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SEASONAL SOURCE–RECEPTOR RELATIONSHIPS IN ASIA

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Abstract—Seasonal source-receptor relationships are estimated for countries in Asia and the Indian sub-continent and the impact of long-range transport on countries' deposition are explored through these relationships. The influence of precipitation patterns and changes in flow fields on deposition in southeast Asia and the Indian sub-continent is also demonstrated. Many of the small sulfur emitting countries in the region are found to receive more sulfur deposition than they emit, with the majority of their deposition coming from neighboring or even distant countries. For example, Vietnam accounts for 35% of its own deposition while Thailand contributes 19% and China 39%. Similarly, over 60% of the sulfur deposited on Nepal is due to Indian emissions. China's contribution to Japan's deposition is shown to exhibit strong seasonal dependence, with winter and spring contributions 2.5 times higher than summer and autumn. China and South Korea are found to play a major role in the deposition in southern and western Japan while volcanoes and domestic sources dominate in the north and east. The impact of Chinese emissions on Japan's deposition is found to be highly sensitive to wet removal rates. © 1998 Published by Elsevier Science Ltd. All rights reserved

Key word index: Acid deposition, Asia, long-range transport, seasonality, source–receptor relationships, sulfur deposition.

1. INTRODUCTION

The rapid growth in many Asian economies in recent years has resulted in significant growth in the region's energy needs (Kato and Akimoto, 1991). Asia boasts some of the fastest growing economies in the world. This economic expansion has resulted in widespread degradation of air quality in the region. Many urban centers in China have air pollution levels exceeding WHO ambient standards (Florig, 1995) and many locations in Asia are now reporting acidic precipitation (Khemani *et al.*, 1989; Wang and Wang, 1995; Mohamed and Kamsah, 1993). A major source of the increased acidification in Asia is the rise in sulfur emissions from coal combustion in the region (Hordijk *et al.*, 1995). Over the past two decades China's SO_2 emissions have grown by more than a factor of three (Cofala, 1995) and this trend is expected to continue. Asia's total SO_2 emissions may increase by another factor of three between 1990 and the year 2020 (Foell *et al.*, 1995).

How and where this sulfur will be deposited is an area of increasing environmental concern (Rodhe *et al.*, 1992; Hordijk *et al.*, 1995). A number of models have been developed to understand the transport and deposition of sulfur in the region (Robertson *et al.*, 1995; Sato *et al.*, 1996; Arndt *et al.*, 1997; Kotamarthi and Carmichael, 1990). These models assist investigators in understanding the impact of current emissions on sulfur deposition in Asia and in anticipating how projected emissions may affect the region's environment in the future.

To better understand how future emissions may affect acidification in Asia, it is necessary to develop relationships between sources of sulfur and their resulting deposition patterns. As part of the RAINS-ASIA Project (Foell *et al.*, 1995) a long-range transport model (ATMOS) was developed for studying the transport and deposition of sulfur in Asia (Arndt *et al.*, 1997; Arndt and Carmichael, 1995). The ATMOS model calculates the deposition from each emission source directly, allowing deposition from a specific point source, region, or country to be analyzed separately. This information can be used to identify which sources contribute to the deposition at a specific receptor site. This data can then be consolidated to provide source–receptor information at a region-to-region or country-to-country level.

In this article the influence of seasonality on Asia's eposition is presented, with emphasis on intra-annual changes in source–receptor relationships.

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Comparison of model calculated deposition and concentrations with observations from several monitoring networks is performed. The changes in receptor countries' total deposition amounts and in the sources of their deposition on an intra-annual basis are presented. The predicted changes in seasonal deposition are compared with meteorological and emission patterns for Asia to illustrate the role that these factors play in Asia's sulfur budget. Furthermore, variations in deposition sources' contributions as a function of location within Japan are shown. Finally, the predicted influence of China on Japan's deposition is compared with other source–receptor model calculations.

2. MODEL DESCRIPTION AND COMPARISON WITH OBSERVED DATA

ATMOS is a modified version of NOAA's Air Resources Laboratory Branching Atmospheric Trajectory model (Heffter, 1984). This three-layer Lagrangian model calculates SO_2 and sulfate concentrations and their wet and dry deposition on a $1^\circ \times 1^\circ$ resolution. Within the modeling domain, 10° South to 55° North latitude and 60° East to 150° East longitude, SO_2 emission plumes are modeled as puffs released every three hours from the emission source location. Each puff is assigned a mass proportional to the source strength, and is assumed to mix uniformly in the vertical throughout an assigned layer, and to diffuse with a Gaussian distribution in the horizontal. Individual emission puffs are followed throughout their transport and deposition "lifetimes". Each puff's transport is followed for up to five days, until its mass falls below a cut-off value, or until it is transported beyond the modeling domain, whichever occurs first. As the puffs are transported, SO_2 is chemically converted to sulfate, and SO_2 and sulfate are deposited to the surface in the form of wet and dry species. Concentrations and deposition are calculated on a one hour basis which can then be aggregated to a monthly or annual basis. A detailed description of the ATMOS model including selection of the chemical conversion and wet and dry removal parameters used in the ATMOS model base year calculations and comparison of the rates used with those used in other models is presented in Arndt *et al.* (1997).

