objective, and platform of these field experiments. Based on the observational data, there was extensive evidence of pollution transport from East Asia to the North Pacific. Substantial increases of oxidized S and N pollutants were observed at the upper tropospheric levels in 1991 during the PEM-West A experiment (Singh et al., 1996; Talbot et al., 1996). Further, concentrations of NO_x and SO_2 in the Asian boundary layer outflow were about 2–5 times higher during the TRACE-P experiment than during PEM-West B, due to the economic growth of Asia during 1994–2001 (Dibb et al., 2003; O'Sullivan et al., 2004; Talbot et al., 2003; Liu et al., 2003).

2.2.2. Satellite remote sensing
Satellite observations can be used to provide new evidence for long-range transport of air pollutants by expanding spatial and temporal scales (Yu et al., 2008; Li et al., 2010; Clarisse et al., 2011; Hsu et al., 2012; Yu et al., 2012, 2013). Tropospheric columns of SO_2 and NO_2 can be derived from satellite retrievals of the global ozone monitoring experiment (GOME), the scanning imaging absorption spectrometer for atmospheric cartography (SCIAMACHY), the ozone monitoring instrument (OMI), and the infrared atmospheric sounding interferometer (IASI), among others. In addition, satellite measurements from the moderate-resolution imaging spectroradiometer (MODIS), multi-angle imaging spectroradiometer (MISR), and the cloud-aerosol lidar with orthogonal polarization (CALIOP), have been used to retrieve the distributions of aerosol particles and their properties, with an improved accuracy of aerosol optical depth (AOD) and enhanced capability of characterizing the aerosol type.

Many remote sensing observations have suggested that the long-range transport of sulfur species from East Asia occurs quite frequently and could exert strong impacts on

Table 3 – Comparison of parameters used in various studies for source–receptor relationship of oxidized S/N pollutants in East Asia.

Reference	Model	Period	Calculated emissions of S and N in China	Wet removal rates	Oxidation rates used	Contribution of China's emissions	
						To Japan	To Korea
Huang et al. (1995)	3-D Eulerian model	January 1989	9.1 Tg S/year	$W_{SO_2} = (4.3 + 0.78 \ln C_{SO_2}) + (0.14-0.019 \ln C_{SO_2}) \times P/3600$; $W_{SO_4^{2-}} = 9.2 \times 10^{-5} \times P^{0.83}$	Look-up table for oxidation rates	3.5%	7.9%
Ichikawa and Fujita (1995)	CRIEPI Trajectory Model	October 1988–September 1989	9 Tg S/year	$W_{SO_2} = 2.8 \times 10^{-5} \times P$, $W_{SO_4^{2-}} = 2.8 \times 10^{-5} \times P$	Parameterization for oxidation rates	40%	–
Arndt et al. (1998)	ATMOS-2	1990	11 Tg S/year	$W_{SO_2} = 1.1 \times 10^{-4} \times P$ (low latitude); $W_{SO_2} = (8.3 \times 10^{-5} + 2.7 \times 10^{-5} \times \sin(2 \times \pi \times \frac{\text{Julian day}-80}{365})) \times P$; $W_{SO_4^{2-}} = 7.0 \times 10^{-4} \times P^{0.7}$ (for in-cloud scavenging); $W_{SO_4^{2-}} = 6.0 \times 10^{-5} \times P$ (for below-cloud scavenging)	Parameterization for oxidation rates	17%	–
Holloway et al. (2002)	ATMOS-N	1990	2.5 Tg N/year	$W_{HNO_3} = 7.0 \times 10^{-4} \times P^{0.87}$ (for in-cloud scavenging); $W_{HNO_3} = 6.0 \times 10^{-5} \times P$ (for below-cloud scavenging)	Look-up table for oxidation rates	18%	26%
Lin et al. (2008)	CMAQ	2001	10.6 Tg S/year; 3.5 Tg N/year	$W_{gas} = \frac{1}{\tau_{washout}(1+\frac{\rho_{H_2O}}{W_T R H_i})}$, $W_{aerosol} = \frac{1}{\tau_{washout}}$ [a]	Chemical reactions with OH	15.4% 20.6%	– 39.1%
Aikawa et al. (2010)	based on CMAQ	2003–2005	20.2 Tg S/year; 11.6 Tg S/year	$W_{gas} = \frac{1}{\tau_{washout}(1+\frac{\rho_{H_2O}}{W_T R H_i})}$, $W_{aerosol} = \frac{1}{\tau_{washout}}$	Chemical reactions with OH	~50%–70% ~40%–60%	–
Kajino et al. (2013)	RAQM2	2006	6.3 Tg N/year	$W_{HNO_3} = 0.17 \times 10^{-4} \times P^{0.74}$	Chemical reactions with OH	~50%–60%	
Ge et al. (2014)	NAQPMS	2007	13.1 Tg S/year; 6.2 Tg N/year	$W_{gas} = \frac{1}{\tau_{washout}(1+\frac{\rho_{H_2O}}{W_T R H_i})}$, $W_{aerosol} = \frac{1}{\tau_{washout}}$	Chemical reactions with OH	13% 7%	–

P: the precipitation rate in mm/hr; H_i: Henry's Law coefficient for the pollutant i; $\tau_{washout}$: the washout time ($\tau_{washout} = \frac{W_T \Delta z_{dd}}{\rho_{H_2O} P_r}$); ρ_{H_2O}: the density of water; W_T (kg/m^3): the mean total water content; R: the universal gas constant, T (K): the in-cloud air temperature; P_r: the specified precipitation rate; Δz_{dd}: the cloud thickness; "–" refers to not reported in the literature.
[a] The formulas are from Byun and Ching (1999).

large areas downstream. Li et al. (2010) observed the transport and evolution of a plume containing substantial SO_2 and high loadings of dust particles from China to the Northwest Pacific, using OMI and MODIS satellite sensors. Clarisse et al. (2011) observed that a SO_2 plume formed over northeastern China and was transported to North America in five days, using IASI measurements. Hsu et al. (2012) analyzed SO_2 data observed by OMI during 2005–2008, and reported 16 SO_2 long-range transport episodes, out of 62 Asian outflow events.

