

## SULFUR DEPOSITION IN ASIA: SEASONAL BEHAVIOR AND CONTRIBUTIONS FROM VARIOUS ENERGY SECTORS

SARATH K. GUTTIKUNDA<sup>1</sup>, NARISARA THONGBOONCHOO<sup>1</sup>, RICHARD L. ARNDT<sup>1</sup>, GIUSEPPE CALORI<sup>1</sup>, GREGORY R. CARMICHAEL<sup>1\*</sup> and DAVID G. STREETS<sup>2</sup>

<sup>1</sup> Center for Global and Regional Environmental Research, Department of Chemical and Biochemical Engineering, The University of Iowa, Iowa City, Iowa 52242, U.S.A.; <sup>2</sup> Decision and Information Sciences Division, Argonne National Laboratory, 9700, South Cass Avenue, Argonne, Illinois 60439, U.S.A.

(\* author for correspondence, e-mail: gcarmich@engineering.uiowa.edu)

(Received 26 October 1999; accepted 4 October 2000)

**Abstract.** Sulfur transport and deposition in Asia, on an annual and seasonal basis, is analyzed using the ATMOS model. Calculations are performed for two complete years (1990 and 1995). Deposition amounts in excess of  $0.5 \text{ g S m}^{-2} \text{ yr}^{-1}$  are estimated for large regions in Asia, with values as high as  $10 \text{ g S m}^{-2} \text{ yr}^{-1}$  in southeastern China. Annual averaged  $\text{SO}_2$  concentrations in excess of  $20 \mu\text{g SO}_2 \text{ m}^{-3}$  are calculated for many urban and suburban areas of eastern China and S. Korea, with an average of  $5 \mu\text{g SO}_2 \text{ m}^{-3}$  over most of the emitter regions. Sulfur deposition by major source categories is also studied. Southeast Asia (Indonesia, Malaysia, Philippines, Singapore) receives  $\sim 25\%$  of its sulfur deposition from shipping activities. Sulfur deposition from bio-fuel burning is significant for most of the underdeveloped regions in Asia. Volcanoes are a major source of sulfur emissions in the Pacific Ocean, Papua New Guinea, Philippines and Southern Japan. Sulfur deposition is shown to vary significantly throughout the year. The monsoons are found to be the largest factor controlling sulfur transport and deposition in the Indian sub-continent and Southeast Asia. India receives over 35% of its total deposition during the summer months. In East Asia, sulfur deposition is estimated to be 10% higher during summer and fall than winter and spring. Model results are compared with observations from a number of monitoring networks in Asia and are found to be generally consistent with the limited observations.

**Keywords:** asia, annual sulfur deposition, long range transport, seasonal sulfur deposition, sectoral analysis

### 1. Introduction

Due to its burgeoning population, high growth rate of industrialization, intensification of agricultural practices, and continued transport sector expansion, Asia has emerged as an economic force. This rapid economic growth has resulted in significant growth in the Asia's energy demand (Akimoto and Narita, 1994). In 1987, coal accounted for 76, 54, and 35% of China, India, and South Korea's primary energy consumption, respectively (Shrestha and Bhattacharya, 1991) and still remains the primary choice of energy demand for most of the Asian countries. This growth has not come without environmental consequences. Over the last two decades, China's



*Water, Air, and Soil Pollution* **131**: 383–406, 2001.

© 2001 Kluwer Academic Publishers. Printed in the Netherlands.

sulfur emissions have grown by more than a factor of three (Streets *et al.*, 1995; Akimoto and Narita, 1994). As the sulfur emissions have increased, the transport and fate of sulfur in Asia has become a greater environmental issue (Carmichael and Arndt, 1995; Arndt *et al.*, 1997; Sharma *et al.*, 1995). Furthermore, as countries receive growing amounts of sulfur from neighboring and even distant countries (Ichikawa and Fujita, 1995; Arndt *et al.*, 1998), awareness of transboundary movement of sulfur and other pollutants means that the domestic energy needs have become regional environmental concerns (Streets *et al.*, 1999). The management of sulfur emissions in Asia requires an understanding of the contribution to sulfur deposition and concentrations from different energy sectors, viz., domestic fuel usage, transport, industry, power plants, etc., and information on the mechanisms and nature of transboundary pollution in Asia. Long-range transport models play a critical role in studying these sulfur transport mechanisms and characteristics. Long-range transport models also play a key role in environmental management, in that they link emissions to deposition and thus are used to evaluate the impact of energy policies and trends on future sulfur deposition levels.

This work extends the study of Arndt *et al.* (1998), by constructing and calculating sulfur deposition in Asia with an emphasis on the seasonal variation of sulfur transport and deposition, and the contributions from the various energy emission sectors. The analysis utilizes the same long-range transport model, but makes use of different meteorological inputs. In this study, meteorological data was based on the gridded NCEP reanalysis fields (Kalnay *et al.*, 1996), instead of the interpolated data from radiosondes used in Arndt *et al.* (1998). In addition, the dry deposition and scavenging parameters have been modified based on recent reviews (Xu and Carmichael, 1998). Furthermore, the analysis is based on simulation of two complete years, 1990 and 1995. Finally, the sulfur emissions inventory for Asia has also been updated (Streets *et al.*, 1998, 1999, 2000).

## 2. Model: Domain and Inputs

Sulfur transport and deposition was studied using the ATMOS regional-scale trajectory model. This three-dimensional, multiple-layer lagrangian model is described in detail elsewhere (Arndt *et al.*, 1997; Arndt, 1997). Using 1990 and 1995 sulfur emission inventories that were compiled as part of the RAINS-ASIA project (Streets *et al.*, 1995), and 1990 and 1994 meteorological fields, sulfur deposition, and sulfur dioxide (SO<sub>2</sub>) and sulfate concentrations were calculated with a 1° × 1° spatial and a one hour temporal resolution. The simulation domain included the whole of Asia, excluding Afghanistan and countries of the former Soviet Union.

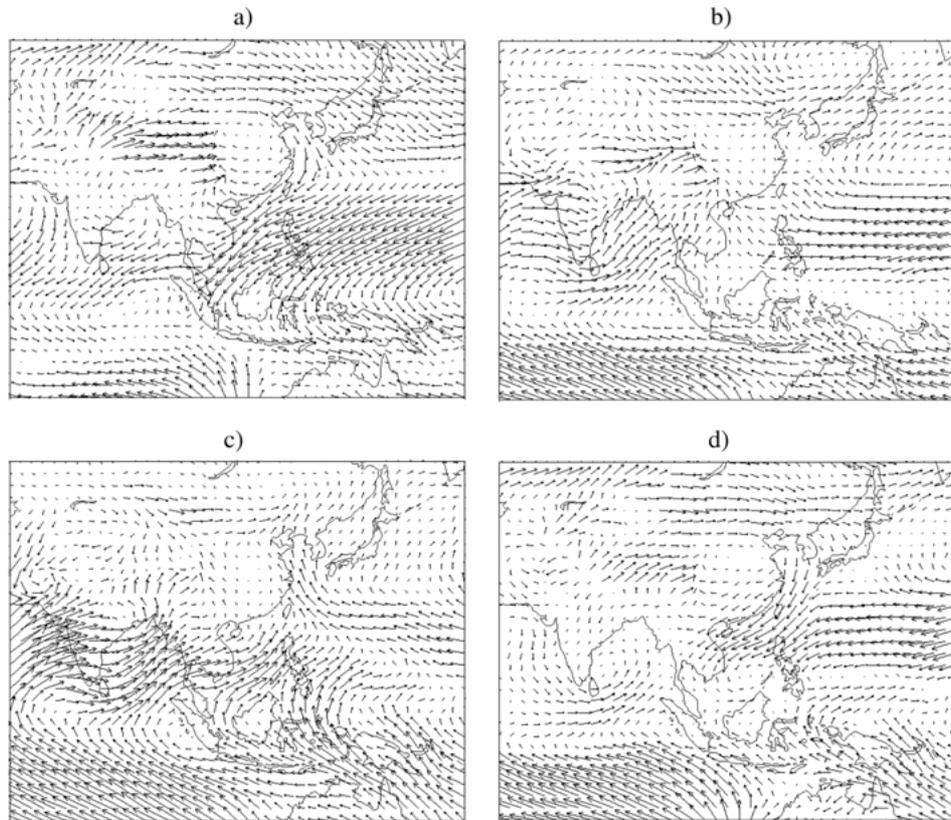


Figure 1. Seasonal averaged flow fields for 1990 in Asia at 0.1 km above ground level, obtained from NCEP/NCAR reanalysis data (Kalnay *et al.*, 1996): (a) Winter (DJF) (b) Spring (MAM) (c) Summer (JJA) (d) Fall (SON).

## 2.1. METEOROLOGY

The meteorological fields used were 6 hr time averaged horizontal winds, precipitation and temperature from the NCEP/NCAR reanalysis project (Kalnay *et al.*, 1996). Figure 1 presents the 1990 seasonal flow conditions in Asia. During the winter months, offshore winds dominate as a result of high pressure centered over Mongolia, in contrast to onshore winds during the summer months when low pressure is centered over the Indian sub-continent. Also, during the fall and the winter months, winds are generally northeasterly over the Indian sub-continent, while southwesterly winds predominate in the spring and the summer months. During winter and spring, persistent westerly winds over northern China result in continental outflow moving across the Korean Peninsula and Japan. During the same period, northeasterly winds blow over southern China and Southeast Asia, in contrast to onshore southwesterly winds during the summer and fall.