The base year for model calculations was 1990. Meteorological data used for this year was from upper air sondings (rawinsonde and pibal observations of wind speed and temperature from the surface to 500 mb) provided by the National Climatic Center (NMC) of the U.S.A. National Oceanic and Atmospheric Administration (NOAA) and precipitation data was NMC analyzed fields at 1.45° resolution obtained from the U.S.A. National Center for Atmospheric Research (NCAR). The emissions inventory was the SO_x inventory compiled as part of the RAINS-ASIA project (Streets *et al.*, 1995). This $1^\circ \times 1^\circ$ degree inventory provides anthropogenic emissions calculated for 1990 for the area between 10° south and 55° north latitude and 60° east and 150° east longitude excluding Afghanistan and the countries of the former Soviet Union. Anthropogenic emissions include both elevated large point sources and surface sources. In addition, SO_x emissions from non-erupting volcanoes in the region were also included (Fujita, 1992; Spiro *et al.*, 1992). The spatial distribution of the 1990 emissions inventory is presented by Arndt and Carmichael (1995).

The calculated 1990 deposition is presented in Fig. 1. Model calculated sulfur deposition in Asia exhibits widespread variation with areas of maximum deposition coinciding with areas of maximum emissions. The dense emission regions in eastern and southern China, northern Thailand, central Japan, S. Korea, Taiwan, and eastern India all have elevated levels of sulfur deposition. Additionally, many of the major urban centers experience high levels of deposition as they are the source of large SO_2 emissions.

A detailed evaluation of model performance is not possible due to a lack of a comprehensive observational data set for acid deposition throughout Asia. However, the model has been compared to various data sets in the region.

Researchers have monitored wet sulfate deposition throughout India, China, Japan, Malaysia, and Thailand. The results of some of these observations are over-laid on model calculated wet deposition in Fig. 2. Comparison of Figs 1 and 2 demonstrates the importance of wet removal processes on Asia's deposition pattern. Overall, wet deposition accounts for nearly 2/3 of the region's sulfur deposition. Hence, reasonable prediction of Asia's wet deposition provides a good source for model evaluation. Observations from the WMO Background Atmospheric Monitoring network (BAPMon) in India during the late 1980s (Ayers and Hara, 1992) range from 0.1 g $S\,m^{-2}\,yr^{-1}$ at Jodpur to 1.6 g $S\,m^{-2}\,yr^{-1}$ at Visakhapatnam. Throughout most of India, observed deposition is below 0.5 g $S\,m^{-2}$ annually and the model provides similar results with deposition between 0.1 and 0.5 g $S\,m^{-2}\,yr^{-1}$ predicted for most of the Indian subcontinent. The model does under-predict deposition along the Bay of Bengal and in the extreme eastern portion of India. Annual sulfur deposition in central Thailand in 1992 was monitored by Granat *et al.* (1996). The observed value (0.2 g $S\,m^{-2}\,yr^{-1}$) and model estimate are within a factor of two of one another. Observed deposition in northern (Beijing and Tianjin) and southwestern (Sichuan and Guizhou provinces) China (in 1982–1985) and Hong Kong (in 1990) have been reported by Ayers and Hara (1992). Observations and model calculations are similar in magnitude at both the northern China and Hong Kong locations. While calculated deposition in southwestern China is significantly higher than observed values, it is important to note that emissions in this

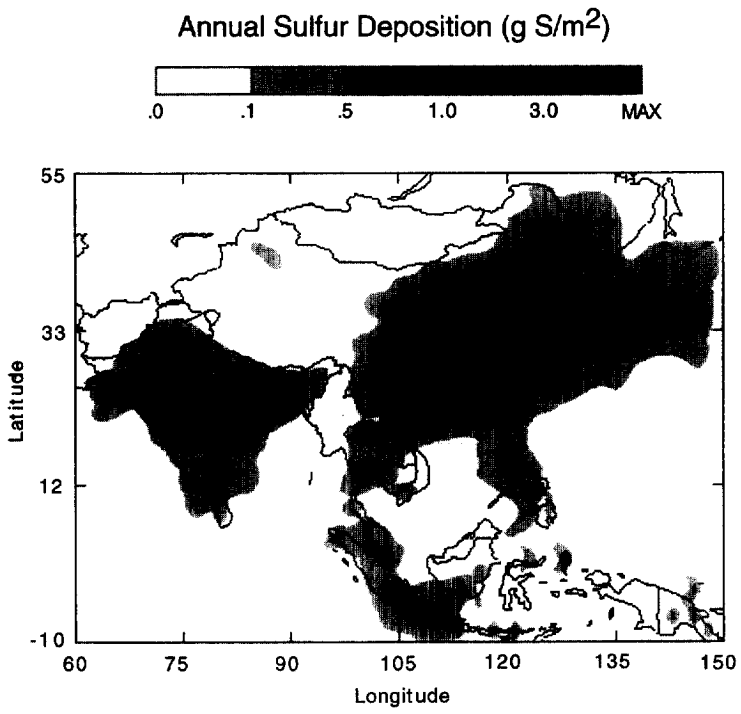

Fig. 1. Calculated annual sulfur deposition for 1990. Units are in g- S m^{-2} yr^{-1}.