2.2.3. Quantified transport

Large influences of the outflow of the S pollutants from East Asia have been confirmed by many model studies using the source-receptor method. An analysis by Liu et al. (2008) showed that sulfate from East Asia contributed approximately 20%–80% of the northern hemisphere sulfate found near the surface. Zhang et al. (2004) estimated that about 42% of S compounds (~25% as SO_2) emitted in East Asia were transported out of the region. Calculations by Satake et al. (2004) indicated that the eastward outflow of S accounted for 27% of all S emitted, during the ACE-Asia intensive observation period in spring 2001. Tan et al. (2002) showed that 20% of the anthropogenic SO_x ($SO_2 + SO_4^{2-}$) emitted over East Asia were exported from the region, primarily in the form of sulfate aerosols, during the late winter/early spring period.

2.3. Transport processes and influencing factors

2.3.1. Transport pattern

Intensive field campaigns over the western and eastern Pacific Ocean (e.g., TRACE-P, ACE-Asia, PEM-West) have shown that Asian aerosols are a complex mixture of dust and anthropogenic particles. Asian dust events from desert or loess areas in China and Mongolia occur frequently in Korea and Japan, and sulfate and nitrate have been predominantly found in the fine dust particles (Sugimoto et al., 2003; Zhang et al., 2003; Shimizu et al., 2004; Takami et al., 2005; Kim et al., 2012; Lee et al., 2013; Kunwar and Kawamura, 2014; Choi and Lee, 2014). Luan and Jaeglé (2013) found that 2/3 of the East Asian sulfate aerosol export events during spring were associated with substantial dust export.

SO_2 is not transported far away from the source regions itself, but it can be converted to sulfate (SO_4^{2-}) via heterogeneous and homogeneous reactions during transport (Arimoto et al., 1996; Zhang and Gao, 2007). Similarly, NO_x is usually dominant near its sources and its export is facilitated by converting into filterable nitrate (NO_3^-) or by the export of peroxyacetylnitrate (PAN), which can be transported long distances at cold temperatures (Singh et al., 1986). Field observations have shown that most of the increases in the nitrogen-containing reactive compounds within the outflow occur as PAN (only 0.5% as NO_x), and most of the SO_x occur as sulfate. Furthermore, export efficiencies to the marine boundary layer and the free troposphere were different, at around 20%–40% and 15%, respectively, for the nitrogen-containing reactive compounds, and around 25%–45% and 10%–20%, respectively, for the SO_x (Koike et al., 2003; Miyazaki et al., 2003).

2.3.2. Transport pathways

Lifting in the warm conveyor belt (WCB) of mid latitude cyclones, associated with rapid transport by the strong westerly winds, was considered the dominant pathway for the outflow of pollutants from Asia to the Pacific (Jacob et al., 2003; Singh et al., 2009). The relative lack of precipitation in the WCB means that sulfate and nitrate aerosols were efficiently injected into the middle troposphere; thus trans-Pacific transport of Asian pollutants usually occurs in the free atmosphere. Observations over the Pacific from the TRACE-P and INTEX-B experiments showed that the trans-Pacific transport of Asian pollutants (e.g., sulfate) generally occurred at higher altitudes (around 2–5 km). Luan and Jaeglé (2013) examined the vertical profiles of aerosol extinction coefficients, using satellite observations together with a global chemical transport model, and concluded that the outflows of sulfate from Asia occur at an altitude of about 2–6 km. Xiao et al. (1997) suggested that anthropogenic S, emitted from source regions in East Asia, is largely limited to the lower 4 km of the atmosphere, based on a regional-scale chemical model. In addition to the WCB mechanism, the local weak trough and saddle field over northeastern Asia were found to be another important uplifting mechanism of

Table 4 – Large field measurement networks or missions that have investigated long-range transport of pollutants.

Name	Period	Region	Objective	Platform	Reference
INTEX-B	17 April 2006–15 May 2006	Pacific Ocean and western North America	Trans-Pacific Asian pollution transport	Airborne	Singh et al. (2009)
EANET	2001–now	Ten countries in East Asia	Acid deposition in East Asia	Ground stations	An et al. (2002)
TRACE-P	February 2001–April 2001	Northwest Pacific	Asian pollution outflow and transport across the Pacific Ocean	Airborne	Jacob et al. (2003)
ACE-Asia	March 2001–May 2001	East Asia and Northwest Pacific	Aerosol properties	Airborne, ship, ground stations	Huebert et al. (2003)
PEM-West B	February 1994–March 1994	Northwest Pacific	Long-range transport of ozone and its precursors, sulfur species	Airborne and ground stations	Hoell et al. (1997)
PEM-West A	September 1991–October 1991				Hoell et al. (1996)

INTEX: the intercontinental chemical transport experiment; EANET: the acid deposition monitoring network in East Asia; ACE-Asia: the Asian Pacific regional aerosol characterization experiment; PEM: the Pacific Exploratory Mission; TRACE-P: transport and chemical evolution over the Pacific.

pollutants from continental Asia to the northwestern Pacific Rim (Li et al., 2013).

It should be mentioned that Huang et al. (1996) found some SO_2 and SO_4^{2-} transport paths on different vertical layers in East Asia, using a sulfur deposition and transport model. They showed that both SO_2 and SO_4^{2-} were usually transported from west to east under the influence of westerly flows in higher layers. In lower layers, S transport was different, dependent on season; the pathway was from northwest to southeast in winter and from southwest to northeast in summer. Arndt et al. (1998) showed that China's influence was most evident in southern and western Japan, with the smallest impact on areas in the north.