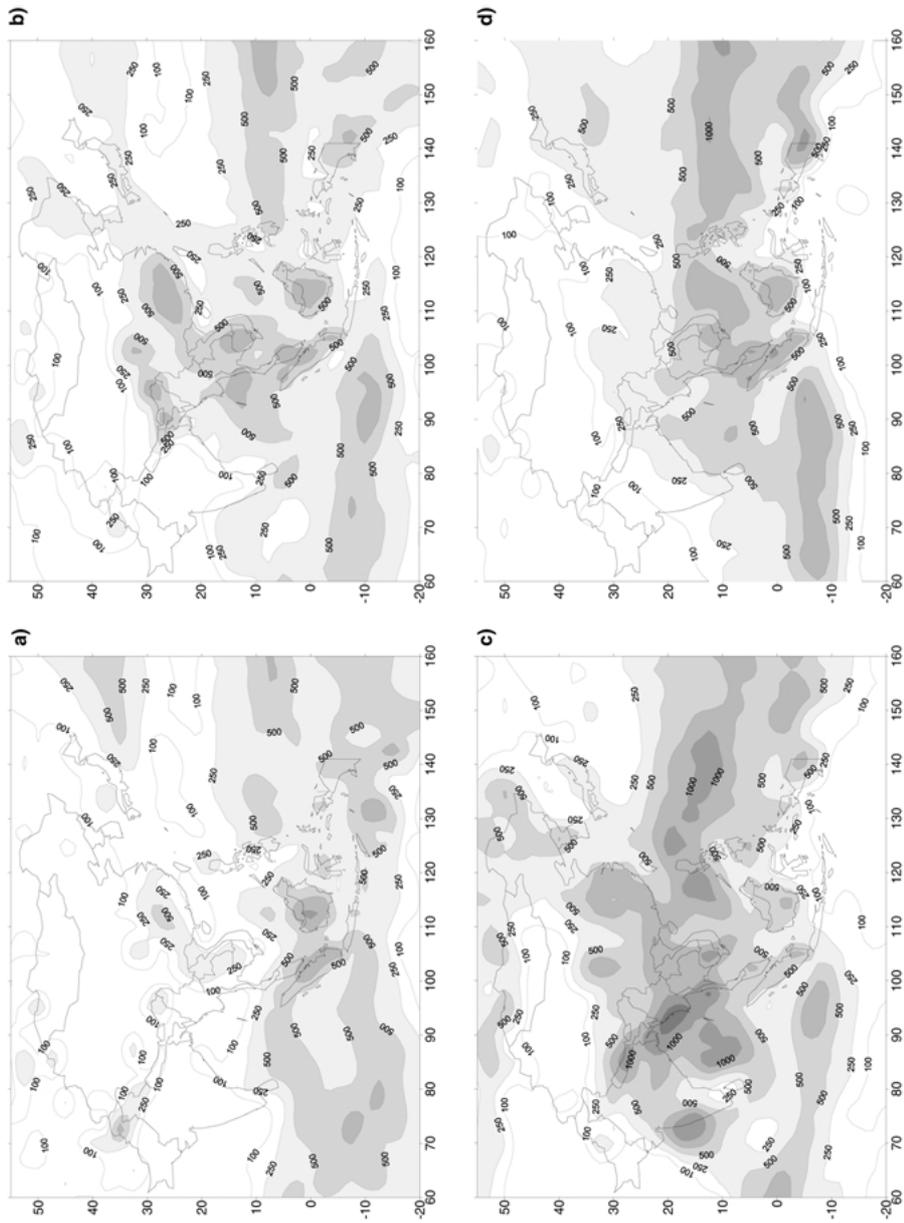


Figure 2. Accumulated seasonal precipitation for 1990 in Asia, in mm, obtained from NCEP/NCAR reanalysis data (Kalnay *et al.*, 1996): (a) Winter (DJF) (b) Spring (MAM) (c) Summer (JJA) (d) Fall (SON).

Asia experiences significant variations in its intra-annual precipitation patterns. Southeast Asia and the Indian sub-continent's annual precipitation amounts are dominated by the monsoon seasons. China, the Koreas, and Japan also experience wet and dry seasons. Accumulated precipitation fields (in mm) for 1990 are presented in Figure 2 (Source: Kalnay, *et al.*, 1996). During the winter months, many locations in the middle and upper latitudes experience little precipitation. However, northern India, southeastern China, and Southeast Asia receive large amounts of precipitation during this time of the year. Precipitation patterns shift northward during the summer months as the summer monsoon brings heavy rains to Indo-China. Indian sub-continent receives most of its precipitation during the summer and fall months. The mid latitude regions ( $0^{\circ}$ – $30^{\circ}$ N) receive heavy precipitation during the summer; however, the Yellow and Japan Seas do not experience significantly higher precipitation during the summer as compared to winter.

## 2.2. EMISSIONS INVENTORY

Sulfur emissions from both surface and elevated sources were included in the analysis. This study utilized the  $1^{\circ} \times 1^{\circ}$  SO<sub>2</sub> emissions inventory compiled as part of the RAINS-ASIA project (Streets *et al.*, 1995), including both natural and anthropogenic sources. Natural sources consist of the active volcanoes in the region (Fujita, 1992; Spiro *et al.*, 1992) and the anthropogenic sources consist of emissions from regional shipping activities, regional domestic fuel usage, and elevated Large Point Sources (LPS's). Emissions from shipping activities include emissions from regional shipping lanes and port activities (Streets *et al.*, 1997, 2000). Area sources are separated into the 94 RAINS-ASIA region designations (Foell *et al.*, 1995), and include emissions from industrial, domestic, and transportation sectors. The LPS's are electrical power plants and industrial sources, which exceed 500 MW.

Table I presents the 1990 and 1995 annual sulfur emissions from the anthropogenic and the volcanic activities in Asia in  $10^6$  kg SO<sub>2</sub>. Annual emissions are estimated to have increased by  $\sim 15\%$  from 1990 to 1995. The largest sources of anthropogenic emissions in Asia are China and India, which accounted for  $\sim 60$  and  $\sim 13\%$  in 1990 and for  $\sim 61$  and  $\sim 14\%$  in 1995, respectively. LPS's accounted for  $\sim 16\%$  of the anthropogenic emissions and volcanoes for  $\sim 6\%$  of the total sulfur emissions in Asia during this period. While the shipping activities accounted for  $\sim 2\%$  of the total emissions in Asia, they accounted for  $\sim 7\%$  of Southeast Asia's sulfur emissions. Also presented are the 1987 estimate of SO<sub>2</sub> emissions for Asia (Akimoto and Narita, 1994). The 1990 estimates are  $\sim 15\%$  higher than those for Akimoto *et al.* Several factors contribute to these differences including the rapid growth in SO<sub>2</sub> emissions between 1987 and 1990 and the inclusion of bio-fuel sources, viz., field residue, animal waste and wood burning (Streets *et al.*, 1998). Russian emissions were not included in this analysis, nor were natural emissions associated with Di-Methyl Sulfide (DMS).

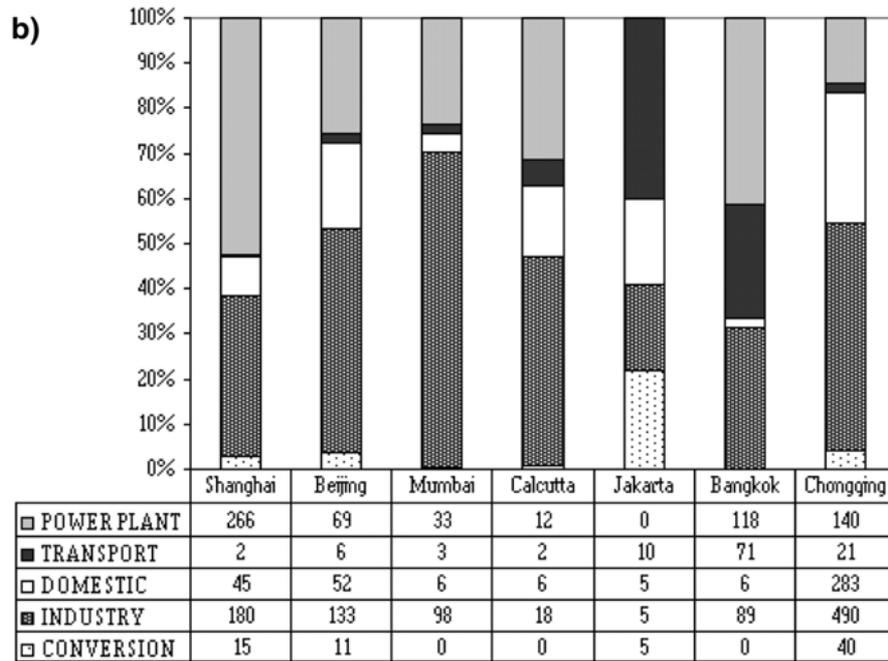
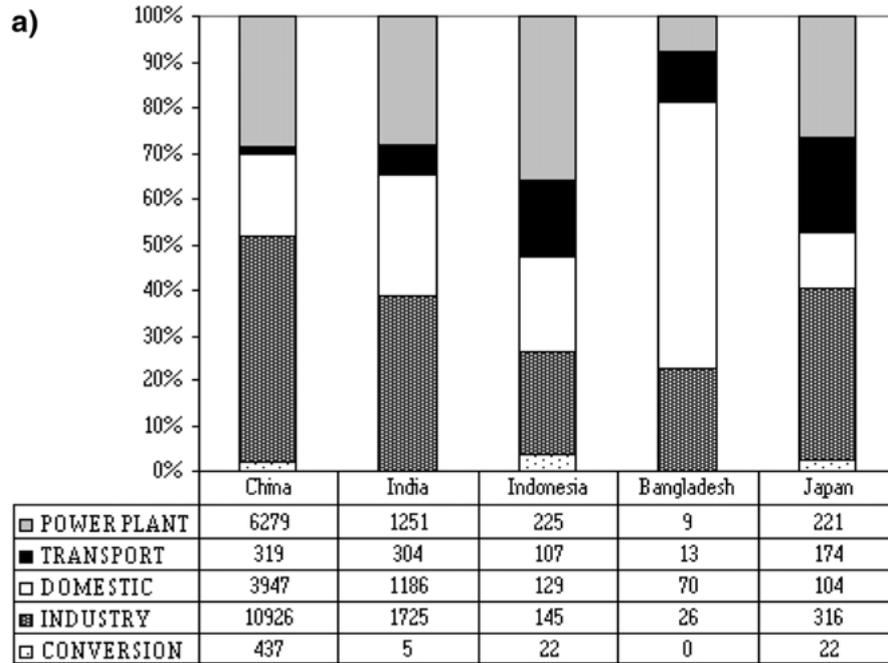


