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INVITED PAPER

INTERANNUAL VARIABILITY IN SULFUR DEPOSITION IN ASIA

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Abstract

Source-receptor relationships for sulfur deposition in Asia are explored over a period of thirteen years (1985-97) by means of a multi-layer Lagrangian model. Calculations are driven by year-by-year emissions inventories and meteorology. Over this time total deposition of sulfur has increased throughout most of Asia, by more than a factor of two in Central India and North-Central Indochina; but has decreased slightly in South Korea, Southwest Japan, and parts of Taiwan and the Philippines. The interplay between emission trends and meteorological factors in year-to-year variability is also demonstrated. Examples are given in which meteorological factors partially or completely mask the expected deposition trends due to changes in emissions. The influence of precipitation anomalies is clearly visible throughout most of the area, and especially during the El Niño years 1994 and 1997 in the Indian subcontinent and in Southeast Asia. Yearly country total depositions are shown to vary due to changes in meteorology by more than 20% from the average values in the Indian subcontinent, generally less than 10% in Southeast Asia and to an intermediate degree in Indochina and North and East Asia.

KEYWORDS: *sulfur deposition, SO_2 emissions, interannual variability*

1. Introduction

The adverse effects caused by the growth of emissions levels throughout most of Asia have drawn increasing attention of researchers and policy makers. Population increase accompanied by fast expanding economies during the last two decades have boosted energy demand throughout the continent. The primary energy demand in Asia is currently doubling every twelve years, a pace much more rapid than the world average of every twenty-eight years (Downing *et al.*, 1997). Presently ~80% of the demand is satisfied by fossil fuels, with coal being the primary energy source. Most of the energy scenarios up to 2020 are

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characterized by a further increase in energy use, with fossil fuels remaining the dominate source (WEC, 1993; WEC-IIASA, 1995). In Asia especially, the demand for coal and oil is expected to double or triple in the next thirty years. As a consequence, current sulfur emissions throughout the continent, already nearly equal to those from Europe and North America combined (EMEP, 1999; Streets *et al.*, 1999), are likely to increase even more in the next decades, even though in some regions the emissions are likely to decrease. The resulting environmental impacts, including acid rain, are already extensively documented (Foell and Sharma, 1991; Foell and Green, 1992; Rodhe *et al.*, 1992; Hordijk *et al.*, 1995), and are expected to intensify.

This growth in emissions has stimulated various atmospheric modeling studies, investigating different aspects of transport, transformation and deposition mechanisms of sulfur compounds throughout the region (Huang *et al.*, 1995; Ichicawa and Fujita, 1995; Ikeda *et al.*, 1997; Arndt *et al.*, 1998; Hayami *et al.*, 1999). To better understand the consequences of future scenarios and emission abatement strategies, atmospheric models need to be coupled with consistent energy projections, related control technologies information and environmental impacts, as is done in integrated assessment models. This is the main focus of the RAINS-Asia Project (Foell *et al.*, 1995; Downing *et al.*, 1997; Shah *et al.*, 2000), currently in its second phase, aimed to provide a set of tools to study future air pollution and acid deposition scenarios in Asia. Within this framework, source-receptor relationships have also been developed, using the ATMOS model (Arndt and Carmichael, 1995), and incorporated in the RAINS-Asia integrated assessment model as a yearly region-to-grid transfer matrix. The atmospheric calculations so far have focused on the base year 1990.

The present work explores sulfur trends in Asia and assesses inter-annual variability by calculating sulfur deposition over the years 1985-1997, using year-to-year meteorology and emissions inventories. During this period most Asian countries have experienced dramatic changes in their emission levels. This long time frame allows the investigation of the combined effects of emission changes and inter-annual meteorological variability on depositions levels and source-receptor relationships throughout Asia. In the paper the main features of the modeling system are briefly summarized, together with the data sources employed for the study. Regional deposition trends are linked to the corresponding ones of emissions, analyzing country balances and meteorological factors. The magnitude of the inter-annual meteorological variability is finally estimated and its effect on source-receptor relationships assessed.

2. Methodology

2.1 Transport-deposition model

Atmospheric calculations were carried out by means of the ATMOS model (Arndt and Carmichael, 1995; Arndt *et al.*, 1997, 1998). ATMOS is a multi-layer source-oriented Lagrangian trajectory model, simulating emission, transport, chemical transformation and deposition of sulfur. The model assumes that during daytime the atmosphere is subdivided in two vertical layers, namely the planetary boundary layer (PBL) and the free troposphere above it, up to 6000 m. During nighttime the PBL becomes the residual boundary layer, and a surface layer of 300 m is also present. During every simulation time step (typically 3 hours) a new puff is released from each modeled source, and injected into the proper layer according to source type; area sources are injected in the lowest modeling layer (the PBL during daytime or the surface layer during nighttime), while elevated sources are always injected into the PBL. Puffs are followed for up to five days forward in time. Sulfur dioxide and sulfate are considered by the model, with a conversion rate depending on latitude and day of the year which accounts for gas-phase as well as in-cloud transformations. Both species are removed via dry and wet deposition processes, with parameters depending on land-sea terrain type, season and precipitation intensity. Model parameters have been updated for the current work, based on recent reviews and studies (Huang *et al.*, 1995; Ichikawa and Fujita 1995; Ichikawa and Hayami, 1998; Phadnis and Carmichael; 1998; Xu and Carmichael, 1998). Complete details about the model formulation are presented in (Arndt and Carmichael, 1995; Arndt *et al.*, 1997; Guttikunda *et al.*, 2001)

2.2 Simulation conditions and data sources

Calculations were performed for the years 1985 and 1990 through 1997, on a domain of 60° to 150° E and from 20° S to 55° N. Concentrations and depositions were computed on a reference grid at one degree resolution, the same resolution as the emissions data set. Major data sources employed in the study are described below.