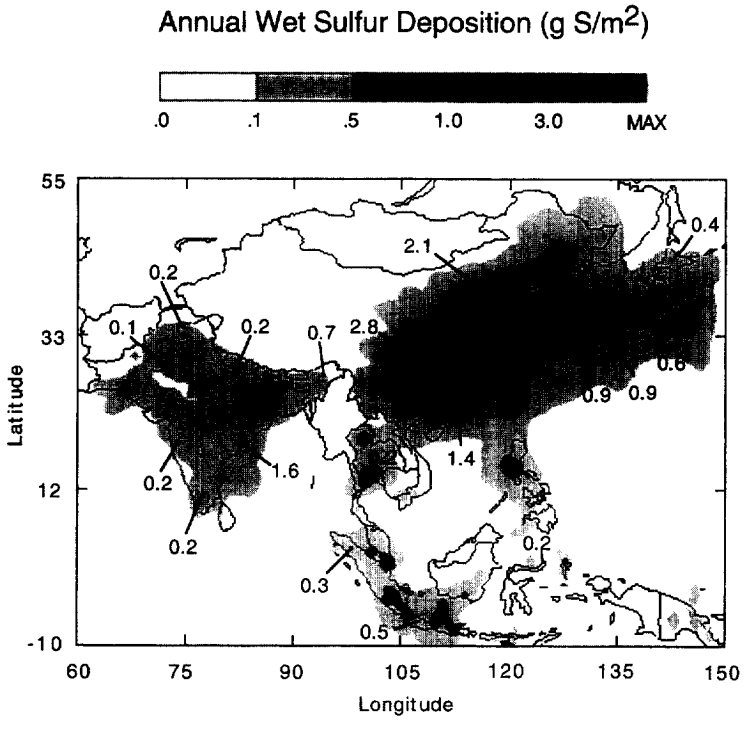

Fig. 2. Calculated annual wet sulfur deposition for 1990 and observed wet sulfur deposition at several locations. Units are in g- S m^{-2} yr^{-1}.

area grew dramatically from 1985 to 1990. The Central Research Institute of Electrical Power Industry (CRIEPI) has an extensive network of monitoring stations in Japan (CRIEPI, 1992) which collected wet sulfate deposition from 1987 to 1990. A more extensive comparison of CRIEPI's observations with model predictions is presented in Arndt and Carmichael (1997). The model under-predicts deposition

on the northern island of Hokkaido (due in part to the absence of Russian emissions in the inventory) but does a reasonable job of calculating deposition on Kyushu, Shikoku, and Honshu. Deposition at several locations in Indonesia in 1989 were reported by Soedomo (1990), and are also shown. Again the model predicts the right magnitude of the deposition, except at Manado, where deposition is under-predicted.

The model calculated deposition values have also been compared with monitoring data from the Japanese EPA (Arndt *et al.*, 1997). While the model has a tendency to under-predict deposition, especially at those monitoring sites located near major urban areas, the model does accurately capture the spatial variability in the sulfur deposition. Furthermore, Murano (1994) found that wet sulfate deposition accounts for over half of the sulfate deposition on Japan. Of the estimated 1 Tg S deposited on Japan each year, 0.62 Tg S is due to wet deposition (Fujita, 1996). The ATMOS model results are consistent with these, predicting that 60% of the total deposition in Japan is due to wet removal processes.

CRIEPI has developed an air quality monitoring network in East Asia with locations in Japan, South Korea, China, and Taiwan (CRIEPI, 1994). This network reflects a widespread variation in geographic locations and emission source magnitudes. The observed annual averaged sulfate concentrations at 16 locations throughout these four countries are presented along with model calculated concentrations in Fig. 3. The model captures both the spatial variation and magnitude of the observed values at most locations. The model significantly under-predicts sulfate concentrations at two locations, Nanjing, China, and Taichung, Taiwan. This may be due to the model failing to capture local emission effects due to the model's coarse resolution. For example, Nanjing is a highly industrialized city with large SO_2 emissions surrounded by agricultural lands.

These preliminary results, while identifying the need to perform a rigorous model evaluation, provide confidence that the model can provide a reasonable representation of Asia's deposition pattern. Hence, the model has been applied to study source–receptor relationships in the region.