2.3.3. Seasonal variation

Transport patterns of oxidized S and N significantly vary throughout the year. Generally, the long-range transport of oxidized S and N pollutants is more pronounced in spring and winter, and trans-boundary oxidized S and N deposition levels peak during the two seasons (Kim et al., 2003; Lin et al., 2008; Luan and Jaeglé, 2013; Lee et al., 2014). A comparison between PEM-West A (in autumn) and PEM-West B (in spring) observations found that the western Pacific is more strongly impacted by sources of pollutants from continental Asia in spring, when westerly winds prevail (Kondo et al., 1997). The levels of reactive nitrogen in the continental air mass were found to be significantly higher during PEM-West B than during PEM-West A (Kondo et al., 1997). Some studies have attempted to apply backward trajectory models to identify the long-range transport and sources of observed polluted air masses that may affect air quality in South Korea and Japan (He et al., 2003; Kang et al., 2010, 2013; Kitayama et al., 2010; Takashima et al., 2011; Sakata et al., 2013; Zaizen et al., 2014). The backward trajectories in winter and spring, which were attributed to enhanced emissions and dominant westerly winds, were considered to carry more oxidized S and N pollutants than in summer and autumn. For example, Kitayama et al. (2010) detected remarkable seasonal changes and found that winter concentrations of SO_4^{2-} in precipitation in Japan were two to four times larger than summer ones, due to the long-range transport of the pollutant from continental Asia. Zaizen et al. (2014) tracked the air parcels of Mt. Kiso-Komagatake (3000 m above sea level) and suggested that the main sources of aerosols (including sulfate) in winter and spring were from continental Asia.

3. Recommendations

Trans-boundary issues have been serious concerns in East Asia, in particular the long-range transport of air pollutants from China to South Korea and Japan, due to persistent westerlies in this region. Increased energy demand will result in a large increase in SO_2 and NO_x emissions in East Asia (especially in China) in the future; there are strong research needs to mitigate NO_x emissions in future studies of air pollution.

The source–receptor relationships for oxidized S and N pollutants have been integrated with chemical transport models. In view of different conclusions drawn by different researchers about the source–receptor relationships, three recommendations were identified here. Firstly, tagged species methods, such as particulate matter source apportionment technology (PSAT) (Wagstrom et al., 2008), can significantly reduce the nonlinear effects that occur due to large changes in model inputs; however, few studies have applied such methods to identify the source–receptor relationships. Secondly, the uncertainties of SO_2 and NO_x emissions among the inventories and studies have greatly affected the concentrations of SO_2 and NO_x. There is an urgent requirement to improve field and satellite measurements of SO_2 and NO_x, and VOC emissions. Lastly, model comparison studies, such as the Model Inter-Comparison Study Asia Phase I and II (Carmichael et al., 2002; Carmichael and Ueda, 2008), have resulted in a better understanding of model performance and uncertainties. It may be true that the models would be highly consistent in identifying the main source–receptor relationships, if the same model inputs and parameters were used.

Furthermore, satellite-based studies have been generally used to assess air quality across oceans or continents, and few studies have centered in country-to-country scale pollutant transport due to coarse spatial resolution. High resolution data (e.g., 3 km) are needed to examine pollutant outflow and evaluate the influence of upwind countries on downwind countries.

It is obvious that observational studies and model simulations in isolation are insufficient to answer all the questions that arise in estimating trans-boundary air pollution. Field measurements, satellite observations, and model evaluations need to be linked to allow us to understand the mechanisms of long-range transport across global, regional, and country-to-country scales.

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CRIEPI: Central Research Institute of Electric Power Industry; YU-SADM: Yonsei University-sulfur acid deposition model; ATMOS: atmosphere; RIAM: Research Institute for Applied Mechanics; RAMS: regional atmospheric modeling system; OPU: Osaka Prefecture University; STEM: sulfur transport Eulerian model; MATCH: mesoscale atmospheric transport and chemistry; CFORS: the regional-scale aerosol transport model Chemical Weather Forecasting System; RADM: the regional acid deposition model; MSSP: a regional-scale Eulerian Model System for Soluble Particles; Polair3D: the 3D Eulerian chemistry-transport-model POLAIR; RAQM: an improved regional air quality model; NAQPMS: the nested air quality prediction modeling system; CMAQ: the models-3 community multiscale air quality; CAMx: the comprehensive air quality model with extensions; WRF: weather research forecast; CGRER: the Center for Global and Regional Environmental Research; EPA: Environmental Protection Agency; ENVIRON: ENVIRON International Corporation; NCAR: National Center for Atmospheric Research.

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REFERENCES

Aikawa, M., Ohara, T., Hiraki, T., Oishi, O., Tsuji, A., Yamagami, M., et al., 2010. Significant geographic gradients in particulate sulfate over Japan determined from multiple-site measurements and a chemical transport model: impacts of transboundary pollution from the Asian continent. Atmos. Environ. 44 (3), 381–391.

Akimoto, H., 2003. Global air quality and pollution. Science 302 (5651), 1716–1719.

An, J.L., Huang, M.Y., Wang, Z.F., Zhang, X.L., Ueda, H., Cheng, X.J., 2001. Numerical regional air quality forecast tests over the mainland of China. Water Air Soil Pollut. 130 (1–4), 1781–1786.

An, J.L., Ueda, H., Wang, Z.F., Matsuda, K., Kajino, M., Cheng, X.J., 2002. Simulations of monthly mean nitrate concentrations in precipitation over East Asia. Atmos. Environ. 36 (26), 4159–4171.

Arimoto, R., Duce, R.A., Savoie, D.L., Prospero, J.M., Talbot, R., Cullen, J.D., et al., 1996. Relationships among aerosol constituents from Asia and the North Pacific during PEM-West A. J. Geophys. Res. 101 (D1), 2011–2023.

Arndt, R.L., Carmichael, G.R., 1995. Long-range transport and deposition of sulfur in Asia. Water Air Soil Pollut. 85 (4), 2283–2288.