Figure 3. Energy sector wise emissions for 1990 in  $10^9$  g  $\text{SO}_2$   $\text{yr}^{-1}$ : (a) Countries; (b) Megacities.

TABLE I  
SO<sub>2</sub> emissions in Asia for 1990, 1995, and 1987 in 10<sup>9</sup> SO<sub>2</sub> yr<sup>-1</sup>

Country	1990			1995			1987 <sup>a</sup>
	Area sources	LPS's Total	Total	Area sources	LPS's	Total	Total
Bangladesh	100		100	140		140	49
Bhutan	2		2	2		2	
Brunei	6		6	9	9		
Burma	19		19	21		21	29
Cambodia	23		23	26		26	3
China	18865	3360	22225	20942	4756	25698	19990
Hong Kong	45	108	153	40	115	155	150
India	3238	1199	4437	3734	1875	5609	3070
Indonesia	473	89	562	606	78	684	485
Japan	1011	17	1028	939	16	955	1140
Laos	3		3	5		5	2
Malaysia	199	57	256	169	98	267	263
Mongolia	81		81	75		75	101
Nepal	17		17	33		33	30
North Korea	353		353	257		257	333
Pakistan	685		685	908		908	381
Philippines	403	9	412	466	10	476	370
Singapore	191		191	230		230	155
South Korea	1608	98	1706	1174	45	1219	1290
Sri Lanka	26		26	38		38	28
Taiwan	483	22	505	439	15	454	605
Thailand	495	469	964	621	628	1249	612
Vietnam	113		113	133		133	39
Sea Lanes	607		607	817		817	
Volcanoes			2246			2246	
Total	31292	5428	36720	34070	7636	41706	29121

<sup>a</sup> Akimoto *et al.*, 1994.

Figure 3a, shows a comparative distribution of sulfur emissions from China, India, Indonesia, Bangladesh and Japan in 1990. Emissions from the Industrial sector dominated in China and Japan and accounted for ~50 and ~35%, respectively. Sulfur emissions from domestic fuel burning, like dung, agricultural and animal waste accounted for ~60% of Bangladesh's annual emissions. Contributions from the power sector varied from 25 to 50%. A similar analysis for seven of the Asian

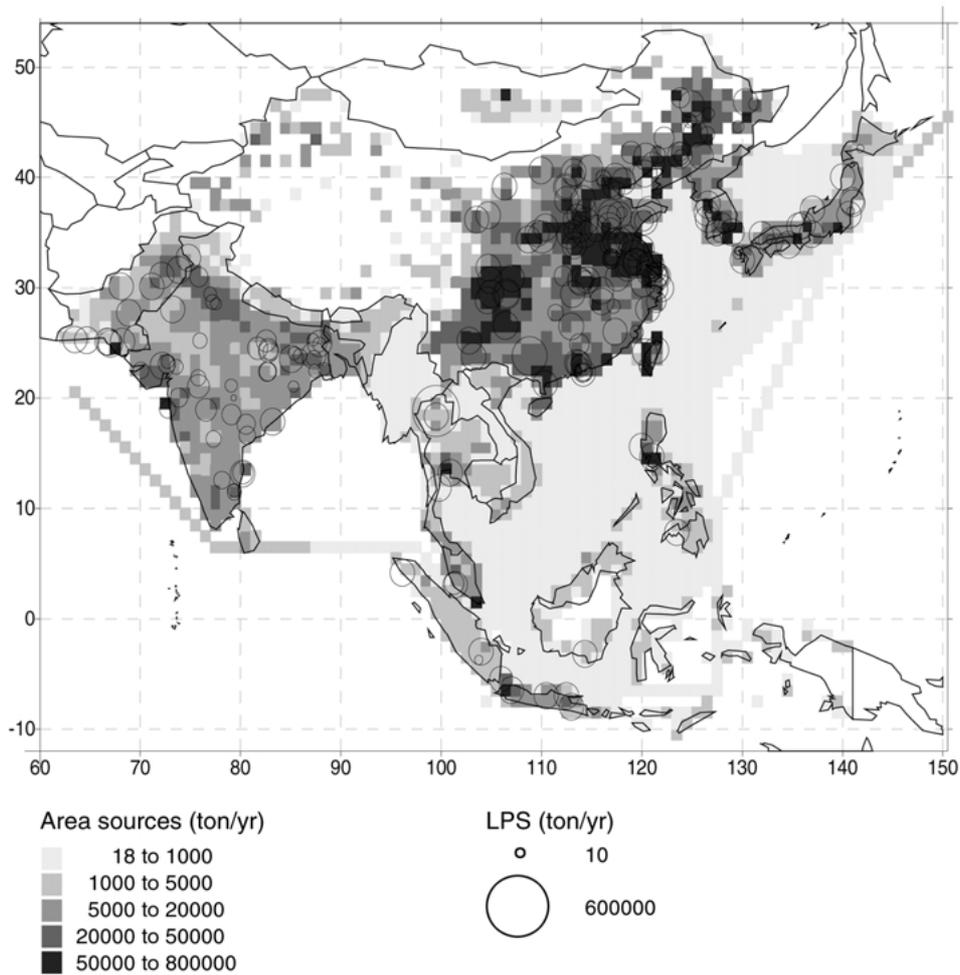


Figure 4. Estimated annual sulfur emissions for 1990 in  $10^{12}$  g  $\text{SO}_2$   $\text{yr}^{-1}$ .

mega cities (Figure 3b) shows major contributions coming from the industrial and power sectors, reflecting the economic activity of the cities. Mumbai, has the largest contribution from the industrial sector,  $\sim 70\%$ . Contribution from domestic sources varies dramatically from a few percent in Bangkok to  $\sim 30\%$  in Chongqing, reflecting a considerable variation in the importance of bio-fuel usage in Asia. Also represented in the figure are the amounts of sulfur emissions in  $10^9$  g  $\text{SO}_2$   $\text{yr}^{-1}$  from each of the source categories.

The spatial distribution of the gridded emissions is presented in Figure 4. The spatial patterns do not change markedly from 1990 to 1995. Large emission densities are seen in and around the major urban areas of Asia. Grid cells containing Karachi, Mumbai, Calcutta, Bangkok, Singapore, Jakarta, Hong Kong, Beijing,

Taipei, Manila, Seoul, and Tokyo all have emissions in excess of  $0.1 \times 10^{12}$  g SO<sub>2</sub> per grid. Additionally, areas of heavy industrialization or electrical generation also have elevated emissions. The Ganges river basin in India, south central and the eastern coastal regions of China, northern Thailand, Taiwan, and most of South Korea, have large sulfur emissions. The contribution from the major shipping lanes, running between Middle East and Japan, and the small-scale cargo shipping lanes between the Southeast Asian nations are clearly shown (Streets *et al.*, 2000). Throughout the islands of Southeast Asia, the Philippines, and Japan, volcanoes are a significant point source of sulfur. Most notable is Mt. Sakurajima on the island of Kyushu, with an estimated emission of  $\sim 0.5 \times 10^{12}$  g SO<sub>2</sub> yr<sup>-1</sup> (Fujita, 1992).

For modeling purposes, area emissions were located at the center of the grid and were released into the well-mixed layer during the day and the surface layer at night. The elevated sources (LPS) were emitted into the well-mixed layer during the day and above the surface layer at night. Volcanic emissions were emitted above the mixed layer at all times. Since elevated sources are individual facilities (e.g., electrical power plants) or volcanoes, their emissions are designated at their actual location (on the hundredth of a degree resolution).