2.3 Meteorological data

The NCEP Reanalysis data, provided by the NOAA-CIRES Climate Diagnostics Center (Boulder, Colorado), were used throughout all the simulated years. The data consisted of wind and precipitation fields at four-times daily values at ~2.5° resolution at the surface and at standard pressure levels. The PBL height was computed according to local heat flux, temperature profile, surface wind speed and surface properties using Carson (1973) and Venkatram (1980) methods for thermal and mechanical components, respectively. Surface properties were assigned according to the NASA Goddard DAAC ISLSCP Initiative I global land cover database (DeFries and Townshend, 1994).

2.4 Emission inventories

Anthropogenic emission fluxes were based on the work of Streets *et al.* (1999) and Streets (1999), and include ground-based sources as well as contributions from maritime traffic. The 1990 base year inventory (Streets *et al.*, 1995) uses detailed data on energy and fuel use by sector for the 99 regions listed in Table 1. The inventory spanning the years from 1985 to 1997 were derived from the baseline inventory for the year 1990 using country-based energy use statistics and emission control levels (in the form of reduced sulfur content of fuels and post-combustion abatement measures). Emissions were subdivided into area sources, arranged on a 1° resolution grid, and ~ 250 large point sources (Figure 1). Emission totals by country for each year are presented in Figure 2. During the nineties, the annual average growth rate of emissions for Asia as a whole was 2.9%; but with substantial regional differences, including higher growth rates in the Indian subcontinent (5.6% average), lower rates in East Asia (2.2%) and intermediate behavior in Southeast Asia (4.3%). The lower rates in East Asia are the consequence of control measures applied to various degrees in China, Japan, South Korea and Taiwan. The last three countries in fact have experienced reductions in SO_2 emissions throughout the nineties, with yearly average changes in sulfur emission rates of -1.4%, -5.7% and -2.0%, respectively. For some other countries the downward trend has started later, as is the case of Sri Lanka (-4.6%, after 1993), Singapore and Vietnam (-1.0% and -1.3%, both after 1994).

The emissions from the portion of the former Soviet Union (herein after referred as FSU) falling inside the simulation domain were taken from Ryaboshanko *et al.* (1996). According to that inventory the total emissions from the part of FSU falling into the simulation domain varied from 5.65 Tg SO_2 yr^{-1} in year 1985 to 5.30 Tg SO_2 yr^{-1} in year 1990.

Volcanic sources throughout Asia were assigned according to Andres and Kasgnoc inventory (1998) also integrated with Japanese data from Fujita (1992). Total volcanic emissions within the domain amount to 3.33 Tg SO_2 yr^{-1}, and were held constant throughout the period studied.