Arndt, R.L., Carmichael, G.R., Streets, D.G., Bhatti, N., 1997. Sulfur dioxide emissions and sectorial contributions to sulfur deposition in Asia. Atmos. Environ. 31 (10), 1553–1572.

Arndt, R.L., Carmichael, G.R., Roorda, J.M., 1998. Seasonal source: receptor relationships in Asia. Atmos. Environ. 32 (8), 1397–1406.

Biscaye, P.E., Grousset, F.E., Svensson, A.M., Bory, A., 2000. Eurasian air pollution reaches eastern North America. Science 290 (5500), 2258–2259.

Boutahar, J., Lacour, S., Mallet, V., Quelo, D., Roustan, Y., Sportisse, B., 2004. Development and validation of a fully modular platform for numerical modelling of air pollution: POLAIR. Int. J. Environ. Pollut. 22 (1–2), 17–28.

Byun, D.W., Ching, J.K.S., 1999. Science Algorithms of the EPA Models-3 Community Multiscale Air Quality (CMAQ) Modeling System, EPA/600/R-99/030, US EPA, Research Triangle Park, NC.

Carmichael, G.R., Ueda, H., 2008. MICS-Asia II: the model intercomparison study for Asia phase II. Atmos. Environ. 42 (15), 3465–3467.

Carmichael, G.R., Peters, L.K., Saylor, R.D., 1991. The STEM-II regional scale acid deposition and photochemical oxidant model: I. An overview of model development and applications. Atmos. Environ. 25 (10), 2077–2090.

Carmichael, G.R., Hayamib, H., Caloria, G., Uno, I., Cho, S.Y., Engardtd, M., et al., 2001. Model intercomparison study of long range transport and sulfur deposition in East Asia (MICS-ASIA). Water Air Soil Pollut. 130 (1–4), 51–62.

Carmichael, G.R., Calori, G., Hayami, H., Uno, I., Cho, S.Y., Engardt, M., et al., 2002. The MICS-Asia study: model intercomparison of long-range transport and sulfur deposition in East Asia. Atmos. Environ. 36 (2), 175–199.

Chang, S.C., Lee, C.T., 2007. Evaluation of the trend of air quality in Taipei, Taiwan from 1994 to 2003. Environ. Monit. Assess. 127 (1–3), 87–96.

Chang, J.S., Brost, R.A., Isaksen, I.S.A., Madronich, S., Middleton, P., Stockwell, W.R., et al., 1987. A three-dimensional Eulerian acid deposition model: physical concepts and formulation. J. Geophys. Res. 92 (D12), 14681–14700.

Che, H.Z., Zhang, X.Y., Li, Y., Zhou, Z.J., Qu, J.J., 2007. Horizontal visibility trends in China 1981–2005. Geophys. Res. Lett. 34 (24), L24706.

Chiwa, M., Enoki, T., Higashi, N., Kumagai, T., Otsuki, K., 2013. The increased contribution of atmospheric nitrogen deposition to nitrogen cycling in a rural forested area of Kyushu, Japan. Water Air Soil Pollut. 224, 1763.

Choi, H., Lee, A.S., 2014. Double compressions of atmospheric depth by geopotential tendency, vorticity, and atmospheric boundary layer affected abrupt high particulate matter concentrations at a coastal city for a yellow dust period in October. Adv. Meteor., Article ID 756230

Clarisse, L., Fromm, M., Ngadi, Y., Emmons, L., Clerbaux, C., Hurtmans, D., et al., 2011. Intercontinental transport of anthropogenic sulfur dioxide and other pollutants: an infrared remote sensing case study. Geophys. Res. Lett. 38 (19), L19806.

Cofala, J., Bertok, I., Borken-Kleefeld, J., Heyes, C., Klimont, Z., Rafaj, P., et al., 2012. Emissions of Air Pollutants for the World Energy Outlook 2012 Energy Scenarios. International Institute for Applied Systems Analysis, Laxenburg, Austria.

Cogbill, C.V., Likens, G.E., 1974. Acid precipitation in the Northeastern United States. Water Resour. Res. 10 (6), 1133–1137.

Dibb, J.E., Talbot, R.W., Scheuer, E.M., Seid, G., Avery, M.A., Singh, H.B., 2003. Aerosol chemical composition in Asian continental outflow during TRACE-P campaign: comparison with PEM-West B. J. Geophys. Res. 108 (D21), 8815.

ENVIRON, 2011. User's Guide: Comprehensive Air Quality Model With Extensions Version 5.40. ENVIRON International Corporation, Novato, California.

Foell, W., Green, C., Amann, M., Bhattacharya, S., Carmichael, G., Chadwick, M., et al., 1995. Energy use, emissions, and air pollution reduction strategies. Water Air Soil Pollut. 85 (4), 2277–2282.

Fujita, S., 1996. An estimation for atmospheric sulfur budget over the Japanese archipelago. Environ. Sci. 9 (2), 185–199.

Ge, B.Z., Wang, Z.F., Xu, X.B., Wu, J.B., Yu, X.L., Li, J., 2014. Wet deposition of acidifying substances in different regions of China and the rest of East Asia: modeling with updated NAQPMS. Environ. Pollut. 187, 10–21.

Gietl, J.K., Klemm, O., 2009. Analysis of traffic and meteorology on airborne particulate matter in Münster, Northwest Germany. J. Air Waste Manage. Assoc. 59 (7), 809–818.

Grell, G.A., Peckham, S.E., Schmitz, R., McKeen, S.A., Frost, G., Skamarock, W.C., et al., 2005. Fully coupled "online" chemistry within the WRF model. Atmos. Environ. 39 (37), 6957–6975.

Grousset, F.E., Ginoux, P., Bory, A., Biscaye, P.E., 2003. Case study of a Chinese dust plume reaching the French Alps. Geophys. Res. Lett. 30 (6), 1277.