### 3. Results and Discussion

#### 3.1. ANNUAL SULFUR DEPOSITION AND CONCENTRATIONS

The annual-averaged surface concentrations of SO<sub>2</sub> and sulfate, and the dry and wet deposition of SO<sub>2</sub> and sulfate, were calculated using the ATMOS model for 1990 and 1994. The model calculated annual total deposition in g S m<sup>-2</sup> yr<sup>-1</sup> is presented in Figure 5a. The average deposition fields presented include contributions from the anthropogenic sources, regional shipping activities and volcanoes. The high sulfur deposition regions follow closely the spatial distribution and the density of the emissions. For example, the dense emission regions in the eastern and southern China, S. Korea, northern Thailand, Taiwan, and central Japan, all show sulfur deposition in excess of 0.5 g S m<sup>-2</sup> yr<sup>-1</sup>. Southern Malaysia, Singapore, and parts of Indonesia also show elevated sulfur deposition. The highest calculated annual deposition ( $\sim 10.5$  g S m<sup>-2</sup> yr<sup>-1</sup>) occurs around the city of Chongqing in the Sichuan province. Sulfur deposition in excess of 1.0 g S m<sup>-2</sup> yr<sup>-1</sup> is also found around the major urban centers in Asia.

Figure 5b presents the model predicted annual averaged SO<sub>2</sub> surface concentrations. Similar to the deposition patterns, major industrial and urban centers show the highest SO<sub>2</sub> concentrations. Most of the Indian sub-continent, south central and eastern China, South Korea, southern Japan, Taiwan, and the northern Thailand, all show annual average SO<sub>2</sub> concentrations as high as 5 μg m<sup>-3</sup>. The major cities, Bangkok, Singapore, Hong Kong, Seoul, etc., are clearly depicted by their high SO<sub>2</sub> concentrations. It is important to note that the sulfur deposition and concentrations reflect the average value for the entire grid cell, and within the urban part

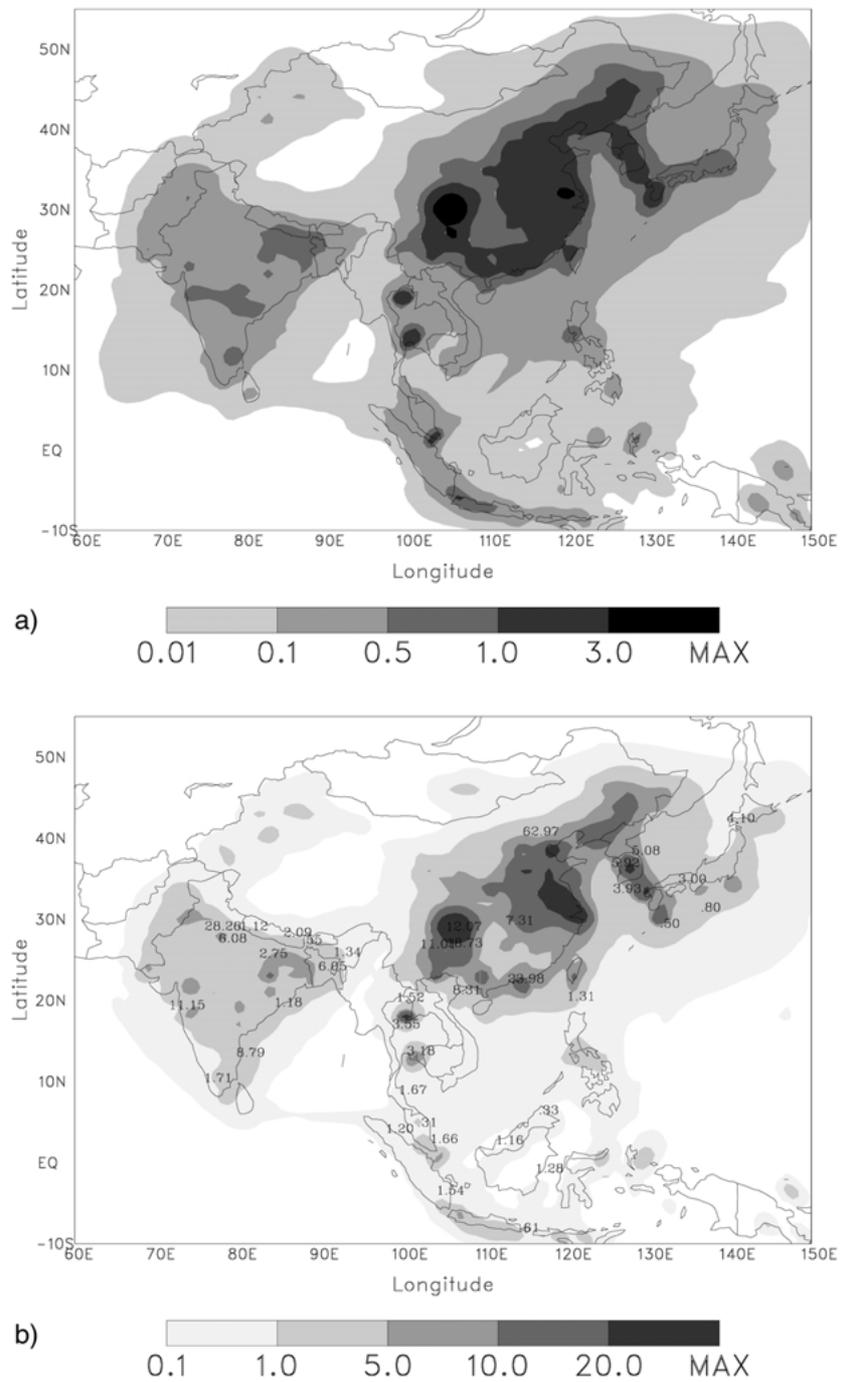


Figure 5. (a) Calculated annual total sulfur deposition for 1990 in  $\text{g S m}^{-2} \text{ yr}^{-1}$ . (b) Calculated annual averaged  $\text{SO}_2$  concentrations for 1990 overlaid by measurements obtained from various monitoring networks in Asia (Carmichael, *et al.*, 1995b) in  $\mu\text{g SO}_2 \text{ m}^{-3}$ .

of the grid they are expected to be significantly higher than these average values (Calori and Carmichael, 1999).

From the annual sulfur budget calculations, over one-third of sulfur emitted from anthropogenic sources was transported out of the region. This is similar to transport trends over North America and Western Europe (Welpdale, 1996). Of the remaining two-third's deposited in the region, 60% was removed by wet processes and the remainder by dry removal processes. For the case of SO<sub>2</sub>, the domain-integrated total deposition due to dry and wet removal mechanisms are of similar importance. For most locations below 20°N, wet deposition of sulfate dominates in accordance with heavy precipitation rates.

### 3.2. SECTOR WISE CONTRIBUTION TO SULFUR DEPOSITION

Sulfur deposition by major source category is presented in Figure 6. The percentage contribution to sulfur deposition due to shipping activities (Figure 6a) is over 10% for broad regions below 20°N. Besides dominating sulfur deposition levels over the ocean areas, ships also contribute significantly to on-shore deposition; Malaysia, Indonesia, Vietnam, and the Philippines. Islands west of Borneo, south of Sumatra receive up to 50% of their on-shore sulfur deposition from shipping activities. Most of the contribution is attributed to the heavy small-scale cargo shipping activities in these regions. An in-depth analysis on ship's contribution to sulfur deposition in Asia is presented in Streets *et al.* (1997, 2000).

Bio-fuels form a major source of energy for most of the underdeveloped regions in Asia (Streets *et al.*, 1998). Bangladesh, Nepal, and parts of northeastern India receive ~25–50% of their annual sulfur deposition from bio-fuel usage (Figure 6b). The contribution of large point sources (Figure 6c) varies significantly throughout Asia with most of the deposition occurring in eastern China, central India and northern Thailand. The maximum amount of deposition due to point sources is ~70%, where as on average ~10% is accounted by LPS's. This is consistent with the LPS emissions, which comprise ~16% of the total anthropogenic sulfur emissions in Asia. LPS's have a greater tendency to be transported out of the region than the area sources, and as a result, the fraction of sulfur deposition from LPS emissions in Asia is lower than the fraction of LPS emissions. Throughout eastern and southeastern Asia, volcanoes dominate the amount of sulfur deposited (Figure 6d). Volcanoes account for ~30% of the total deposition in Japan, ~30% in Indonesia, and ~25% in the Philippines. The largest impact of volcanic emissions is over southern Sumatra, Indonesia, Papua New Guinea and the southern tip of Kyushu, Japan. The percentage contribution to total sulfur deposition from the rest of the emission sources, viz., transport, industrial and small-scale power generation, domestic sources, etc is presented in Figure 6e. The urban centers are clearly dominated by these source sectors. For example, more than 75% of sulfur deposition in Mumbai and its surroundings is from these sources. On an average, most of the Asian countries receive over 50% from transport, industries and small