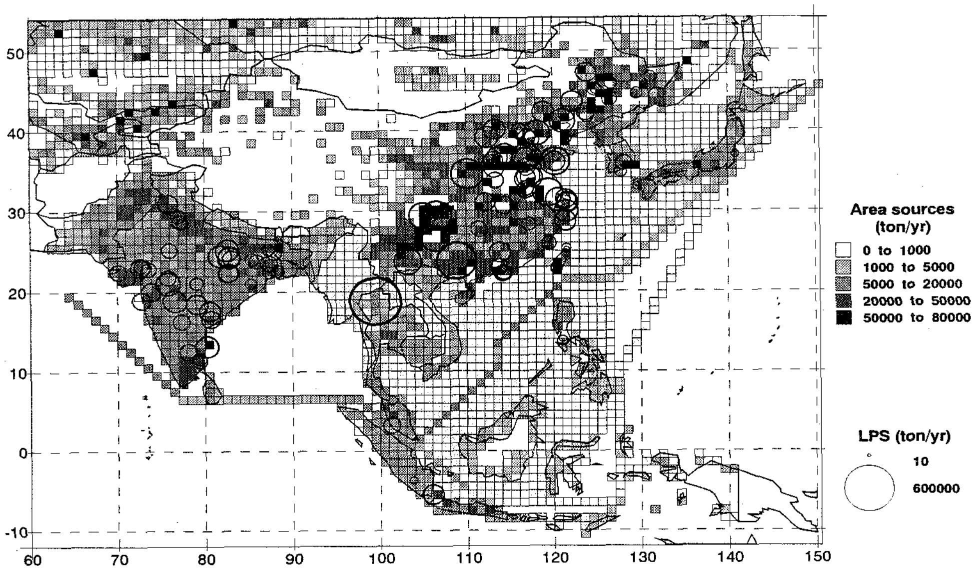


Figure 1. Emissions from area and large point sources, year 1990

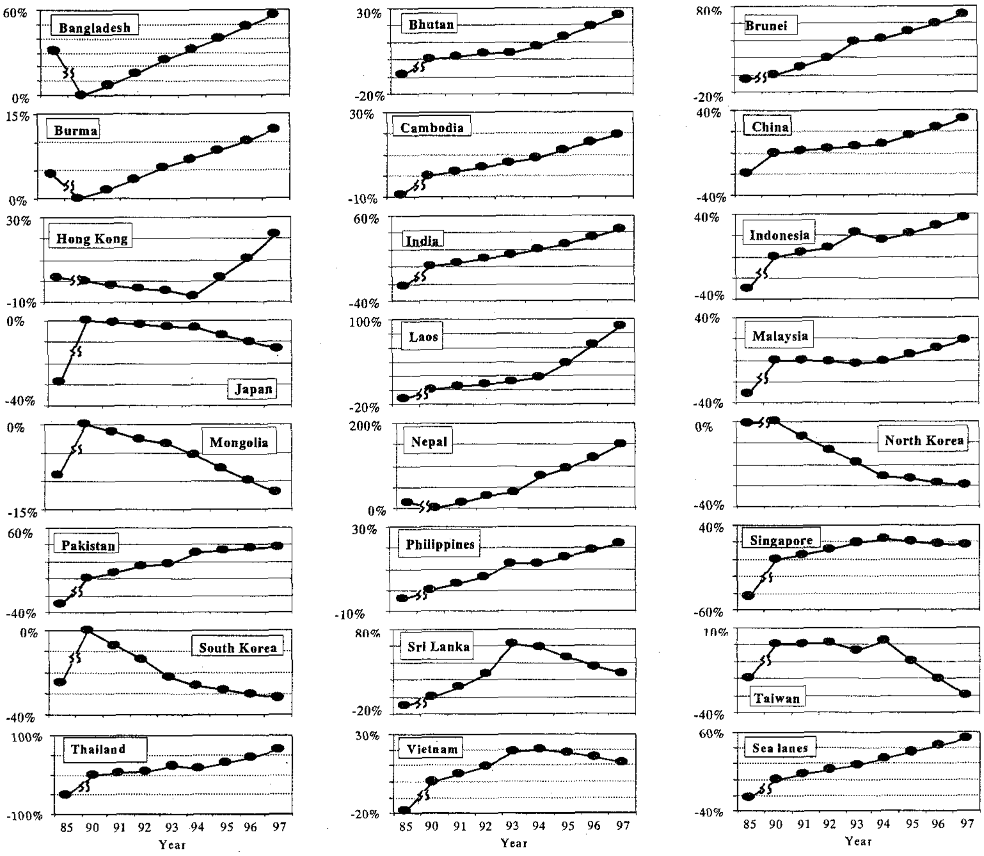


Figure 2. Trends in SO_2 emissions, by country (percentage change from 1990 levels)

Table 1.Yearly anthropogenic emissions, by country (Gg SO_2 yr^{-1})

Country	**Code**	**1985**	**1990**
Bangladesh	BANG	131	100
Bhutan	BHUT	1	1
Brunei	BRUN	6	6
Burma	BURM	20	19
Cambodia	CAMB	21	23
China	CHIN	17897	22225
Hong Kong	HONG	155	153
India	INDI	3402	4437
Indonesia	INDO	387	562
Japan	JAPA	729	1028
Laos	LAOS	3	3
Malaysia	MALA	176	256
Mongolia	MONG	74	81
Nepal	NEPA	19	17
North Korea	NKOR	349	353
Pakistan	PAKI	474	685
Philippines	PHIL	394	412
Singapore	SING	105	191
South Korea	SKOR	1281	1706
Sri Lanka	SRIL	23	26
Taiwan	TAIW	404	505
Thailand	THAI	454	964
Vietnam	VIET	92	113
Sea lanes	SEAL	469	607
Asia		27065	34474

3. Results

A detailed analysis of model performance including a comparison with observations in the region is presented elsewhere (Guttikunda *et al.*, 2001). These results indicate that ATMOS is able to capture many of the features and the long-term variability of deposition levels in different areas of Asia, as already emerged during previous applications (Arndt and Carmichael, 1995; Arndt *et al.*, 1997, 1998). Recently the new form of ATMOS model was evaluated against observational data for January and May 1993, and was intercompared with six other long-range transport models, including detailed Eulerian codes (Carmichael *et al.*, 2000). ATMOS performance in predicting sulfur deposition was shown to be very similar to these more detailed models.

3.1 Trends in country and regional balances

(1) East Asia

The results for East Asia are shown in Figure 3. In the 1985-1990 period the net increase of emissions of all these countries shown in Table 1 is clearly reflected in the deposition trends: increasing by 1.2% in North Korea, 34% in South Korea and 39% in Japan. Total sulfur deposition over China (not shown here) during the same period increased by 30%. During the nineties, the picture is more complex. According to the inventory estimations, as of year 1997 emissions had declined in both Koreas by about 30% from their 1990 levels, by about 14% in Japan, while Chinese emissions increased by more than 30%.

Emission trends in different regions also have clear effects in changing the relative contributions of different sources to depositions in specific receptor areas. For example in 1990 Japanese anthropogenic emissions contributed about 50% to the total sulfur deposition over Japan, with the remaining sources attributed to emissions from China (21%), volcanic sources (13%) and South Korea (13%). After 1990, Japanese emissions decreased continuously. However sulfur deposition continued to increase above 1990 levels. The significant increase in Chinese emissions counterbalanced the effect of decreasing Japanese emissions, resulting in a slight increase in total deposition over Japan in the 1990-97 period. This is also evidenced by the opposite trends of Chinese and Japanese shares, with Japan contribution to its deposition falling from 48% to 38%, and Chinese contribution to Japan deposition increasing from 26% to 40%.

A similar effect can be seen in the case of North Korea. The economic problems in North Korea during the nineties are reflected in a significant decrease in emissions since 1990. However sulfur deposition has not decreased and this is due to the increase in Chinese emissions.