Han, B., Kim, J., Cho, S.Y., 2001. A numerical simulation of acid deposition in East Asia. Water Air Soil Pollut. 130 (1–4), 487–492.

Han, Z.W., Ueda, H., Sakurai, T., 2006. Model study on acidifying wet deposition in East Asia during wintertime. Atmos. Environ. 40 (13), 2360–2373.

Hao, J.M., Wang, S.X., Liu, B.J., He, K.B., 2000. Designation of acid rain and SO_2 control zones and control policies in China. J. Environ. Sci. Health AToxic/Hazard. Subst. Environ. Eng. 35 (10), 1901–1914.

Hao, J.M., Tian, H.Z., Lu, Y.Q., 2002. Emission inventories of NO_x from commercial energy consumption in China, 1995–1998. Environ. Sci. Technol. 36 (4), 552–560.

Hatakeyama, S., Hanaoka, S., Ikeda, K., Watanabe, I., Araki, T., Sadanaga, Y., et al., 2011. Aerial observation of aerosols transported from East Asia-chemical composition of aerosols and layered structure of an air mass over the East China Sea. Aerosol Air Aual Res. 11 (5), 497–507.

Hayami, H., Ichikawa, Y., 2001. Sensitivity of long-range transport of sulfur compounds to vertical distribution of sources. Water Air Soil Pollut. 130 (1–4), 283–288.

He, Z., Kim, Y.J., Ogunjobi, K.O., Hong, C.S., 2003. Characteristics of $PM_{2.5}$ species and long-range transport of air masses at Taean background station, South Korea. Atmos. Environ. 37 (2), 219–230.

Hoell, J.M., Davis, D.D., Liu, S.C., Newell, R., Shipham, M., Akimoto, H., et al., 1996. Pacific exploratory Mission-West A (PEM-West A): September–October 1991. J. Geophys. Res.-Atmos. 101 (D1), 1641–1653.

Hoell, J.M., Davis, D.D., Liu, S.C., Newell, R.E., Akimoto, H., McNeal, R.J., et al., 1997. The Pacific exploratory mission west phase B: February–March, 1994. J. Geophys. Res.-Atmos. 102 (D23), 28223–28239.

Holloway, T., Levy, H., Carmichael, G., 2002. Transfer of reactive nitrogen in Asia: development and evaluation of a source-receptor model. Atmos. Environ. 36 (26), 4251–4264.

Hsu, N.C., Li, C., Krotkov, N.A., Liang, Q., Yang, K., Tsay, S.C., 2012. Rapid transpacific transport in autumn observed by the A-train satellites. J. Geophys. Res. 117 (D6), D06312.

Huang, M.Y., Wang, Z.F., He, D.Y., Xu, H.Y., Zhou, L., 1995. Modeling studies on sulfur deposition and transport in East Asia. Water Air Soil Pollut. 85 (4), 1921–1926.

Huang, M.Y., Wang, Z.F., He, D.Y., Xu, H.Y., Zhou, L., Gao, H.W., 1996. Studies on cross-boundary transport of sulfur in China and East Asia. Clim. Environ. Res. 1 (1), 55–62.

Huebert, B.J., Bates, T., Russell, P.B., Shi, G.Y., Kim, Y.J., Kawamura, K., et al., 2003. An overview of ACE-Asia: strategies for quantifying the relationships between Asian aerosols and their climatic impacts. J. Geophys. Res. -Atmos. 108 (D23), 8633.

Ichikawa, Y., Fujita, S., 1995. An analysis of wet deposition of sulfate using a trajectory model for East Asia. Water Air Soil Pollut. 85 (4), 1927–1932.

Ichikawa, Y., Hayami, H., Fujita, S., 1998. A long-range transport model for East Asia to estimate sulfur deposition in Japan. J. Appl. Meteor. 37 (10), 1364–1374.

Jacob, D.J., Crawford, J.H., Kleb, M.M., Connors, V.S., Bendura, R.J., Raper, J.L., et al., 2003. Transport and chemical evolution over the Pacific (TRACE-P) aircraft mission: design, execution, and first results. J. Geophys. Res. 108 (D20), 9000.

Jaffe, D., Anderson, T., Covert, D., Kotchenruther, R., Trost, B., Danielson, J., et al., 1999. Transport of Asian air pollution to North America. Geophys. Res. Lett. 26 (6), 711–714.

Kajino, M., Ueda, H., Satsumabayashi, H., An, J.L., 2004. Impacts of the eruption of Miyakejima Volcano on air quality over far east Asia. J. Geophys. Res. 109 (D21), D21204.

Kajino, M., Sato, K., Inomata, Y., Ueda, H., 2013. Source-receptor relationships of nitrate in Northeast Asia and influence of sea salt on the long-range transport of nitrate. Atmos. Environ. 79, 67–78.

Kang, J., Cho, B.C., Lee, C.B., 2010. Atmospheric transport of water-soluble ions (NO_3^-, NH_4^+ and $nss-SO_4^{2-}$) to the southern East Sea (Sea of Japan). Sci. Total Environ. 408 (11), 2369–2377.

Kang, J., Choi, M.S., Yi, H.I., Jeong, K.S., Chae, J.S., Cheong, C.S., 2013. Elemental composition of different air masses over Jeju Island, South Korea. Atmos. Res. 122, 150–164.

Kim, J., Han, B., Cho, S., 2003. A numerical simulation of annual acid deposition amount in Korea. Atmos. Environ. 37 (12), 1703–1713.

Kim, H.S., Kim, D.S., Kim, H., Yi, S.M., 2012. Relationship between mortality and fine particles during Asian dust, smog-Asian dust, and smog days in Korea. Int. J. Environ. Health Res. 22 (6), 518–530.

Kitayama, K., Murao, N., Hara, H., 2010. PMF analysis of impacts of SO2 from Miyakejima and Asian Continent on precipitation sulfate in Japan. Atmos. Environ. 44 (1), 95–105.