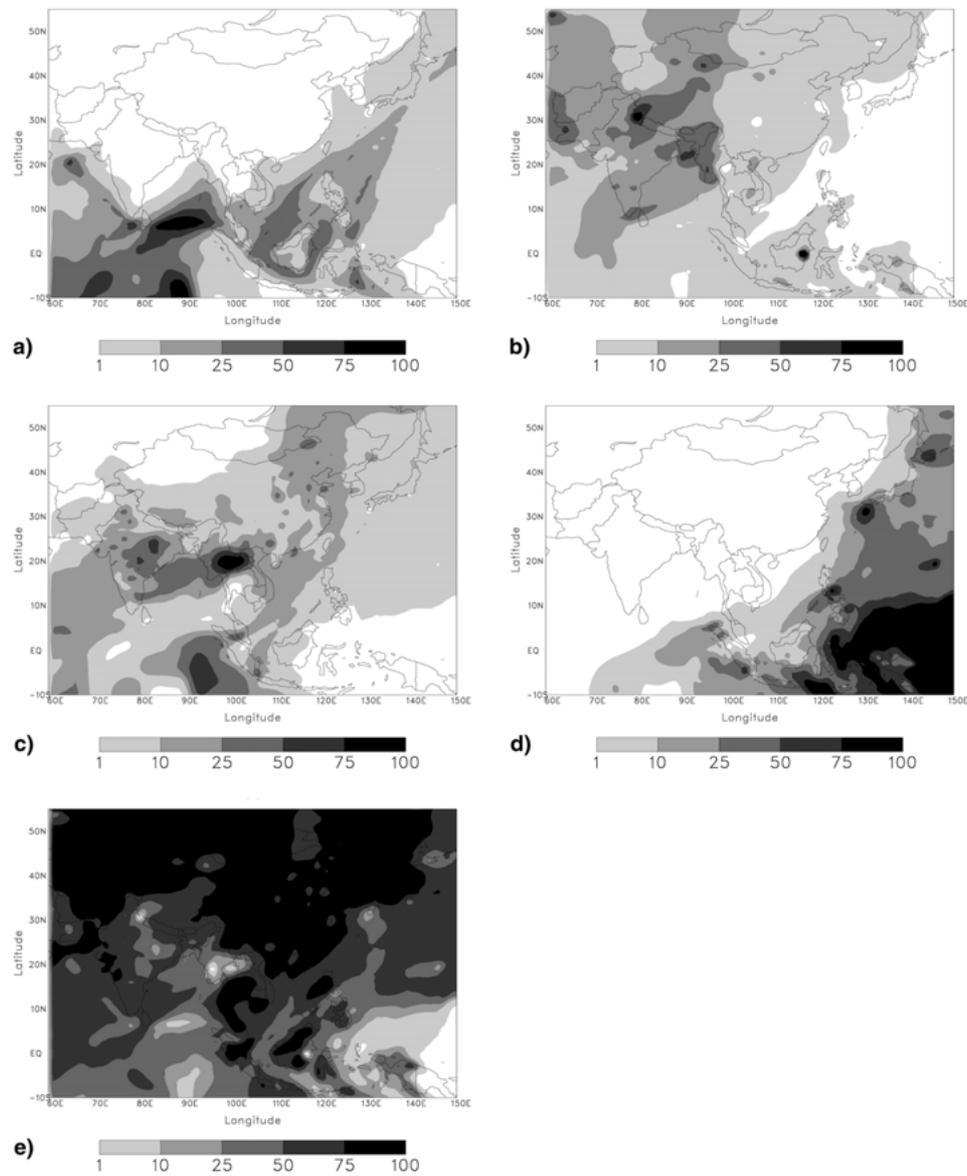


Figure 6. Percentage contributions to annual total sulfur deposition by source for 1990 in Asia: (a) Shipping lanes; (b) Bio-fuel burning; (c) Large Point Sources; (d) Volcanoes; (e) Rest of the sources.

scale domestic and power generation sectors. Most of the Indian sub-continent receives between 50 to 75% of its sulfur deposition from sources other than bio-fuel burning, large point sources, shipping activities and volcanoes, emphasizing the importance of these sources in this region. In contrast to most of China and Japan, which receives over 75% of its sulfur deposition from sources other than those mentioned above.

Coal is by far the dominant energy source followed by the biomass and other renewable and Non-renewable energy sources in the developing countries of Asia. Noncommercial energy sources such as wood, dung, agricultural waste and other forms of biomass are the main source of energy for most of the rural population. Regional air pollution could also prove problematic, especially in the rapidly growing, densely populated, coal intensive economies of Asia. Through 2020 in Asia, the principle change in the primary energy mix will be from traditional renewable energy sources to commercial fossil fuels (Nakicenovic *et al.*, 1998). In the future, shift from noncommercial energy and rather inefficient energy use toward higher-quality and efficient fuels, that is, to liquids and grid-dependent forms of energy for industrial and domestic processes will lead to large efficiency improvements and greatly reduced contributions to sulfur deposition from the above sectors.

### 3.3. SEASONAL BEHAVIOR OF SULFUR DEPOSITION IN ASIA

Figure 7 presents the seasonal contribution to sulfur deposition and Table II summarizes the percent deposited by season for each country. In general, sulfur deposition is highest during the summer and lowest in the winter, and is typically ~10% higher in summer and fall than in winter and spring. Higher deposition during the summer months is the result of heavy precipitation and on-shore winds over the large emitter countries.

Seasonal sulfur deposition over the Indian sub-continent reflects the monsoonal precipitation pattern. For example, 42% of Nepal's sulfur deposition occurs during the summer, in contrast to 13% in the winter. Similarly, summer deposition in India is more than double the winter sulfur deposition. Transport away from the continent, combined with low precipitation over the Indian sub-continent and northern China, results in significant levels of sulfur being moved away from the continent in the winter. The transport of sulfur away from the Indian sub-continent is very apparent over the Indian Ocean during the winter months, as much of the Indian Ocean receives its sulfur deposition during this period. Sulfur deposition over the Bay of Bengal has high variability between the two seasons, with almost no deposition occurring in the summer months to over half of its deposition occurring in the winter months. China, the Koreas, and Japan also experiences maximum sulfur deposition during their wet season. China receives nearly one-third of its deposition in the fall season. Similar trends are observed over Southeast Asia. In Thailand, 62% of sulfur deposition occurs in summer and fall, in contrast to 38% in winter and spring. Not all of Asia receives its maximum deposition during the

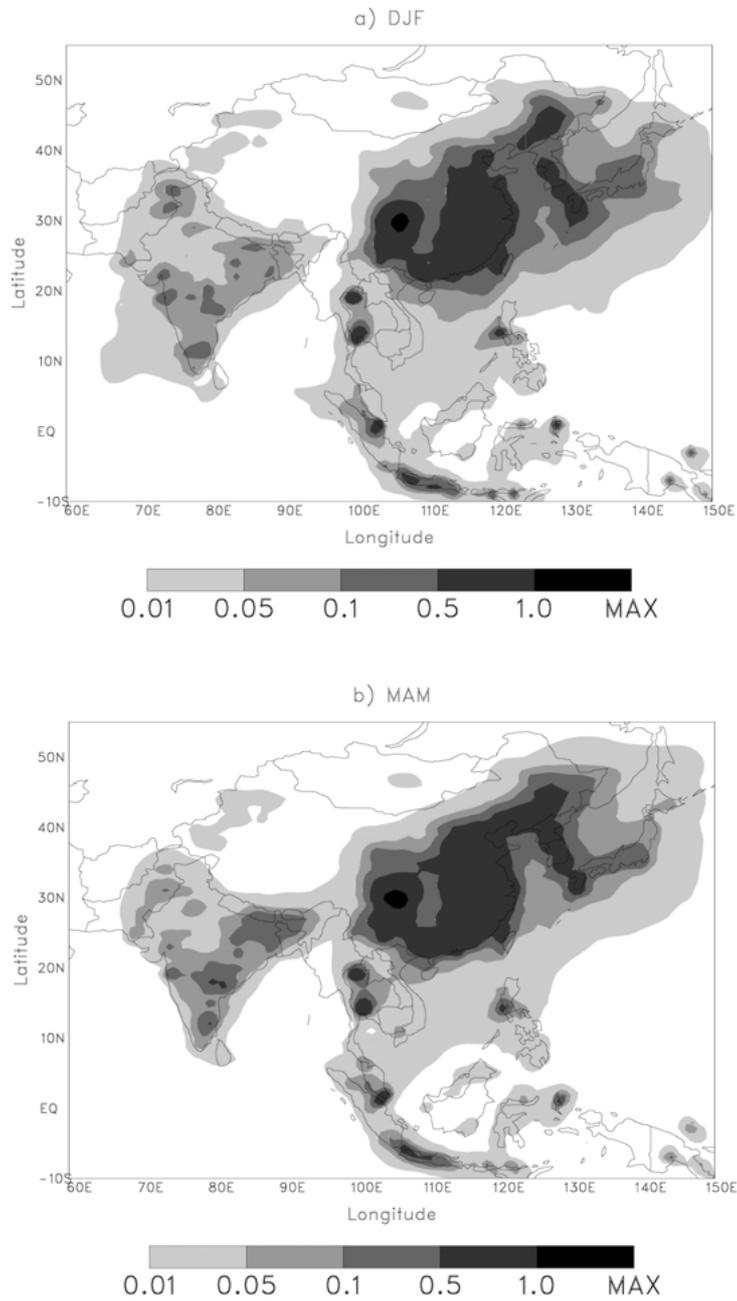


Figure 7a-b. Calculated seasonal sulfur deposition for 1990 in Asia in  $\text{g S m}^{-2} \text{ yr}^{-1}$ : (a) Winter (DJF); (b) Spring (MAM).