3. SOURCE–RECEPTOR RELATIONSHIPS

Based on the 1990 calculations, approximately 70% of the emissions are deposited within the study region with the remainder being transported out of the region. This is similar to estimates for North

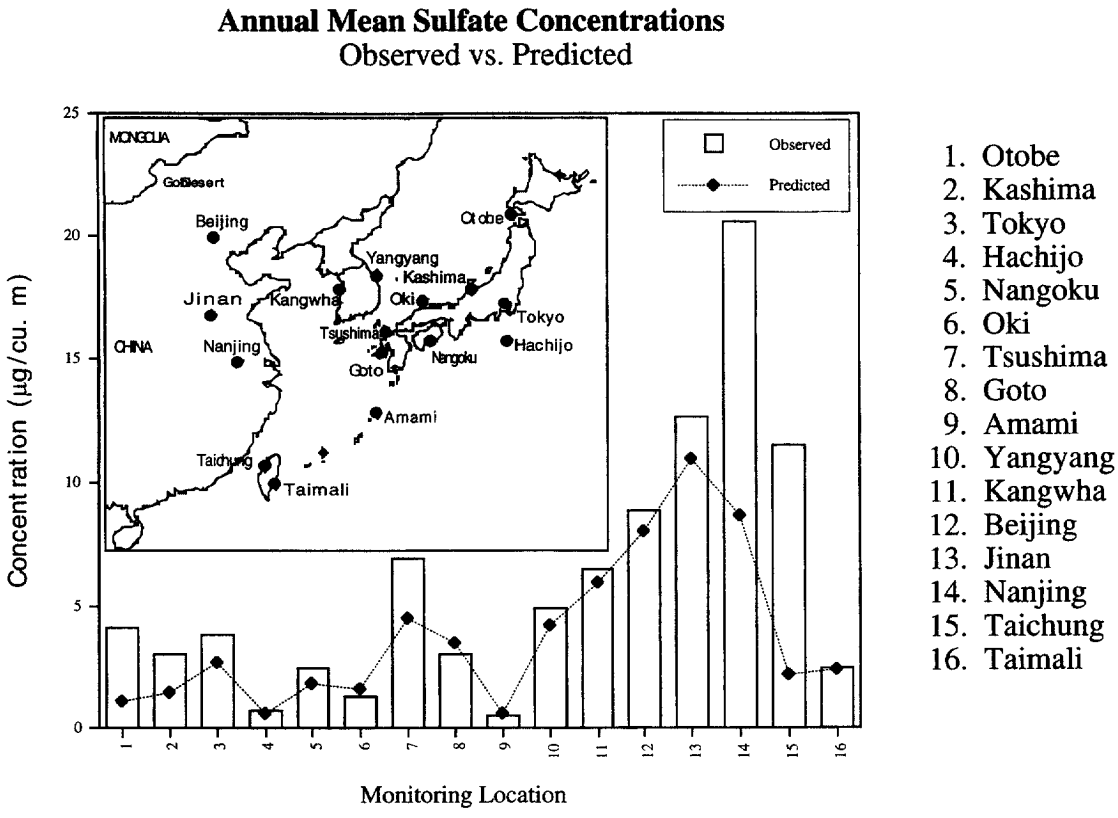

Fig. 3. Observed and calculated annual averaged sulfate concentrations for 16 monitoring sites operated by CRIEPI in east Asia.

American and Western European emissions (Welpdale, 1996). However, the relationship between how much sulfur a particular country emits and the amount of sulfur deposited on that same country varies significantly throughout the region. The largest emitter nations in the region receive approximately half of the magnitude of their emissions in resulting deposition. China's sulfur emissions are nearly 11 Tg S yr^{-1} but only 6.1 Tg of sulfur are deposited on China per year (with the remainder deposited on the seas, other countries, and transported out of the region). Similarly, India emits 2.2 Tg S yr^{-1} while receiving 1.1 Tg S yr^{-1}. In contrast, Nepal and Vietnam receive sulfur deposition exceeding their emissions. The factors causing these differences in emission-to-deposition ratios can be better understood by studying the source–receptor relations between countries.

The ATMOS model allows calculation of source–receptor relationships on individual source, grid cell, sub-nation, or national level. By focusing on seasonal variations in these relationships, we can evaluate how changes in chemical conversions, removal rates, meteorology, and precipitation associated with the different seasons impact the overall deposition patterns in Asia. This information related to source–receptor relationships is of great interest in the region. Below we discuss findings based on our model calculations. These must be viewed with caution, since they are based on only one year of meteorology and the model itself has not been rigorously evaluated. We present the results emphasizing the seasonal variation in these relationships, and identifying regions of potential importance from a long-range transport standpoint.