Koike, M., Kondo, Y., Kita, K., Takegawa, N., Masui, Y., Miyazaki, Y., et al., 2003. Export of anthropogenic reactive nitrogen and sulfur compounds from the East Asia region in spring. J. Geophys. Res. 108 (D20), 8789.

Kondo, Y., Koike, M., Kawakami, S., Singh, H.B., Nakajima, H., Gregory, G.L., et al., 1997. Profiles and partitioning of reactive nitrogen over the Pacific Ocean in winter and early spring. J. Geophys. Res. 102 (D23), 28405–28424.

Kunwar, B., Kawamura, K., 2014. One-year observations of carbonaceous and nitrogenous components and major ions in the aerosols from subtropical Okinawa Island, an outflow region of Asian dusts. Atmos. Chem. Phys. 14 (4), 1819–1836.

Larssen, T., Lydersen, E., Tang, D.G., He, Y., Gao, J.X., Liu, H.Y., et al., 2006. Acid rain in China. Environ. Sci. Technol. 40 (2), 418–425.

Lee, T.Y., Kim, S.B., Lee, S.M., Park, S.U., Kim, D.S., Shin, H.C., 1998. Numerical simulation of air quality and acid deposition for episodic cases in eastern Asia. Korean J. Atmos. Sci. 1 (2), 126–144.

Lee, S., Ho, C.H., Lee, Y.G., Choi, H.J., Song, C.K., 2013. Influence of transboundary air pollutants from China on the high-PM_{10} episode in Seoul, Korea for the period October 16–20, 2008. Atmos. Environ. 77, 430–439.

Lee, H.J., Kim, S.W., Brioude, J., Cooper, O.R., Frost, G.J., Kim, C.H., et al., 2014. Transport of NOx in East Asia identified by satellite and in situ measurements and Lagrangian particle dispersion model simulations. J. Geophys. Res. 119 (5), 2574–2596.

Li, C., Krotkov, N.A., Dickerson, R.R., Li, Z.Q., Yang, K., Chin, M., 2010. Transport and evolution of a pollution plume from northern China: a satellite-based case study. J. Geophys. Res. 115 (D7), D00K03.

Li, J., Wang, Z.F., Yamaji, K., Takigawa, M., Kanaya, Y., Taketani, F., et al., 2013. Uplifting of Asian continental pollution plumes from the boundary layer to the free atmosphere over the Northwestern Pacific rim in spring. Sola 9, 40–44.

Lin, M.Y., Oki, T., Bengtsson, M., Kanae, S., Holloway, T., Streets, D.G., 2008. Long-range transport of acidifying substances in east Asia — part II: source-receptor relationships. Atmos. Environ. 42 (24), 5956–5967.

Liu, H.Y., Jacob, D.J., Bey, I., Yantosca, R.M., Duncan, B.N., Sachse, G.W., 2003. Transport pathways for Asian pollution outflow over the Pacific: interannual and seasonal variations. J. Geophys. Res. 108 (D20), 8786.

Liu, J., Mauzerall, D.L., Horowitz, L.W., 2008. Source-receptor relationships between East Asian sulfur dioxide emissions and Northern Hemisphere sulfate concentrations. Atmos. Chem. Phys. 8 (14), 3721–3733.

Liu, X.G., Li, J., Qu, Y., Han, T., Hou, L., Gu, J.W., et al., 2013. Formation and evolution mechanism of regional haze: a case study in the megacity Beijing, China. Atmos. Chem. Phys. 13 (9), 4501–4514.

Lo, J.C.F., Lau, A.K.H., Fung, J.C.H., Chen, F., 2006. Investigation of enhanced cross-city transport and trapping of air pollutants by coastal and urban land-sea breeze circulations. J. Geophys. Res. 111 (D14), D14104.

Lu, Z., Streets, D.G., Zhang, Q., Wang, S., Carmichael, G.R., Cheng, Y.F., et al., 2010. Sulfur dioxide emissions in China and sulfur trends in East Asia since 2000. Atmos. Chem. Phys. 10 (13), 6311–6331.

Luan, Y., Jaeglé, L., 2013. Composite study of aerosol export events from East Asia and North America. Atmos. Chem. Phys. 13 (3), 1221–1242.

Ma, J.Z., Liu, H.L., Hauglustaine, D., 2002. Summertime tropospheric ozone over China simulated with a regional chemical transport model 1. Model description and evaluation. J. Geophys. Res. 107 (D22) ACH 27-1-ACH 27-13.

McGregor, G.R., Bamzelis, D., 1995. Synoptic typing and its application to the investigation of weather air pollution relationships, Birmingham, United Kingdom. Theor. Appl. Climatol. 51 (4), 223–236.

Miyazaki, Y., Kondo, Y., Koike, M., Fuelberg, H.E., Kiley, C.M., Kita, K., et al., 2003. Synoptic-scale transport of reactive nitrogen

over the Western Pacific in spring. J. Geophys. Res. 108 (D20), 8788.

Monks, P.S., Granier, C., Fuzzi, S., Stohl, A., William, M.L., Akimoto, H., et al., 2009. Atmospheric composition change-global and regional air quality. Atmos. Environ. 43 (33), 5268–5350.

NADP (National Atmospheric Deposition Program), 2000. National Atmospheric Deposition Program Annual Data Summary: Precipitation Chemistry in the United States. University of Illinois, Champaign, NADP Program Office, Illinois State Water Survey.

National Research Council, 1991. Rethinking the Ozone Problem in Urban and Regional Air Pollution. National Academies Press, Washington, D. C.

Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., et al., 2007. An Asian emission inventory of anthropogenic emission sources for the period 1980–2020. Atmos. Chem. Phys. 7 (16), 4419–4444.

O'Sullivan, D.W., Heikes, B.G., Snow, J., Burrow, P., Avery, M., Blake, D.R., et al., 2004. Long-term and seasonal variations in the levels of hydrogen peroxide, methylhydroperoxide, and selected compounds over the Pacific Ocean. J. Geophys. Res. 109 (D15), D15S13.