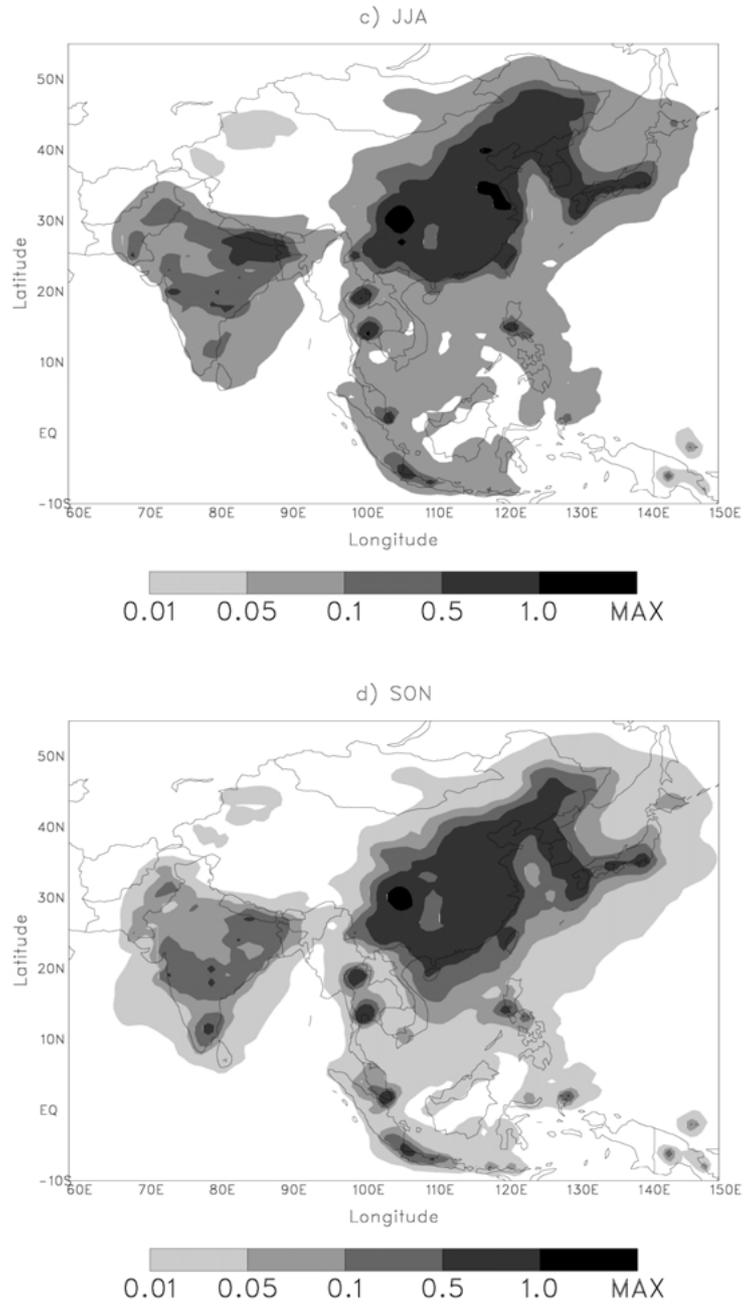


Figure 7c-d. Calculated seasonal sulfur deposition for 1990 in Asia in  $\text{g S m}^{-2} \text{ yr}^{-1}$ : (c) Summer; (JJA) (d) Fall (SON).

TABLE II

Country-wise calculated seasonal sulfur deposition for 1990 in Asia, as a percentage of their annual total sulfur deposition, using estimated 1990 sulfur emissions and 1990 meteorology from NCEP/NCAR (Kalnay *et al.*, 1996)

Country	Percentage of annual sulfur deposition by season			
	DJF	MAM	JJA	SON
Ocean	26	25	26	24
Bangladesh	17	30	29	24
Bhutan	8	35	39	18
Brunei	38	22	15	25
Myanmar	22	29	19	29
Cambodia	13	20	34	33
China	22	22	32	25
Hong Kong	36	21	17	26
India	16	20	35	30
Indonesia	29	25	26	21
Japan	21	22	30	26
N. Korea	18	22	36	24
S. Korea	20	20	32	27
Laos	19	34	30	18
Malaysia	17	26	28	30
Mongolia	27	20	31	23
Nepal	13	23	42	22
Pakistan	21	21	37	20
Philippines	24	23	26	8
Sea Lanes	33	23	18	26
Singapore	38	24	7	30
Sri Lanka	18	19	33	30
Taiwan	24	21	29	26
Thailand	16	22	31	31
Vietnam	20	26	21	33
Total	22	22	30	25

summer or fall months; Singapore receives nearly 40% of its deposition in winter compared to 7% in fall. This is due to the winter monsoons over Southeast Asia. Due to heavy precipitation throughout the year, much of the Southeast Asia shows less variation in their intra-annual deposition.

Figure 8 presents the seasonal variation of the wet and dry removal processes for (a) China, Mongolia, and Hong Kong, (b) Japan, Taiwan and the Korean Pen-

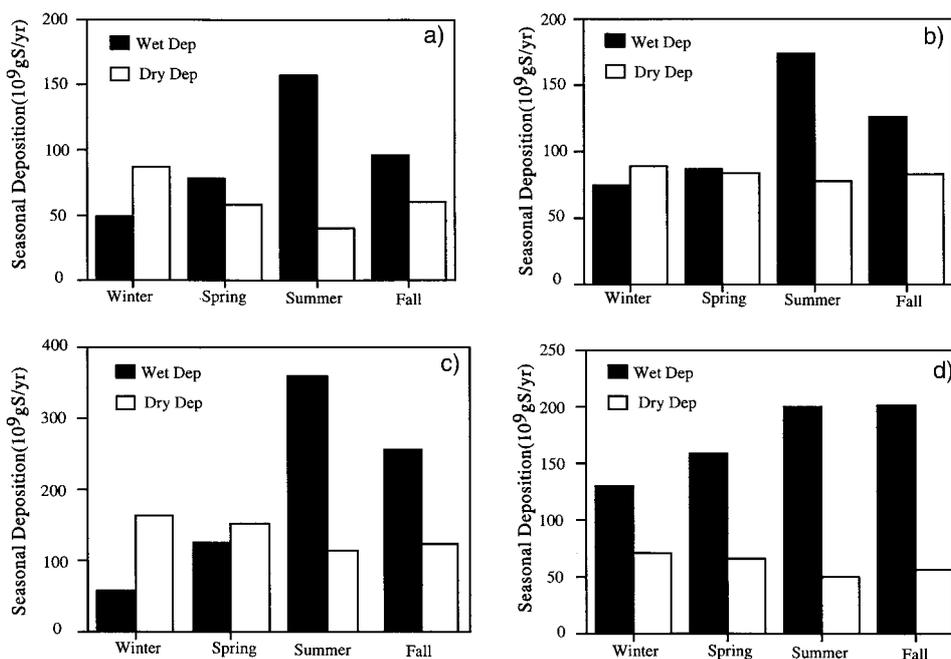


Figure 8. Calculated seasonal wet and dry sulfur deposition for 1990 in  $10^9 \text{ g S yr}^{-1}$  for four sub-regions within Asia: (a) China, Hong Kong, and Mongolia; (b) Japan, N. Korea, S. Korea, and Taiwan; (c) the Indian sub-continent; (d) Southeast Asia.

insula, (c) the Indian sub-continent, and (d) Southeast Asia. As expected, the wet deposition of sulfur follows the seasonal precipitation pattern. The high level of wet removal experienced during the summer months in the northern latitudes (China, and Mongolia) and the Indian sub-continent can be contrasted with the low deposition values experienced during the winter months as discussed previously. The Indian sub-continent's wet sulfur deposition is more than six times higher for the summer months as in the winter months. The relative importance of dry and wet removal also shows considerable variation. Unlike the rest of Asia, in Japan, dry deposition remains fairly constant for all the seasons. In Southeast Asia, wet deposition exceeds dry deposition for all four seasons. A comparison of wet and dry deposition fluxes in India indicates that the majority of sulfate deposition in this country is the result of wet removal processes. The model calculated wet-to-dry deposition ratios and the overall mass balance are consistent with observation-based analysis (Whelpdale, 1996).

### 3.4. OBSERVED SULFUR DEPOSITION AND CONCENTRATIONS IN ASIA

Observations provide a basis for evaluating the model's ability to capture spatial variations in sulfur deposition and concentrations. Unfortunately, the observations of sulfur deposition in Asia are sparse and many measurements are for relatively

TABLE III

Observed to Modeled annual wet sulfur Deposition in China, Japan, Indonesia and Malaysia, along with the range of wet sulfur deposition values from the surrounding eight grid cells, in  $\text{g S m}^{-2} \text{yr}^{-1}$