3.1. *Source–receptor relationships in the Indian sub-continent and Southeast Asia*

Figure 4 presents the major anthropogenic contributors of sulfur deposition on Bangladesh, Nepal, Vietnam, Malaysia, China, and Japan separated into winter (December, January, and February), spring (March, April, and May), summer (June, July, and August), and autumn (September, October, and November). These countries' deposition sources, while not providing a comprehensive picture of Asia's deposition patterns, do provide insight into how deposition patterns in the region vary seasonally.

Bangladesh's deposition reflects what is seen in much of the Indian sub-continent, where domestic and Indian sources dominate the smaller countries' deposition patterns. Bangladesh experiences its maximum deposition during the spring and summer with

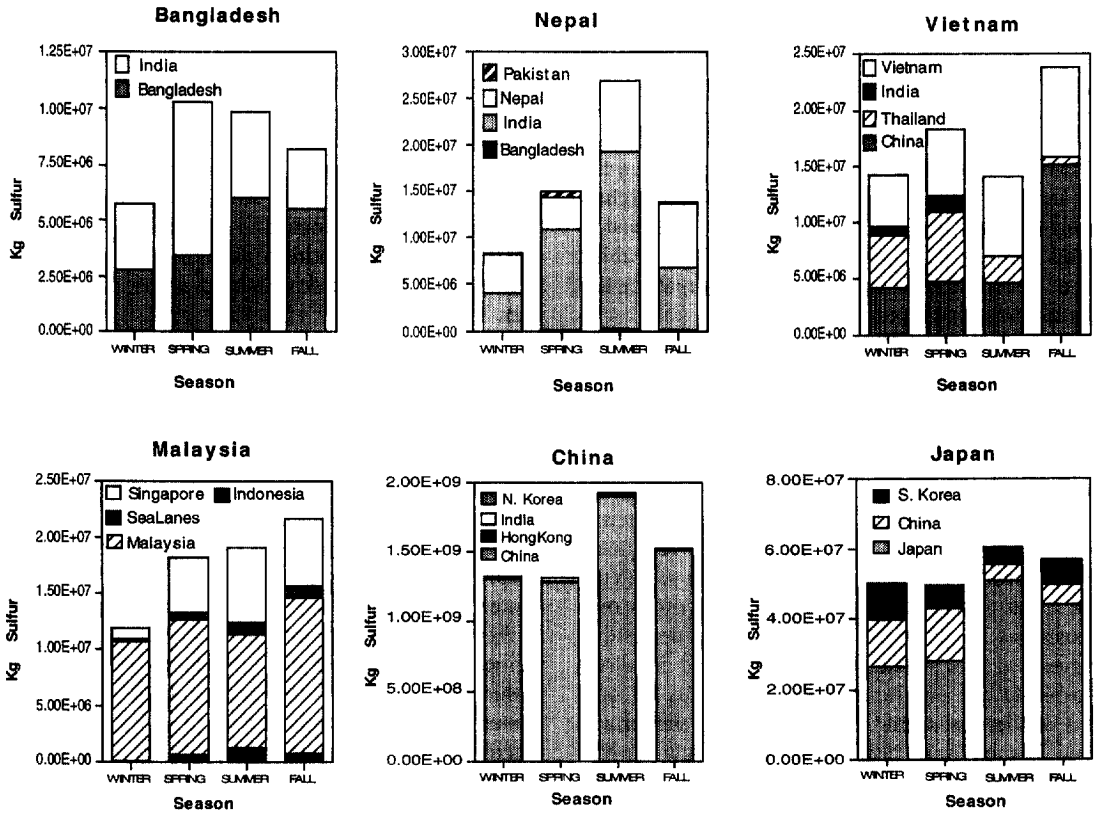

Fig. 4. Calculated seasonal sources and their magnitudes of anthropogenic sulfur deposition for Bangladesh, Nepal, Vietnam, Malaysia, China, and Japan.

minimum deposition occurring during the winter months. The contribution of Indian emissions to Bangladesh's deposition shows clear seasonal variation. Indian emissions are the primary source of sulfur during the spring. During this time, lack of precipitation over India and prevailing westerlies result in significant amounts of India's emissions being deposited over Bangladesh. During the summer months, when India and Bangladesh receive a large portion of their annual precipitation, Indian and Bangladesh emissions are removed closer to their source locations and thus India's contribution decreases while Bangladesh's increases. In contrast to the high deposition experienced during the other three seasons, deposition over Bangladesh during the winter is quite small, 17% of the annual. This is a combination of low precipitation and northeasterly winds carrying most of India's emissions away from Bangladesh.