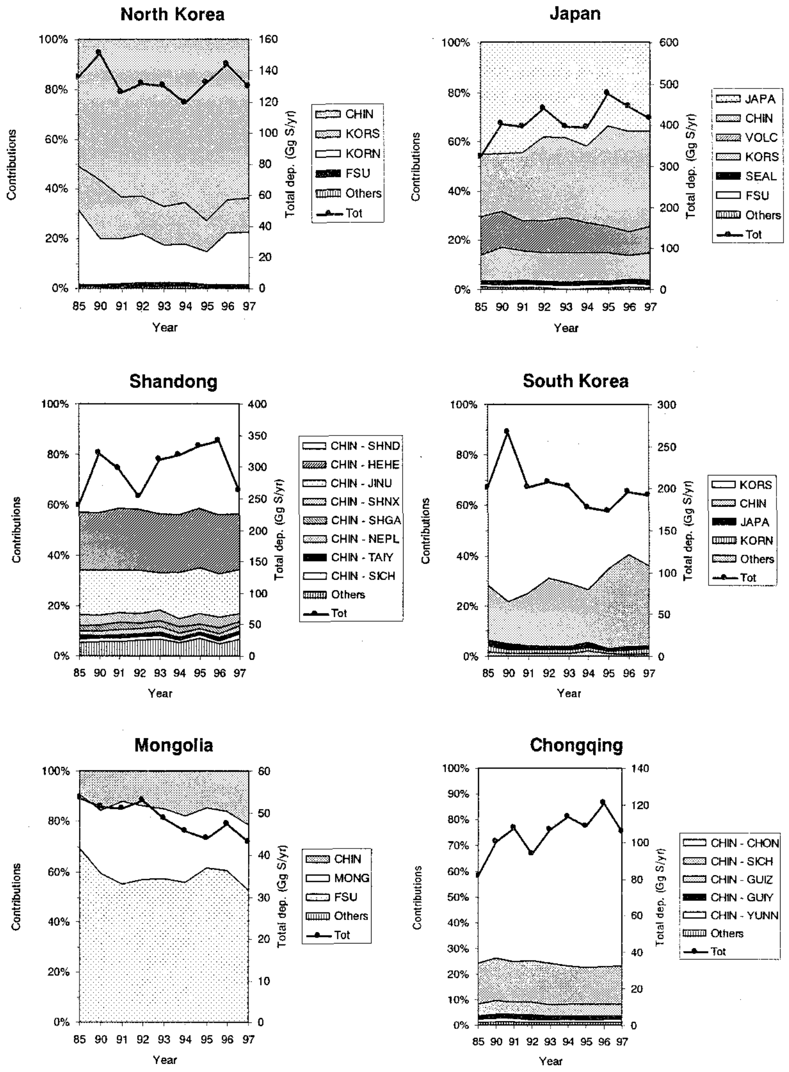


Figure 3. Yearly country and regional balances: total depositions (thick line with markers, right axis) and countributions shares, by country or region (dashed areas, left axis). Regions codes: CHON = Chongquing; GUIY = Guiyang; GUIZ = Guizhou; HEHE = Anhui, Hebei, Henen; JINU = Jiangsu (except Shanghai); SHND = Shandong; SHNX = Shanxi (except Taiyuan); SHGA = Gansu and Shaanxi; SICH = Sichuan (except Chongquing); TAIY = Taiyuan; YUNN = Yunnan. For country codes, refer to Table 2

The case of South Korea is different. As shown the indigenous emissions were responsible for the major fraction of sulfur deposition over the country (about 75% in year 1990). Here the decrease in South Korea emissions since 1990 has resulted in a net decrease in local sulfur deposition, even though the relative contribution of Chinese emissions increased during the nineties (from about 20% in 1990 to 38% in 1997). This downward trend over the country is also revealed by measured sulfate wet deposition and SO_2 air concentration measurements in large urban areas (Table 2).

The deposition trends are not a simple function of emissions. Deviations of deposition trends from the corresponding trends of emissions can be noticed in the figures (for example the decrease in sulfur deposition at Shandong by 10% in 1992 and 1997). Some of these discrepancies can be explained by year-to-year variations in precipitation fields. Figure 4 shows the relative anomalies in yearly precipitation, computed as relative deviations with respect to the average values over the studied period. The lower sulfur deposition over Japan in years 1993 and 1994 corresponds to lower precipitation over Japan during these years. In addition higher precipitation (10 to 20% higher) over the upwind Chinese emission areas (mainly Shandong, Hebei and Southern Heilonjiang) also caused less pollutant to be available for long-range transport and thus resulted in a lower contribution of Chinese sources to Japan and South Korea during this period. The opposite phenomenon can be observed during 1992 and 1995-97, when lower precipitation over North-East China enhanced the long-range transport of pollutants in these years. This is recognizable in the peak in deposition over Japan and Korea during 1992 and in the enhanced contribution of Chinese emissions to the deposition at these countries during 1995-97.

Table 2. Evidence of long-term trends in available measurements.

Country	**Measurements**	**Period**	**Trend over the period**	**Reference**
China	SO_4 in precipitation	1986-1994	+60 in Chongquing	Bai *et al.*, 1997
China	SO_4 wet deposition	1988-96	+50% increase in the late eighties; maximum in early nineties, then decreasing to previous levels	Hong Kong Env. Protection Dept., 1998
Indonesia	SO_4 wet deposition	1992-96	+20% in Jakarta	Gillett *et al.*, 1999
Malaysia	SO_4 in precipitation	1985-1994	from +10% in internal areas of Malay peninsula to +100% on the West coast	Malaysian Meteorological Service, 1995
S. Korea	SO_2 concentrations	1988-92	-40% in Seoul and –25% in Pusan	Korean Ministry of Environment, 1998
S. Korea	SO_4 wet deposition	1994-96	-60% in rural areas in the northern part of the country	Korean Ministry of Environment, 1998

The influence of precipitation anomalies can also be seen in the sulfur deposition trends in Shandong, a region which is dominated by local sources and from pollutants coming from surrounding Chinese regions. Enhanced deposition in 1990 and reduced deposition in 1992 and '97 correspond to positive and negative precipitation anomalies, respectively.

In Taiwan (Figure 5), after a substantial increase in emissions between 1985 and the early nineties, emissions have decreased, and are now below 1985 levels. However sulfur deposition, while now relatively stable, is higher than that in 1985. During the nineties the contribution of Chinese emissions to Taiwan deposition increased from 17% to 33%, accompanied by a corresponding decrease from 76% to 60% of the indigenous contribution. The effects of precipitation variability are also visible: the peaks in depositions in 1992 and 1994 correspond to positive anomalies of precipitation over the country, while the lower values of 1990, 1991 and especially 1993 coincide with negative anomalies.

Sulfur deposition in Hong Kong (Figure 5) is dominated by local sources (Hong Kong and surrounding regions), as is the case of Chongqing (Sichuan, China; Figure 3). In the case of Hong Kong deposition reaches a maximum in the early nineties, then slightly decreases and then rise again in 1996-1997 according to the emission trend. For Chongqing sulfur deposition increased throughout the simulated period. These trends are generally consistent with the available monitoring data (Table 2). Sources from the FSU dominate Mongolian deposition, with a contribution to national balance as high as about 60% in 1990, and progressively diminishing in the following years due to an increased Chinese influence. For other Asian countries in the domain the contribution of FSU sources is generally not significant at the level of national balance, but can be locally important. This happens in the northern part of the domain, and is enhanced where local emissions are low, emphasizing the role of the long-range transport. As far as China is concerned, FSU emissions account for from 20% to more than 60% of total sulfur deposition in the Western part of the country, and also affect significantly the deposition along the Northeastern frontier. The contribution of FSU sources to deposition in Northern Japan can account for from 5% to 20% of total sulfur deposition