Location	Observed	Modeled	Range	Location	Observed	Modeled	Range
<b>China<sup>a</sup></b>				<b>Malaysia<sup>c</sup></b>			
Ulumuqi	1.20	0.01	0.01–0.02	Bayan Lepas	0.30	0.28	0.15–0.35
Kunming	0.78	0.76	0.67–1.00	Bukit Kledang	0.10	0.39	0.23–0.47
Lanzhou	0.81	0.16	0.13–0.20	Petaling Jaya	0.26	0.78	0.38–1.05
Chengdu	2.89	2.85	1.48–3.73	Senai	0.10	1.84	0.28–2.32
Nanchong	3.65	5.64	3.50–5.86	Tawau	0.01	0.04	0.02–0.04
Baise	0.63	0.69	0.60–0.97	Kuching	0.14	0.04	0.03–0.04
Guiyang	0.30	2.72	1.53–3.19	Bukit Tinggi	0.05	0.05	0.03–0.06
Nanning	1.40	1.09	0.56–2.15	Melaka	0.16	0.61	0.26–0.80
Xian	2.82	0.69	0.54–0.88	Kuala Terengganu	0.02	0.17	0.13–0.30
Guilin	1.86	1.05	0.89–1.63	Alor Setar	0.01	0.20	0.11–0.31
Maoming	0.44	0.33	0.24–0.41	<b>Indonesia<sup>d</sup></b>			
Taishan	2.96	0.60	0.33–0.76	Lhokseumawe	0.08	0.06	0.05–0.08
Xiangfan	1.39	1.32	1.07–1.51	Medan	0.07	0.18	0.08–0.21
Changsha	0.90	1.05	0.87–1.28	Palembang	0.36	0.16	0.09–0.20
Yingde	3.70	0.89	0.67–1.06	Tanjung Karang	0.14	1.37	0.32–1.69
Guangzhou	2.23	1.26	0.67–1.67	Baai Island	0.14	0.10	0.05–0.14
Yichun	3.78	0.77	0.62–0.93	Jambi	0.04	0.15	0.10–0.19
Shantou	1.28	0.58	0.21–0.69	Pakanbaru	0.12	0.19	0.12–0.19
Chengde	1.99	0.91	0.73–1.08	Bandung	0.50	0.65	0.37–0.77
Chongqing	5.33	2.17	1.69–2.59	Cisarua	0.88	0.97	0.40–1.34
Xuzhou	2.49	2.02	1.71–2.32	Surabaya	0.89	0.27	0.10–0.36
Jinan	1.42	1.25	1.11–1.53	Banjarbaru	0.23	0.04	0.03–0.05
Hefei	1.54	1.81	1.56–2.16	Pontianak	0.11	0.04	0.03–0.04
Xiamen	1.64	0.55	0.31–0.62	Samarinda	0.12	0.03	0.02–0.04
Qinhuangdao	2.81	0.51	0.45–0.57	Dilli	0.22	0.01	0.01–0.01
Suzhou	3.48	1.44	1.05–1.64	Manado	0.28	0.02	0.01–0.02
Shanghai	1.82	1.92	1.31–2.83	Palu	0.03	0.02	0.01–0.03
Siping	0.97	0.94	0.75–1.08	Ambon	0.22	0.01	0.01–0.01
Changchun	0.53	0.29	0.27–0.30	Jayapura	0.08	0.10	0.04–0.23
<b>Japan<sup>b</sup></b>				<b>Japan</b>			
Amagasaki	1.13	0.84	0.69–0.90	Noshiro	1.07	0.34	0.31–0.39
Fukue	1.00	0.50	0.42–0.62	Nonodake	0.50	0.46	0.37–0.60
Tokoro	0.47	0.15	0.13–0.17	Otofuke	0.40	0.18	0.15–0.20
Takamatsu	0.63	0.78	0.69–0.82	Kawasaki	1.77	0.70	0.53–0.80
Oki	1.17	0.80	0.71–0.89	Takaoka	0.93	0.92	0.43–1.14

<sup>a</sup> Bai *et al.*, 1997 (for year 1992–1993).

<sup>b</sup> CREIPI, 1992; Murano, 1994 (for year 1990).

<sup>c</sup> Bureau of Meteorology, Indonesia (for 1995).

<sup>d</sup> Malaysian Meteorological Services (for 1993).

Note: Model predicted values are from the simulation using 1990 emissions and 1990 meteorology and year in the brackets above indicate the year of measurement.

short periods. In Japan this is not a significant problem, as the sulfur emissions are not changing rapidly. But the rapid growth of emissions, elsewhere in Asia, makes it difficult to compare model results for specific years with measurements from different years. With this limitation in mind, comparisons of predicted and observed values do provide valuable insights into spatial variation of sulfur deposition and concentrations and model performance. We have compiled a database of sulfur deposition from Japan, China, Indonesia and Malaysia for the early 1990s. A compilation of this data is presented in Table III. This data contains information from the Japanese Environmental Agency which has maintained an extensive sulfur deposition monitoring network since 1986 (Murano, 1994). Observational data were also obtained from the Central Research Institute for Electric Power Industry (CRIEPI, 1992), Japan, for 1987–1990. Observations for wet sulfur deposition in China were obtained from Bai *et al.* (1997). Data were also compiled from the Bureau of Meteorology and Geophysics (BMG, 1995), Republic of Indonesia and the Malaysian Meteorological Service (1994). The predicted values along with the range of predicted values from the surrounding eight grid cells for 1990 are also presented in Table III.

In Japan, observed sulfur deposition values ranged from 0.5 to 1.8 g S m<sup>-2</sup> yr<sup>-1</sup> while the model predicted values ranged from 0.2 to 1.5 g S m<sup>-2</sup> yr<sup>-1</sup>. Maximum deposition amounts are found near major urban areas, in Kyushu, and along the Japan Sea side of Honshu. In China, observations ranged from 0.1 to 5.3 g S m<sup>-2</sup> yr<sup>-1</sup> while the model predicted sulfur deposition ranged from values as high as 10.5 g S m<sup>-2</sup> yr<sup>-1</sup> around the city of Chongqing to as low as 0.01 g S m<sup>-2</sup> yr<sup>-1</sup> in the regions of northwest China. Figure 9a, provides a spatial distribution of predicted annual wet sulfur deposition in g S m<sup>-2</sup> for 1990, overlaid by some of the observations from Table III. The model has a tendency to under-predict sulfur deposition but succeeds in capturing the spatial variation. The under-prediction is partially the result of most monitoring sites being located in or near urban areas, and hence the one-degree resolution of the model is too coarse to capture local influences.

Figure 9b presents the predicted annual wet sulfur deposition overlaid by some of the observations from Table III in Indonesia and Malaysia. In general, observed and modeled sulfur deposition displayed a similar range (0.01 to 1.8 g S m<sup>-2</sup> yr<sup>-1</sup>) and an appropriate spatial variability. There is a tendency for the model to over predict wet deposition in Malaysia. The largest discrepancy at Senai, Malaysia (0.1 to 1.8 g S m<sup>-2</sup> yr<sup>-1</sup>) exists because this station falls within the 1° grid box that includes Singapore.

In addition to monitoring sulfur deposition, extensive monitoring of airborne concentrations was performed in Japan (CRIEPI, 1994). Data on SO<sub>2</sub> concentrations from 11 Asian countries is also available from a passive sampler network operated in 1994 (Carmichael *et al.*, 1995). This network of samplers was developed with the assistance of domestic environmental agencies and researchers. Some of the measurements from the passive sampler network, Table IV, are over-



TABLE IV

Observed to modeled annual averaged SO<sub>2</sub> concentrations in Asia along with the range of SO<sub>2</sub> concentrations from the surrounding eight grid cells for 1995, in  $\mu\text{g SO}_2 \text{ m}^{-3}$

Country	Location	Observation <sup>a</sup>	Modeled	Range
Bangladesh	Jahangirnagar Sava	6.1	2.2	1.9–2.4
China	Chongqing	12.1	43.5	16.9–154.0
	Yueyang	7.3	12.3	8.6–14.0
	Huitong	11.0	10.3	6.3–15.2
	Liuchongguan	18.7	61.2	15.6–78.2
	Fuye Ding, Beijing	63.0	19.4	11.5–49.8
Hong Kong	Cape, D'Aequier	4.0	23.0	3.3–43.1
India	Madras	8.8	8.4	2.1–15.3
	Berhampur, Univ.	1.2	1.5	0.4–1.9
	Cochin, Univ.	1.7	1.4	0.4–1.9
	BHU, Varanasi	2.8	12.6	3.5–39.6
	Bombay	11.2	8.6	4.4–13.9
	Nehu Shilong	1.3	0.7	0.6–1.5
	Agra	6.1	4.4	2.2–5.6
	Delhi	28.2	12.2	6.9–15.1
	Indonesia	Medan	1.2	0.9
Palembang		1.5	0.7	0.4–1.5
Balikpapan		1.3	0.2	0.1–0.3
Ojung Bandung		0.6	0.8	0.1–0.8
South Korea	Cheju	4.0	1.8	0.9–2.2
	YangYang	5.1	6.9	3.1–19.7
	Taeon	5.9	8.1	2.5–37.5
Malaysia	Tanah Rata	0.3	2.5	1.2–8.8
	Mersing	1.7	3.3	0.8–15.8
	Sibu	1.2	0.4	0.3–0.5
	Lawa Mandau	0.3	0.5	0.2–0.6
Nepal	Dhangadi	1.1	1.6	0.5–1.9
	Dhankuta	0.6	1.4	0.5–2.2
	Janakpur	2.1	0.5	0.2–0.8
Taiwan	Leiu	1.3	0.6	0.2–0.8
Thailand	Surat-Thani	1.5	0.4	0.2–0.8
	Chieng Rai	1.7	11.5	0.5–21.6
	Tak	3.6	0.5	0.2–0.9
	Chonburi	3.2	11.1	1.4–19.9

TABLE IV  
Continued

Country	Location	Observation <sup>a</sup>	Modeled	Range
Vietnam	Kien An	8.3	1.7	1.1–2.6
Japan <sup>b</sup>	Otobe	4.1	0.8	0.6–1.0
	Kashima	3.0	6.8	2.8–7.0
	Amami	0.5	0.4	0.3–0.6
	Hachijyo	0.8	0.6	0.5–0.8

<sup>a</sup> Carmichael *et al.*, 1995b.

<sup>b</sup> CRIEPI, 1994.