Nepal's deposition, which is heavily influenced by Indian emissions, exhibits strong seasonal variation with the maximum deposition occurring in the summer and the minimum during the winter months. Minimums in precipitation and wind flows combine to make Nepal's winter deposition its annual minimum. During the spring, westerlies bring sulfur from Indian sources to Nepal. Similarly, summer southwesterlies also carry sulfur from Indian sources to Nepal. The magnitude of this transport is indicated by India being the primary source of Nepal's deposition during the spring and summer. Indian deposition on Nepal is nearly five times higher in the summer than winter despite the fact that due to the summer monsoon in India there is less pollutant available for transboundary transport. Nepal experiences the majority of its precipitation during the summer and fall and hence deposition due to Nepal sources is highest during this time. However, during the fall northeasterlies transport Indian emissions towards the Arabian Sea, as a result Indian contributions to Nepal are less than a third of summer values.

Vietnam's deposition sources vary significantly as a function of season as does its total deposition. In contrast to countries on the Indian sub-continent, Vietnam experiences a minimum in deposition during the summer, when Vietnam is the primary source of deposition. Maximum deposition occurs during the spring and autumn when contributions from external sources are at their peak. Deposition from Vietnamese sources is roughly constant throughout the year (Vietnam never accounts for more than half of its own deposition), however, its other two major deposition sources vary significantly by season and in direct contrast to one another. Thailand accounts for one-third of Vietnam's total deposition in the winter and spring but less than 5% in the fall. During the fall, China is the primary source of deposition in Vietnam, and accounts for over 60% of the total. It is interesting to note that China's contribution in the fall exceeds Vietnam's total deposition in either the winter or the summer. This is due to northeasterly flows over southern China which carry sulfur from this region to Vietnam in combination with heavy precipitation over Vietnam.

Malaysia's deposition increases consistently from winter through to autumn with Malaysia as the primary source of deposition for all seasons. The seasonal contribution of Singapore to Malaysia's deposition also exhibits strong variation, accounting for less than 10% of Malaysia's total deposition during the winter months, over 20% during the spring and autumn, and over 30% during the summer. The low contribution of Singapore emissions in the winter is the result of meteorological and precipitation patterns over the city-nation. During the winter months, the winter monsoon season results in heavy precipitation over Singapore, thus removing most of Singapore's emissions close to their point of origin. In addition, northeasterlies carry the remaining emissions away from the Malaysian Peninsula. During the summer, lower precipitation and southerly flows combine to maximize both Singapore's and Indonesia's contribution to Malaysia's deposition. In contrast, deposition from Malaysian sources is at a minimum during this time. During the summer Indonesia contributes 6% of Malaysia's deposition, however, during the winter Indonesia accounts for only 1% of Malaysia's deposition.

3.2. *Source–receptor relationships in East Asia*

The interaction between emission sources and their resulting deposition patterns in eastern Asia is of particular interest since this region has the highest sulfur emissions in Asia (Hara, 1994; Huang *et al.*, 1995; Murano, 1993). Transport patterns show significant variation throughout the year with continental outflow during the winter and spring and onshore flows during the summer and fall. These flow fields combine with precipitation patterns to significantly alter the source-receptor relationships during the year.

During the winter and spring months, low precipitation levels over the northern half of China result in the sulfur emitted in this area experiencing very little wet removal. As a result, this sulfur is transported farther from its point of origin then similar emissions released during the summer months when precipitation is higher and the pollutant is more likely to be washed out closer to its point of origin. Additionally, the presence of strong prevailing westerlies during the winter and spring cause the pollutant to be carried farther from its source location. The net effect is that Chinese emissions are more likely to be deposited within China during the summer than during the winter. These factors can be quantified by comparing where Chinese emissions are deposited throughout the year. China's deposition on its own soils is over 50% higher during the summer as compared to winter, despite the fact that due to the domestic heating cycle, sulfur emissions in the winter months are 16%