Park, M.E., Song, C.H., Park, R.S., Lee, J., Kim, J., Lee, S., et al., 2014. New approach to monitor transboundary particulate pollution over Northeast Asia. Atmos. Chem. Phys. 14 (2), 659–674.

Quinn, P.K., Bates, T.S., 2003. North American, Asian, and Indian haze similar regional impacts on climate. Geophys. Res. Lett. 30 (11), 1555–1558.

Richter, A., Burrows, J.P., Nüß, H., Granier, C., Niemeier, U., 2005. Increase in tropospheric nitrogen dioxide over China observed from space. Nature 437 (7055), 129–132.

Robertson, L., Langner, J., Engardt, M., 1999. An Eulerian limited-area atmospheric transport model. J. Appl. Meteor. 38 (2), 190–210.

Sakata, M., Ishikawa, T., Mitsunobu, S., 2013. Effectiveness of sulfur and boron isotopes in aerosols as tracers of emissions from coal burning in Asian continent. Atmos. Environ. 67, 296–303.

Satake, S., Uno, I., Takemura, T., Carmichael, G.R., Tang, Y.H., Streets, D., et al., 2004. Characteristics of Asian aerosol transport simulated with a regional-scale chemical transport model during the ACE-Asia observation. J. Geophys. Res. 109 (D19), D19S22.

Seinfeld, J.H., 2004. Air pollution: a half century of progress. AICHE J. 50 (6), 1096–1108.

SEPA (State Environmental Protection Administration, 2003. Report on the State of the Environment in China. State Environmental Protection Administration of China, Beijing.

Shimizu, A., Sugimoto, N., Matsui, I., Arao, K., Uno, I., Murayama, T., et al., 2004. Continuous observations of Asian dust and aerosols by polarization lidars in China and Japan during ACE-Asia. J. Geophys. Res. 109 (D19), D19S17.

Singh, H.B., Salas, L.J., Viezee, W., 1986. Global distribution of peroxyacetylnitrate. Nature 321 (6070), 588–591.

Singh, H.B., Herlth, D., Kolyer, R., Salas, L., Bradshaw, J.D., Sandholm, S.T., et al., 1996. Reactive nitrogen and ozone over the Western Pacific: distribution, partitioning, and sources. J. Geophys. Res. 101 (D1), 1793–1808.

Singh, H.B., Brune, W.H., Crawford, J.H., Flocke, F., Jacob, D.J., 2009. Chemistry and transport of pollution over the Gulf of Mexico and the Pacific: spring 2006 INTEX-B campaign overview and first results. Atmos. Chem. Phys. 9 (7), 2301–2318.

Stern, D.I., 2006. Reversal of the trend in global anthropogenic sulfur emissions. Global Environ. Change 16 (2), 207–220.

Stohl, A., Forster, C., Huntrieser, H., Mannstein, H., McMillan, W.W., Petzold, A., et al., 2007. Aircraft measurements over Europe of an air pollution plume from Southeast Asia — aerosol and chemical characterization. Atmos. Chem. Phys. 7 (3), 913–937.

Streets, D.G., Waldhoff, S.T., 2000. Present and future emissions of air pollutants in China: SO_2, NO_x, and CO. Atmos. Environ. 34 (3), 363–374.

Streets, D.G., Carmichael, G.R., Amann, M., Arndt, R.L., 1999. Energy consumption and acid deposition in Northeast Asia. Ambio 28 (2), 135–143.

Streets, D.G., Tsai, N.Y., Akimoto, H., Oka, K., 2000. Sulfur dioxide emissions in Asia in the period 1985–1997. Atmos. Environ. 34 (26), 4413–4424.

Streets, D.G., Tsai, N.Y., Akimoto, H., Oka, K., 2001. Trends in emissions of acidifying species in Asia, 1985–1997. Water Air Soil Pollut. 130 (1–4), 187–192.

Streets, D.G., Bond, T.C., Carmichael, G.R., Fernandes, S.D., Fu, Q., He, D., et al., 2003. An inventory of gaseous and primary aerosol emissions in Asia in the year 2000. J. Geophys. Res. 108 (D21), 8809.

Sugimoto, N., Uno, L., Nishikawa, M., Shimizu, A., Matsui, I., Dong, X.H., et al., 2003. Recent heavy Asian dust in Beijing in 2002: observations and model analysis of recent events. Geophys. Res. Lett. 30 (12), 1640.

Takami, A., Miyoshi, T., Shimono, A., Hatakeyama, S., 2005. Chemical composition of fine aerosol measured by AMS at Fukue Island, Japan during APEX period. Atmos. Environ. 39 (27), 4913–4924.

Takashima, H., Irie, H., Kanaya, Y., Akimoto, H., 2011. Enhanced NO_2 at Okinawa Island, Japan caused by rapid air-mass transport from China as observed by MAX-DOAS. Atmos. Environ. 45 (15), 2593–2597.

Talbot, R.W., Dibb, J.E., Klemm, K.I., Bradshaw, J.D., Sandholm, S.T., Blake, D.R., et al., 1996. Chemical characteristics of continental outflow from Asia to the troposphere over the Western Pacific Ocean during September–October 1991: results from PEM-West A. J. Geophys. Res. 101 (D1), 1713–1725.

Talbot, R.W., Dibb, J., Scheuer, E., Seid, G., Russo, R., Sandholm, S., et al., 2003. Reactive nitrogenin Asian continental outflow over the Western Pacific: results from the NASA transport and chemical evolution over the Pacific (TRACE-P) airborne mission. J. Geophys. Res. 108 (D20), 8803.

Tan, Q., Huang, Y., Chameides, W.L., 2002. Budget and export of anthropogenic SO_x from East Asia during continental outflow conditions. J. Geophys. Res. 107 (D13), 4167.