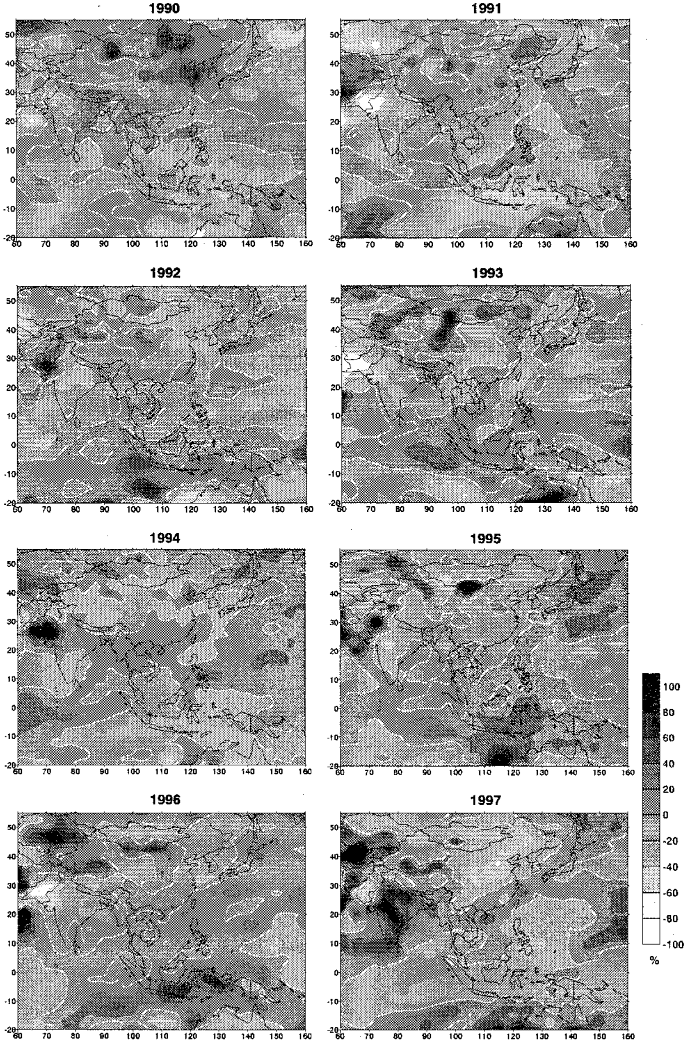


Figure 4. Yearly precipitation anomalies, normalized with respect to the average of 1990-97 values

(2) Southeast Asia

Sulfur deposition in Thailand (Figure 5) strongly increases during the simulated period, due to the very large contribution of indigenous emissions, which have grown significantly during this period. Thailand emissions are significantly larger than those of the surrounding countries, so they influence the behavior of sulfur deposition throughout Indochina. Thailand is the dominating contributor to sulfur deposition in Laos and Cambodia and the second and third largest contributor to Burma and Vietnam, respectively. A typical example is given by Cambodia, where the year to year variation of total depositions over the country is mainly determined by the Thai contribution, oscillating from 47% to 63%.

Malaysian deposition also increased appreciably during 1985-1997, and closely follows the trend in the emissions of the two main contributors: Malaysia itself and Singapore. A progressive increase of the Malaysian contribution is shown, as well as a peak in the Singapore contribution corresponds which to the peak in Singapore's emissions in 1994. This predicted upward trend in sulfur deposition is also revealed by measurements which show that the concentration of sulfate in precipitation and wet deposition over the Malay peninsula have clearly increased during 1985-1994 (Table 2).

Sulfur deposition in Indonesia has increased dramatically, due to the large contribution of indigenous sources (about 73% in year 1990) and a growth rate in emission substantially higher than that in Malaysia. This is confirmed by measurements taken in Jakarta (Table 2).

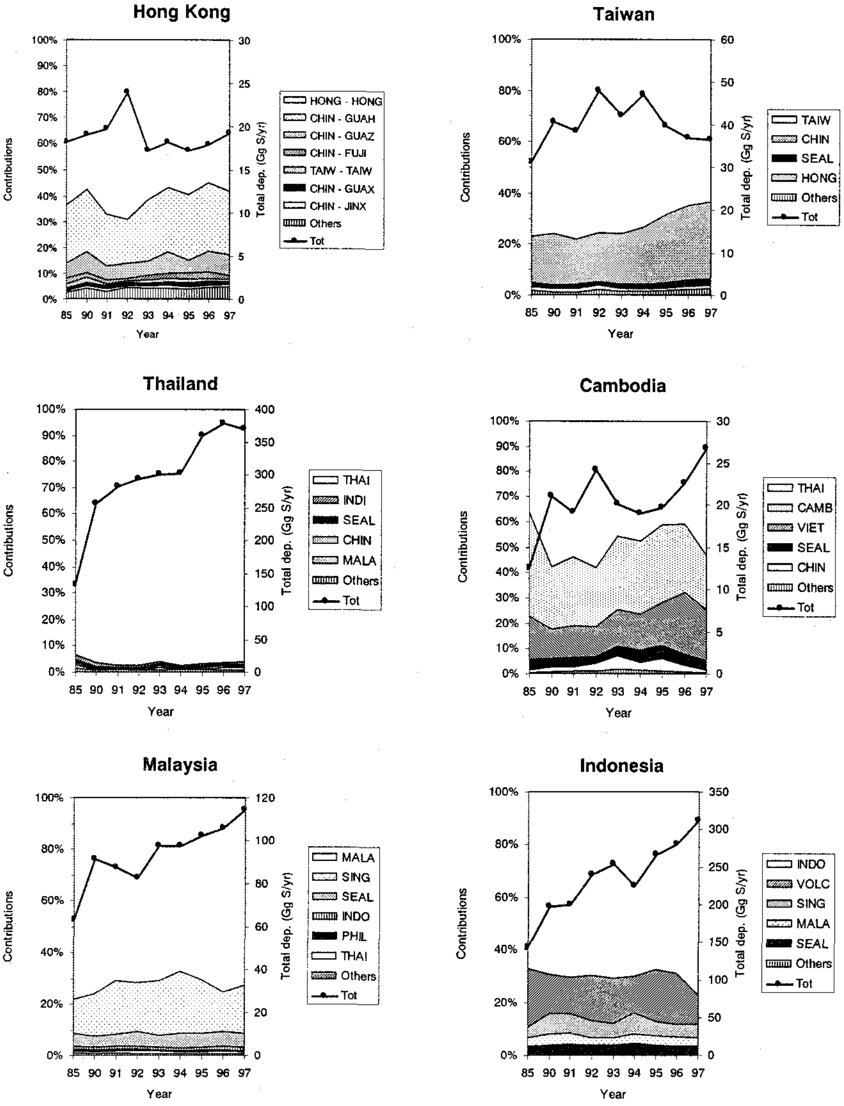


Figure 5. Same as in Figure 3, but for countries and regions of Southeast Asia. Regions codes: FUJI= Fujian; GUAH = Guangdong and Hainan; GUAX = Guangxi; GUAZ = Guagzhou; JINX = Jianxi;i; ZHEJ = Zhejiang