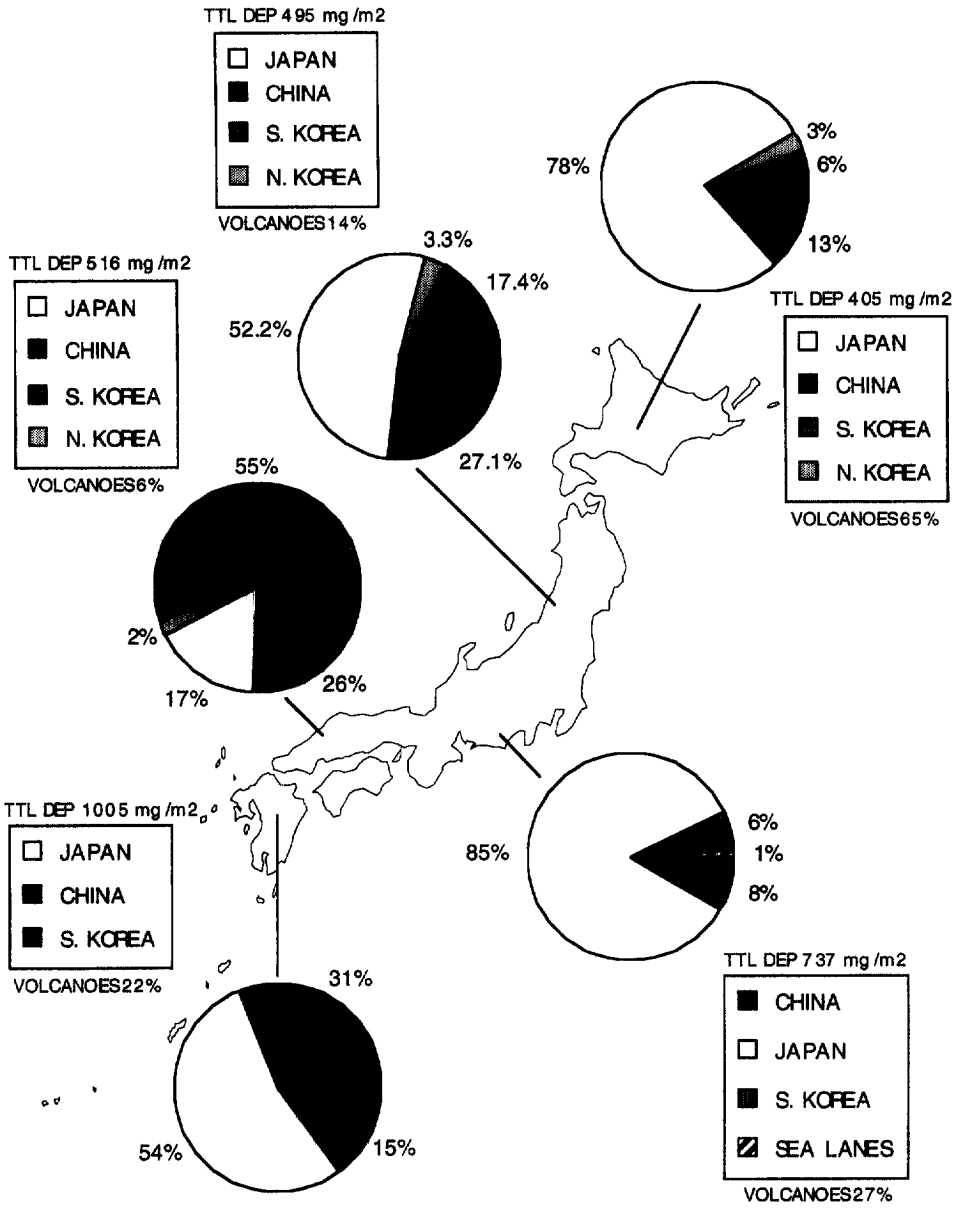

Fig. 5. Calculated annual sulfur deposition sources and their fraction contributions for five locations in Japan. RAINS-ASIA software was used in the analysis.

higher than during the summer months. Rodhe and Granat (1984) reported a similar phenomenon for European sources.

Increased transport of sulfur away from the Chinese mainland during the winter and spring and the elevated precipitation levels along the western coast of Japan during the winter result in deposition on Japan due to Chinese sources being 2.5 times higher for the winter and spring (27.9×10^6 kg S) than for summer and autumn (10.9×10^6 kg S). The same phenomenon is found when comparing winter and summer deposition from S. Korea. Deposition on Japan due to S. Korean sources is over two times higher during the winter (10.5×10^6 kg S) than it is in the summer (4.6×10^6 kg S). In contrast, during the summer, when on-shore winds predominate, over 5.0×10^5 kg of sulfur from Japanese sources reach South Korea. However, during the winter months, only 1.3×10^5 kg S reach S. Korea from Japan.

The sources of Japan's annual deposition vary significantly as a function of location within Japan. Figure 5 presents the primary sources of deposition at five locations within Japan. The variation in Japan's deposition is driven by volcanic, Chinese, S. Korean, and Japanese emissions. (While North Korea is not predicted to be a substantial factor in Japan's

Table 1. Wet removal rates used in: the ATMOS model, by Huang *et al.* (1995), and by Ichikawa and Fujita (1995) (Units are in s^{-1})

SO_2 removal rate constant (s^{-1})	20 South–20 North	20 North–30 North	30 North–55 North
ATMOS Model	1.1×10^{-4} P	$\left\{8.3 \times 10^{-5} + 2.7 \times 10^{-5} \sin\left[\frac{2*\pi*(\text{TOY} - 80)}{365}\right]\right\}$*P	
Ichikawa and Fujita, 1995	$2.8 \times 10^{-5} \times P$		
Huang *et al.* (1995)	$(4.3 + 0.78 \ln C_{SO_2}) + \left[(0.14 - 0.019 \ln C_{SO_2})*\frac{P}{3600}\right]$		
Sulfate removal rate constant (s^{-1})	Constant for all latitudes		
ATMOS model	$4.0 \times 10^{-4} \times P^{0.7}$ {for in-cloud scavenging} $6.0 \times 10^{-5} \times P$ {for below-cloud scavenging}		
Ichikawa and Fujita (1995)	$2.8 \times 10^{-5} \times P$		
Huang *et al.* (1995)	$9.2 \times 10^{-5} \times P^{0.83}$		