Tang, J., Xu, X.B., Ba, J., Wang, S.F., 2010. Trends of the precipitation acidity over China during 1992–2006. Chin. Sci. Bull. 55 (17), 1800–1807.

Tie, X.X., Wu, D., Brasseur, G., 2009. Lung cancer mortality and exposure to atmospheric aerosol particles in Guangzhou, China. Atmos. Environ. 43 (14), 2375–2377.

Uno, I., Carmichael, G.R., Streets, D.G., Tang, Y.H., Yienger, J.J., Satake, S., et al., 2003. Regional chemical weather forecasting system CFORS: model descriptions and analysis of surface observations at Japanese island stations during the ACE-Asia experiment. J. Geophys. Res. 108 (D23), 8668.

van Aardenne, J.A., Carmichael, G.R., Levy, H., Streets, D., Hordijk, L., 1999. Anthropogenic NOx emissions in Asia in the period 1990–2020. Atmos. Environ. 33 (4), 633–646.

van Curen, R.A., Cahill, T.A., 2002. Asian aerosols in North America: frequency and concentration of fine dust. J. Geophys. Res. 107 (D24) AAC 19-1-AAC 19-16.

van Donkelaar, A., Martin, R.V., Leaitch, W.R., Macdonald, A.M., Walker, T.W., Streets, D.G., et al., 2008. Analysis of aircraft and satellite measurements from the Intercontinental Chemical Transport Experiment (INTEX-B) to quantify long-range transport of East Asian sulfur to Canada. Atmos. Chem. Phys. 8 (11), 2999–3014.

Wagstrom, K.M., Pandis, S.N., Yarwood, G., Wilson, G.M., Morris, R., 2008. Development and application of a computationally efficient particulate matter apportionment algorithm in a three-dimensional chemical transport model. Atmos. Environ. 42 (22), 5650–5659.

Wang, T., Kwok, J.Y.H., 2003. Measurement and analysis of a multiday photochemical smog episode in the Pearl River Delta of China. J. Appl. Meteor. 42 (3), 404–416.

Wang, Z., Maeda, T., Hayashi, M., Hsiao, L.F., Liu, K.Y., 2001. A nested air quality prediction modeling system for urban and regional scales: application for high-ozone episode in Taiwan. Water Air Soil Pollut. 130 (1–4), 391–396.

Wang, Z., Xie, F.Y., Sakurai, T., Ueda, H., Han, Z., Carmichael, G.R., et al., 2008. MICS-Asia II: model inter-comparison and evaluation of acid deposition. Atmos. Environ. 42 (15), 3528–3542.

Wang, F., An, J.L., Li, Y., Tang, Y.J., Lin, J., Qu, Y., et al., 2014. Impacts of uncertainty in AVOC emissions on the summer ROx budget and ozone production rate in the three most rapidly-developing economic growth regions of China. Adv. Atmos. Sci. 31 (6), 1331–1342.

Wu, D., Tie, X.X., Li, C.C., Ying, Z.M., Lau, A.K.H., Huang, J., et al., 2005. An extremely low visibility event over the Guangzhou region: a case study. Atmos. Environ. 39 (35), 6568–6577.

Xiao, H., Carmichael, G.R., Durchenwald, J., Thornton, D., Bandy, A., 1997. Long-range transport of SOx and dust in East Asia during the PEM B Experiment. J. Geophys. Res. 102 (D23), 28589–28612.

Yu, H.B., Remer, L.A., Chin, M., Bian, H.S., Kleidman, R.G., Diehl, T., 2008. A satellite-based assessment of transpacific transport of pollution aerosol. J. Geophys. Res. 113 (D14), D14S12.

Yu, H.B., Remer, L.A., Chin, M., Bian, H.S., Tan, Q., Yuan, T.L., et al., 2012. Aerosols from overseas rival domestic emissions over North America. Science 337 (6094), 566–569.

Yu, H.B., Remer, L.A., Kahn, R.A., Chin, M., Zhang, Y., 2013. Satellite perspective of aerosol Intercontinental transport: from qualitative tracking to quantitative characterization. Atmos. Res. 124, 73–100.

Zaizen, Y.J., Naoe, H., Takahashi, H., Igarashi, Y., 2014. Number concentrations and elemental compositions of aerosol particles observed at Mt. Kiso-Komagatake in Central Japan, 2010–2013. Atmos. Environ. 90, 1–9.

Zhang, K., Gao, H.W., 2007. The characteristics of Asian-dust storms during 2000–2002: from the source to the sea. Atmos. Environ. 41 (39), 9136–9145.

Zhang, D.Z., Iwasaka, Y., Shi, G.Y., Zang, J.Y., Matsuki, A., Trochkine, D., 2003. Mixture state and size of Asian dust particles collected at Southwestern Japan in spring 2000. J. Geophys. Res. 108 (D24), 4760.

Zhang, M.G., Uno, I., Yoshida, Y., Xu, Y.F., Wang, Z.F., Akimoto, H., et al., 2004. Transport and transformation of sulfur compounds over East Asia during the TRACE-P and ACE-Asia campaigns. Atmos. Environ. 38 (40), 6947–6959.

Zhang, Q., Streets, D.G., He, K.B., Wang, Y.X., Richter, A., Burrows, J.P., et al., 2007. NOx emission trends for China, 1995–2004: the view from the ground and the view from space. J. Geophys. Res. 112 (D22), D22306.

Zhang, Q., Streets, D.G., Carmichael, G.R., He, K.B., Huo, H., Kannari, A., et al., 2009. Asian emissions in 2006 for the NASA INTEX-B mission. Atmos. Chem. Phys. 9 (14), 5131–5153.

Zhang, X.Y., van Geffen, J., Liao, H., Zhang, P., Lou, S.J., 2012. Spatiotemporal variations of tropospheric SO_2 over China by SCIAMACHY observations during 2004–2009. Atmos. Environ. 60, 238–246.