An important role in the deposition over the country is played by volcanic emissions, which contribute between 15% and 20% to total deposition. Locally share of volcanic sources can be much larger, as in central Java (from 40% to 60%) and especially in Eastern Irian Jaya (from 80% to more than 95%), where anthropogenic sources are very low. Sulfur deposition in Indonesia also show a strong correlation with precipitation anomalies, due to the high incidence of local sources(anthropogenic and volcanic) on depositions. The effect can be clearly observed especially in years 1990, 1993 and 1996 (slightly positive anomalies) as well as in 1991, 1994 and 1997 (marked negative anomalies, in correspondence to El Niño events).

Finally, in both Malaysia and Indonesia there is also a significant contribution of emissions from shipping activities, due to the heavy maritime traffic in the Strait of Malacca and in the South China Sea. Countrywide, shipping emissions contribute between 4 and 6% (Streets *et al.*, 2000). Locally the contribution due to ship emissions can be as high as 20%, as shown along the coastal areas of the Malay Peninsula, Sarawak, Southern Borneo and Sulawesi.

(3) Indian subcontinent

In most parts of India sulfur deposition is dominated by national sources, as evidenced by the balances for Maharashtra and Bihar regions, shown as examples in Figure 6. Again, the link between precipitation and deposition trends is noticeable. The strong enhancement of deposition trends by the 1997 precipitation anomaly is especially evident in both regions. On other years although, the depositions-precipitation link is less pronounced than the one observed in other Asian regions, at least on a yearly basis. This is probably due to the strong seasonality in precipitation caused by the Indian monsoon and is currently the subject of further investigation.

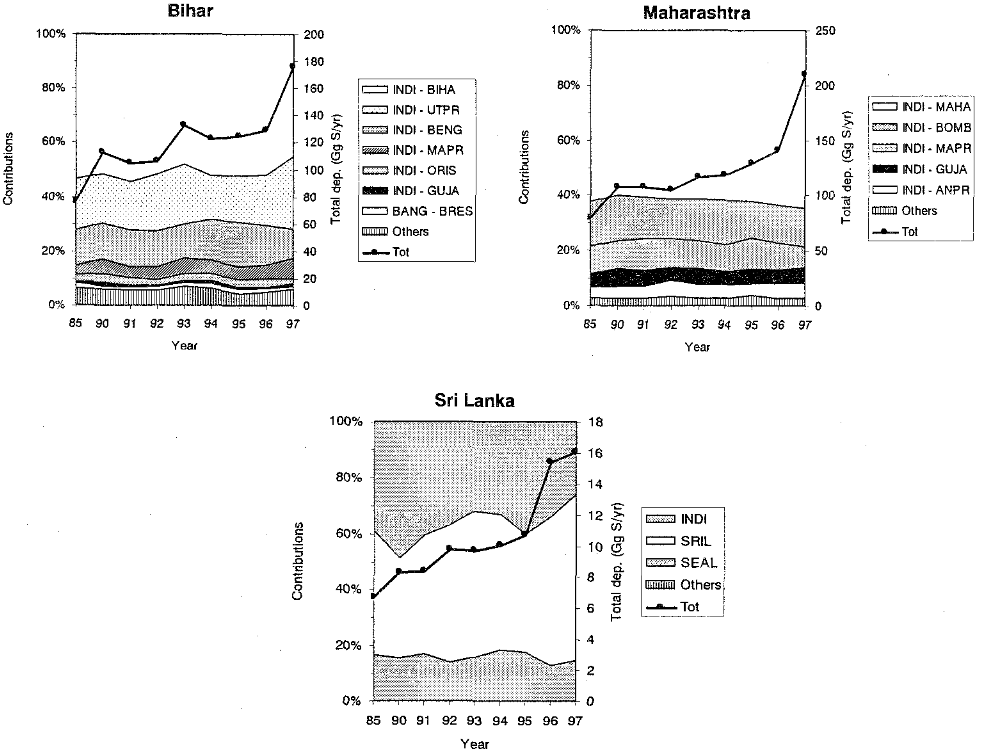


Figure 6. Same as in Figure 3, but for countries and regions of Indian subcontinent. Regions codes: BIHA = Bihar; UTPR = Uttar Pradesh; BENG = West Bengal; MAPR = Madhya Pradesh; ORIS = Orissa ; GUJA = Gujarat; BRES = Bangladesh (except Dhaka); MAHA = Maharashtra; BOMB = Bombay; ANPR = Andhra Pradesh

Indian emissions also heavily influence the deposition of the surrounding countries: India is by far the largest contributor to depositions in Nepal, Bhutan, Bangladesh, and Burma, and also play an important role in the sulfur budget of Sri Lanka. The impact of the large precipitation anomaly of 1997 is also evident in the behavior of Indian and indigenous contributions in the deposition balance of Sri Lanka. During 1994 and 1995 the mentioned contributions follow the respective trends of emissions (increasing in the case of India and slightly decreasing in the one of Sri Lanka). In 1997 stronger precipitation throughout the area enhanced the local deposition over the source regions, depressing the long-range transport, and thus leading to a reversal in the contribution tendencies

Sulfur deposition in Sri Lanka is also impacted by emissions from maritime traffic (about 18% of total sulfur deposition over the country). Locally in various points of the domain the contribution can be even greater, but even when aggregated it is noticeable in the national budgets of different countries of Southeast Asia: Cambodia, Indonesia, Malaysia (about 4% of total depositions), Philippines, Singapore (about 3%) and Brunei (7%).