Note: Where TOY represents the Julian day of the year, P is the precipitation rate in mm/h, and C_{SO_2} is the concentration of SO_2 in $\mu g\,m^{-3}$.

deposition, its influence is unclear due to high uncertainty in the magnitude of that nation's SO_2 emissions (Von Hipple, 1996).) China's influence is shown to be most evident along the western coast of Honshu and the island of Kyushu, with the smallest impact on Hokkaido in the north. While Japanese sources are the primary source of anthropogenic deposition throughout Japan, their contribution is highest in eastern Japan, accounting for over 60% of the total deposition. The influence of volcanic emissions on Japan's deposition pattern is also evident, particularly on Hokkaido. Here large emissions from volcanic sources and small influences from anthropogenic sources result in volcanoes accounting for over 60% of the total deposition. (Note: Russian emissions are not included in the analysis; these emissions would have their largest impact on Japan's northern regions.) It is interesting to note that although Japan's largest volcanic source, Mt. Sakurajima, is located on Kyushu, volcanic sulfur accounts for only 22% of the deposition at this location. This is due to large amounts of anthropogenic deposition from Japan, S. Korea, and China.

Overall, external emissions account for ~17% of the anthropogenic sulfur deposition in Japan. These results can be compared with other model estimates of source–receptor relationships in East Asia by Huang *et al.* (1995) and Ichikawa and Fujita (1995). Their calculated contribution of Chinese sources to Japan's deposition present markedly different estimates of the role that long-range transport plays in Japan's overall deposition. Huang *et al.* (1995) estimated that China accounts for only 3.5% of Japan's total sulfur deposition. They found that over 93% of the sulfur deposited within Japan was from either Japanese anthropogenic or volcanic sources and that only 26% of total sulfur deposition on Japan was due to external sulfur sources. In contrast, Ichikawa and Fujita (1995) estimated China to be a major source of wet sulfate deposition in Japan, accounting for one-half of the anthropogenic deposition.

We have begun to study the reasons for the differences between these findings. Variations are largely due to differences in removal rates and chemical conversion rates in different models. Ichikawa and Fujita (1995) used a wet SO_2 removal rate of 2.8×10^{-5}, the ATMOS model rate used 1.1×10^{-4}, and Huang *et al.*, (1995) used a significantly higher rate of 1.7×10^{-3} (assuming a SO_2 concentration of $10\,\mu g\,m^{-3}$) (see Table 1). For wet sulfate removal, Ichikawa and Fujita (1995) again used a rate of 2.8×10^{-5}, Huang *et al.* (1995) used 9.2×10^{-5}, and the ATMOS model used in cloud and below cloud rates of 7.0×10^{-4} and 6.0×10^{-5}, respectively. (All rates are based on a precipitation rate of $1\,mm\,h^{-1}$ and have units of s^{-1}.) Low removal rates result in greater transport away from source locations and thus higher transboundary pollution. For example, the use of a low removal rate (such as is the case in Ichikawa and Fujita (1995)) results in a greater transport of sulfur away from source locations, and thus a larger contribution to Japan's deposition from emissions in China. There is clearly a need to conduct further model comparisons and fundamental studies to better determine the most suitable parameters for use in modeling studies in Asia. Thus, while the seasonal variations in source–receptor relationships qualitatively capture Asia's deposition picture, caution must be used when viewing the predicted magnitudes of individual countries' contributions.

4. SUMMARY

Preliminary source–receptor relationships have been presented for Asia. These relationships vary significantly throughout the region. Deposition on an

individual country can vary markedly throughout the year, both in magnitude and in the sources of deposition. Using the ATMOS model we developed seasonal source–receptor relationships for several countries in the region. By comparing the seasonal source–receptor relationships for several countries in Asia we have developed a picture of the role that meteorology, precipitation, and neighboring countries play in various countries' overall deposition. These relationships highlight the importance of seasonality in Asia's deposition patterns. The presence or lack of precipitation over a specific country cannot only alter its own deposition but also, through transboundary flow, that of neighboring or even distant countries. Further, as we saw with Vietnam and Nepal, many countries in the region receive greater deposition from external sources than from domestic sources.

Comparison of deposition sources at locations within Japan emphasizes how deposition sources vary within a receptor country. This illustrates the impact of proximity of emission sources, meteorology and precipitation patterns within the receptor country, and geographical features within the receptor country.

The expansion of Asia's economies is causing rapid degradation in the region's air quality. Furthermore, economic growth in one country can cause, through transboundary transport of air pollutants, degradation of other countries' ecosystems. Transboundary issues are already a serious concern in east Asia and can be expected to become an issue in other areas of the region in the coming years. By developing an understanding of source–receptor relationships in the region, we can anticipate how future emissions will affect the region's environment.

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