Note: Model predicted values are from simulation using 1995 emissions and 1994 Meteorology and the observations listed above for 1994.

laid on the predictions in Figure 5b. Observed values show considerable variation in magnitude, as low as  $0.3 \mu\text{g m}^{-3}$  at Lawa Madau and Tanah Rata, Malaysia to as high as  $63 \mu\text{g m}^{-3}$  in Beijing, China. Table IV, presents a direct comparison of measured to model calculated  $\text{SO}_2$  concentrations and the range of predicted values from the surrounding eight grid cells. The model does capture the spatial variation of the observed values, reflecting the spatial distribution of emissions. For example, the model was successful in capturing the high variations in concentration, ranging as high as  $61 \mu\text{g m}^{-3}$  in Liuchongguan, China to as low as  $0.2 \mu\text{g m}^{-3}$  in Balikpapan, Indonesia. Further insight into model performance requires better characteristics of the inter-annual variability in calculated and measured sulfur deposition.

#### 4. Conclusions

Sulfur deposition due to anthropogenic and volcanic emissions in Asia was calculated using the ATMOS model. Annual and seasonal deposition was investigated for 1990 and 1995 emissions and using 1990 and 1994 meteorology. Due to Asia's geographical size, diverse topography, and disparity in emission intensity, the transport patterns and deposition of sulfur vary widely throughout the region. Deposition and concentrations were found to closely reflect the distribution of emissions. Shipping activities dominate sulfur deposition in Southeast Asia with  $\sim 25\%$  resulting from this sector, along with active volcanic activities over Papua New Guinea and the Philippines. Most of the emissions from large point sources are transported out of the source region, with maximum contribution from LPS's over central India and Thailand. Bio-fuels form a major source of energy demand and its contribution to sulfur deposition is significant over the underdeveloped regions of Asia. Species deposition and concentrations are heavily dependent on seasonal changes in precipitation and flow fields. The summer monsoon is the largest factor controlling

deposition in the Indian sub-continent. Similarly, deposition in the mid-latitudes shows a strong wet versus dry seasonal variation. The heavy precipitation over the Southeast Asia resulted in receiving more wet deposition than dry for all four seasons.

Comparison with a variety of observational data sets was performed. While these results present results from two years of meteorology (1990 and 1994) and emissions (1990 and 1995), this study should be considered preliminary. The spatial patterns and dynamic range in sulfur concentrations and deposition are well captured by the model. While the model tends to under-predict deposition, this can be attributed, at least in part, to under-estimation of emissions (e.g. the absence of Russian and DMS emissions), parameterization of wet and dry removal processes, and limitations to capture local influences at  $1^\circ \times 1^\circ$  model resolution. The situation in Asia, with very distinct seasons and much higher precipitation amounts, clearly depicts the need for a detailed study to characterize wet and dry removal processes in Asia.

Asia's economic growth in the coming decades has potentially serious environmental ramifications for the region. Without an increase in emissions control regulations for the region, SO<sub>2</sub> emissions are expected to increase by a factor of two to three by 2020 (Foell *et al.*, 1995). The results presented in this article provide valuable information on the air quality, especially on seasonal variation of sulfur deposition and sector wise contribution from major sources, viz., transportation, bio-fuels, power plants, etc., which can be used as a basis for projecting future risks to ecosystem. Use of higher-quality and efficient fuels in the future, for transport, industrial and domestic purposes will have a profound impact on Asia's environment, in greatly reducing the sulfur deposition contributions from these sectors.

### Acknowledgements

Much of the work upon which this article is based was based on research supported in part by funds from The World Bank and The Asian Development Bank as part of the RAINS-ASIA project. Special thanks to the collaborators on the RAINS-ASIA Phase-I project.

### References

- Akimoto, H. and Narita, H.: 1994, *Atmos. Env.* **28**, 213.  
Arndt, R. L.: 1997, 'The Role of Sulfur Emissions in Asia's Environmental Change: Analysis on a Regional and Urban Scale', Ph.D. Thesis, Department of Chemical and Biochemical Engineering, The University of Iowa, U.S.A., 167 pp.  
Arndt, R. L., Carmichael, G. R., Streets, D. G. and Bhatti, N.: 1997, *Atmos. Env.* **31**, 1553.  
Arndt, R. L., Carmichael, G. R. and Roorda, J. M.: 1998, *Atmos. Env.* **32**, 1397.

- Bai, N., Lui, N. and Wang, X.: 1997, 'About acid rain in China', Proceeding of the 3rd Joint Seminar on the Regional Deposition Processes in the Atmosphere, Nara, Japan, 5–7 November 1997, pp. 149–158.
- BMG, Bureau of Meteorology and Geophysics, Republic of Indonesia: 1995, 'Annual Summary of Air Pollution Observations'.
- Calori, G. and Carmichael, G. R.: 1999, *Atmos. Env.* **33**, 3109.
- Carmichael, G. R. and Arndt, R. L.: 1995a, 'ATMOS Module – Long Range Transport and Deposition of Sulfur in Asia', in W. Foell, M. Amman, G. Carmichael, M. Chadwick, J. Hettelingh, L. Hordicjk and Z. Dianwu (eds.), *RAINS ASIA: An Assessment Model for Acid Rain in Asia*, The World Bank, Washington, DC, U.S.A., pp. V-1–V-58.
- Carmichael, G. R., Ferm, M., Adikary, S., Ahmad, J., Mohan, M., Hong, M. S., Chen, L., Fook, L., Liu, C. M., Soedomo, M., Tran, G., Suksomsank, K., Zhao, D., Arndt, R. and Chen, L. L.: 1995b, *Water, Air, and Soil Pollut.* **85**, 2289.
- CRIEPI: 1992, 'Data Report of CRIEPI Acidic Deposition Project, 1987–1900', Central Research Institute of Electrical Power Industry Report, Tokyo, Japan, Report #T91019.
- CRIEPI: 1994, 'Acidic Deposition in East Asia – International Workshop on Acidic Deposition in East Asia', Central Research Institute of Electrical Power Industry, Tokyo, Japan, Report #ET93002, 79 pp.
- Foell, W., Green, C., Amann, M., Bhattacharya, S., Carmichael, G., Chadwick, M., Cinderby, S., Haugland, T., Hettelingh, J.-P., Hordijk, L., Kuylenstierna, J., Shah, J., Sherestha, R., Streets, D. and Zhao, D.: 1995, *Water, Air, and Soil Pollut.* **85**, 2277.
- Fujita, S.: 1992, 'Acid Deposition in Japan', Central Research Institute of Electrical Power Industry, Tokyo, Japan, Report #ET91005.
- Ichikawa, Y. and Fujita, S.: 1995, *Water, Air, and Soil Pollut.* **85**, 1927.
- Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., Iredell, M., Saha, S., White, G., Woollen, J., Zhu, Y., Chelliah, M., Ebisuzaki, W., Higgins, W., Janowiak, J., Mo, K. C., Ropelewski, C., Wang, J., Leetma, A., Reynolds, R., Jenne, R. and Joseph, D.: 1996, *Bulletin of American Meteorological Society* **77**, 437.
- Malaysian Meteorological Services: 1994, 'Annual Summary of Air Pollution Observations'.
- Murano, K.: 1994, 'Activity of JEA for East Asian Acid Precipitation Monitoring Network', Proceeding of the Workshop on Acid Rain Monitoring Network in South, East, and Southeast Asia, Kuala Lumpur, Malaysia, 17–19 May 1994.
- Nakicenovic, N., Grubler, A. and McDonald, A. (eds.): 1998, *Global Energy Perspectives*, Cambridge University Press, Cambridge, United Kingdom, 299 pp.
- Sharma, M., McBean, E. A. and Ghosh, U.: 1995, *Atmos. Env.* **29**, 2157.
- Shrestha, R. M. and Bhattacharya, S. C.: 1991, 'Coal Use in Electricity Generation in Asia and Emission Control Options', Proceedings of the 3rd Annual Conference on Acid Rain and Emissions in Asia, Bangkok, Thailand, 18–21 November 1991, pp. 108–120.
- Spiro, P. A., Jacob, D. J. and Logan, J. A.: 1992, *J. of Geo. Res.* **97**, 6023.
- Streets, D., Amann, M., Bhatti, N., Cofala, J. and Green, C.: 1995, 'Emissions and Control', in W. Foell, M. Amman, G. Carmichael, M. Chadwick, J. Hettelingh, L. Hordicjk, and Z. Dianwu (eds.), *RAINS ASIA: An Assessment Model for Acid Rain in Asia*, The World Bank, Washington, DC, U.S.A., pp. IV-1–IV-40.
- Streets, D., Carmichael, G. R. and Arndt, R. L.: 1997, *Atmos. Env.* **31**, 1573–1582.
- Streets, D. G. and Waldhoff, S. T.: 1998, *Energy* **23**, 1029.
- Streets, D. G., Carmichael, G. R., Amann, M. and Arndt, R. L.: 1999, *Ambio* **28**, 135.
- Streets, D. G., Guttikunda, S. K. and Carmichael, G. R.: 2000, *Atmos. Env.* **34**, 4425.
- Whelpdale, D. M. and Kaiser, M. S. (eds.): 1996, *Global Acid Deposition Assessment*, World Meteorological Organization, Global Atmospheric Watch, Report #106, 241 pp.
- Xu, Y. and Carmichael, G. R.: 1998, *J. of Applied Met.* **37**, 1084–1099.