3.2 Inter-annual meteorological variability

(1) Deposition levels

To estimate the magnitude of the influence of inter-annual meteorological variability on deposition levels, year-by-year depositions were also computed using fixed emission levels. As a reference, emissions for the year 1990 were used for the whole thirteen year period. Year 1990 is the baseline year in RAINS-Asia model calculations and was used in previous modeling studies. Results are summarized on a country basis in Figure 7. Shown are the year-by-year relative deviations of country total depositions with respect to the average sulfur deposition over all years. The Indian subcontinent shows the highest inter-annual variability, with yearly country total depositions varying by more than 20% from the average. Southeast Asia exhibits the lowest variability, with yearly deviations from the average levels generally below 10%. Indochina and North and East Asia show an intermediate behavior.

The analysis in term of national balances can be influenced by the size of each country, so a map of the standard deviation of the yearly deposition fields, normalized by the local average depositions, is presented in Figure 8. Local differences in the deposition variability within countries can be in this way appreciated. When examined against the corresponding map of average deposition levels, computed with fixed emissions, it is found that the variability is generally lower for regions with high depositions. As for local variations within India, higher and lower variability areas can be located in the central part of the country and East Himalaya. Within China, higher variability areas are in the north-eastern and western parts of the country, as well in Shanxi and Shaanxi to a lesser extent. Variability is also higher in the northern part of Japan than in the rest of the country, and in parts of Indonesia.

As already pointed out in the different country and regional balances, and also demonstrated by Arndt *et al.* (1998) on a seasonal basis, precipitation variability can play an important role in determining the deposition variability throughout Asia. A map of the standard deviation of yearly precipitation, normalized to the average over the thirteen year period, is also presented in Figure 8. Although the relationship between precipitation and deposition variability can not be assumed as general, there is a good correlation between depositions and precipitation variability, especially in the areas where the influence of local sources is high. This confirms some of the analysis illustrated above at the country level. The effects of large precipitation anomalies of 1997 in particular, a strong El Niño year, are evident in the depositions of many countries, especially in the Indian subcontinent, Indochina, Southern China and part of Southeast Asia.

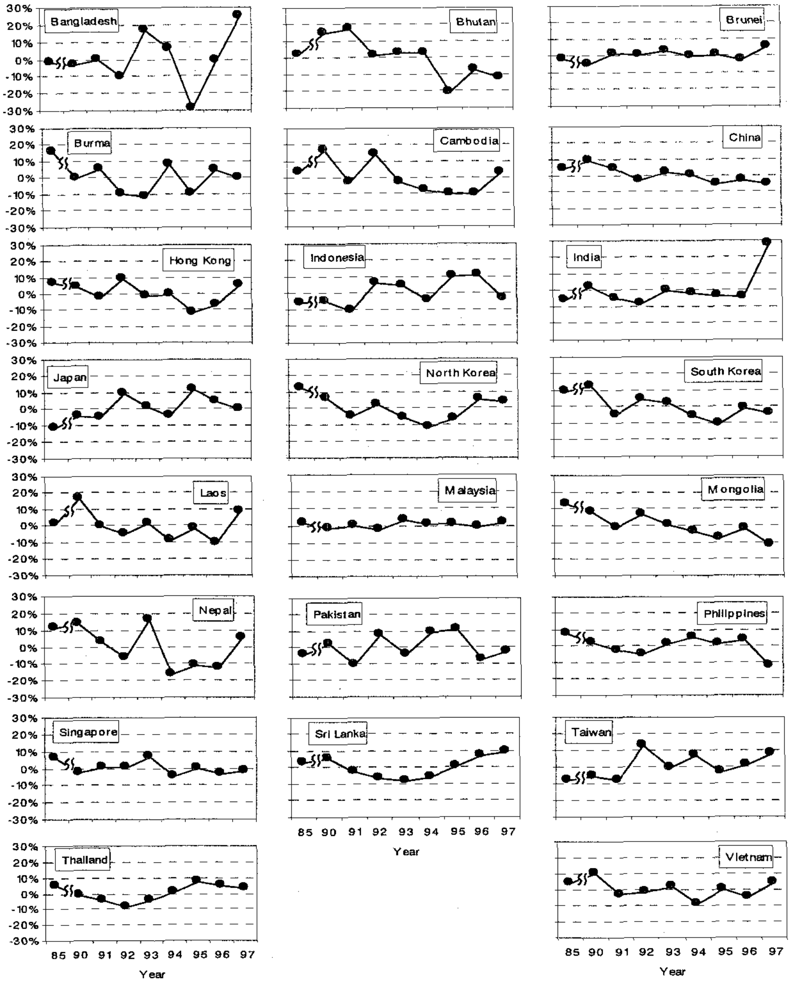
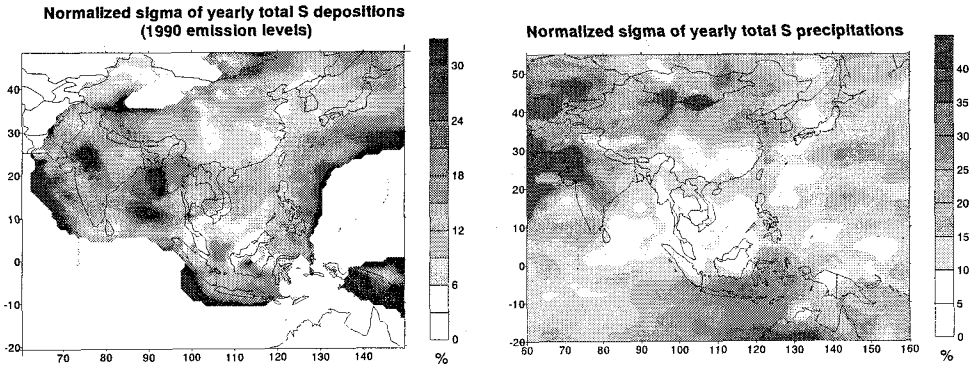


Figure 7. Year-by-year relative deviation of country total depositions, normalized to 1990 emissions levels


Normalized sigma of yearly total S depositions (1990 emission levels)

Normalized sigma of yearly total S precipitations

Figure 8. (a) Standard deviation of yearly total depositions calculated with 1990 emission levels, normalized respect to the average of 1985-97 values; (b) Standard deviation of yearly total precipitation, normalized with respect to the average of 1985-97 values

During the 1990-97 period the normalized total depositions of some countries also exhibit an apparent systematic tendency, underlying the year-by-year meteorological variability. This affects in miscellaneous ways the actual total deposition trends in various countries. In some cases there is a downward tendency that can partially mask the increase in deposition during the years that would result by emissions trends only. This is the case in Bhutan, Cambodia, China, Laos and Nepal. Elsewhere a similar downward tendency can enhance the already existing downward trend in deposition caused by decreasing emissions of the most influential contributors, as in the case of South Korea. Elsewhere, as in Mongolia, the apparent meteorological tendency can even cause a downward trend, where deposition based on emissions only would have otherwise been almost stationary throughout the years. For other countries the tendency due to meteorology plays to the opposite side, enhancing the upward trend of total depositions, as in the case of Indonesia and Japan. This result highlights the complex role played by the meteorology on deposition levels. For these reasons the trends exhibited by the emissions may not necessarily be reflected in the medium term by corresponding tendencies in depositions, and so not clearly revealed by monitoring activities.

(2) Stability of source-receptor relationships

In view of the use of the simulation results in integrated assessment frameworks, such as the RAINS-Asia model, it is interesting to analyze the inter-annual variability of source-receptor relationships. Specifically, we have focused on national balances, looking at how the contributions of emissions from different countries can vary from one year to another due just to the variations in year-by-year meteorology. This has been quantified by taking the year-by-year depositions computed with reference emissions for the year 1990, and calculating national balances in terms of percentage contributions of emissions from different countries. A variability index was computed for each contribution, as the standard deviation normalized to its average value over the years. The results for all countries are shown in Figure 9.

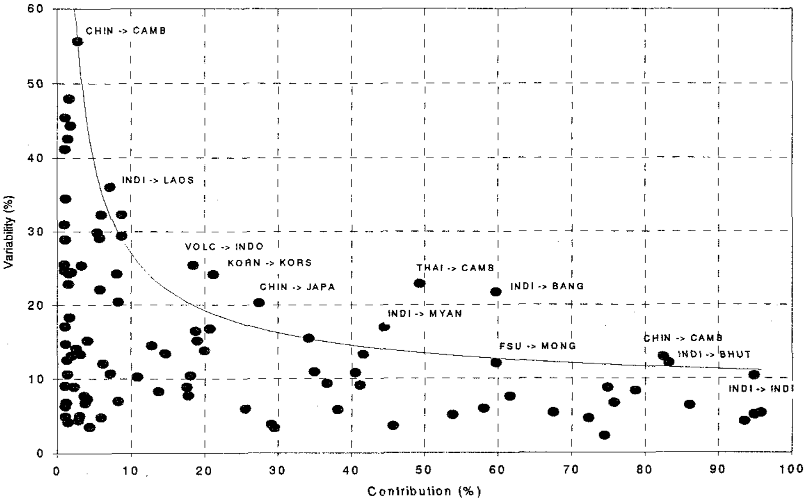


Figure 9. Variability of yearly country-to-country contributions to country total depositions, with constant emission levels; horizontal axis: average yearly contribution to country total depositions (%); vertical axis: contribution variability (as % of contribution).

The variability of small contributions (the ones between 1% and 2% of the national balances) can be large (as high as 40% of their magnitude), while for the largest contributions the variability is typically low. As shown the variability is less than 17% when contributions are in the 10-50%

magnitude class and less than ~10% for the "dominant" contributions (the ones contributing more than 50% to national balances). Notable exceptions are found for India to Bangladesh, Thailand to Cambodia, China to Japan, North Korea on itself and for volcanic sources to Indonesia, where variabilities are in the 20-25% range. In general the year-by-year variability decreases as the relevance of the contributions increases. Furthermore, even at the country-to-country level the variability are on the order of 10-20%, and this fact should be taken into account when source-receptor relationships are used for impact assessment purposes.

4. Conclusions

A multiple year modeling study was conducted for sulfur deposition in Asia. The period of study, spanning thirteen years, is sufficient to show the effects of major emission trends and the year-by-year meteorological variability on regional sulfur deposition. Most of the areas have experienced dramatic increases in deposition levels, with values almost doubled in North-central India and North-central Indochina, parts of South and West China and along the Chinese-Mongolian border. In the rest of China sulfur deposition has increased between 10% and 30%. There has been a decrease in some countries of East Asia (South Korea, Southwest Japan, part of Taiwan and Philippines).

The trends in sulfur deposition were found to be influenced by year-to-year variability meteorological factors, especially precipitation. Yearly precipitation anomalies can partially mask the trends in sulfur deposition expected as a consequence of changes in the emission only. It was also shown that in regions where sulfur deposition is dominated by local sources there is a positive correlation between yearly precipitation and deposition anomalies, whereas in regions with large contributions from long-range transport, precipitation anomalies can sometimes play in the opposite direction.

The magnitude of the signal related to inter-annual meteorological variability was estimated on a country basis and found to be between 10% in Southeast Asia and more than 20% in the Indian subcontinent, with intermediate values in Indochina and North and East Asia. The effects of inter-annual variability in meteorology on country-to-country source-receptor relationships were also evaluated. It was found that the relationships of large source contributions (>30%) to a receptor are less sensitive to inter-annual variability (<10%) than that for the smaller ones; but with some notable exceptions (e.g. India to Bangladesh). This variability should be considered when source-receptor relationships are included in integrated assessment analysis.

These results also demonstrate the need for long term measurements to document changes in emissions and deposition in the region, and the critical role of models in helping to identify the simultaneous influence of meteorology and emissions on the resulting trends.

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