Application of a LRT Model to Acid Rain Control in China[†]

JIMING HAO,* LEI DUAN, XUELONG ZHOU, AND LIXIN FU

Department of Environmental Science and Engineering, Tsinghua University, Beijing 100084, P. R. China

For further control of acid rain and SO_2 pollution in China, acid rain control zones and sulfur dioxide pollution control zones were designated where acid rain or serious SO_2 pollution occurs or may occur. In this study, sulfur deposition in east China was computed through a policy-oriented, two-dimensional Eulerian model for long-range transport and deposition of SO_2 and SO_4^{2-}. The model predictions were in accordance with the wet deposition monitored. Results show that concentrations of SO_2 and SO_4^{2-} are higher in north China than those in the south, and high deposition of sulfur occurs in most areas of North China, in the lower reaches of the Changjiang (Yangtze) River and around Chongqing and Guiyang in southwest China. Total emission of SO_2 from the modeling region (from 19°N to 42°N, and from 104°E to 124°E) was about 20 million tons in 1995. The model predicts that 48% of this deposits within the region as dry deposition, 38% deposits as wet deposition, and only about 14% was transported out of the region. The modeling results of sulfur deposition were directly applied in designating acid rain control zones in China, and the emission−deposition relationship derived was also used to formulate middle- and long-range planning programs for regional acid rain control in China.

1. Introduction

Accompanying the increasing emission of sulfur dioxide in the past decades, acid rain has occurred in large areas of China. Anthropogenic SO_2 emissions from China were about 18.4 Mt a^{-1} in 1990 and reached 23.7 Mt a^{-1} in 1995 (*1*). The region suffering from acid rain has extended northwards from small areas around Chongqing and Guiyang in southwest China in the early 1980s to the whole of eastern China, covering 12 provinces at present. The statistical results from the acid rain survey in 82 cities from 1991 to 1995 also indicated that the annual average pH value of the precipitation was lower than pH 5.6 in nearly half of these cities, with the lowest reaching a pH 3.52 in Changsha of Hunan province (*1*).

The government of China attaches great importance to the pollution caused by acid rain and ambient sulfur dioxide. For further control of acid rain and SO_2 pollution, the National People's Congress amended the Law on the Prevention and Control of Atmospheric Pollution in August 1995, to entitle the State Environmental Protection Administration (SEPA) to designate acid rain control zones and sulfur dioxide pollution control zones (called the two control zones for short) for those areas which are or could become affected by acid deposition or ambient sulfur dioxide pollution. More stringent emission standards will be implemented in the control zones, and thermal power plants and other industries within these areas must take further measures to abate their SO_2 emissions. Accordingly, the industrial layout and energy structure in these zones should also be rearranged soon.

To designate the acid rain control zones, the geographical distribution of concentration and deposition of acidifying substances must be primarily considered, so modeling the long-range transport and deposition of sulfur is of practical significance. Since the 1970s, a number of models designed for different uses have been applied to Europe and North America (*2*−*6*). Although some modeling studies have been carried out in Asia (*7*−*9*), long-range transport and deposition in Asia, especially in China, is less fully understood. From 1986 to 1995, Chinese researchers also developed several models (*10*−*13*). These research-oriented models can give detailed descriptions of the physical and chemical processes in long-range transport and deposition, but may be too complex to be applied in policy-making. Consequently, a simple and practical, policy-oriented model for long-range transport and deposition of sulfur compounds was developed in this study to assess the control scenarios rapidly and to implement the regional control strategy of acid deposition scientifically and efficiently. This two-dimensional Eulerian statistical model predicts the distribution of sulfur deposition in east China, and the results are used in the designation of the acid rain control zones and the sulfur dioxide pollution control zones in China.

2. Model Performance

The detailed description of the two-dimensional Eulerian statistical model is presented in the Supporting Information. To evaluate the performance of the model in estimating sulfur deposition in China, the sensitivity of the model was critically analyzed and the results of the model were compared with monitoring data and those of other models.

2.1 Model Sensitivity. The sensitivity of the model to the variation of parameters was analyzed to demonstrate the rationality of the parameter values. The relative deviation of modeling result was calculated while each parameter was increased or reduced by 25% and others kept unchanged. The sensitivity of the model to each parameter is shown in Table 1. The symbol ± before the numbers indicates that the change is always positive/negative in the whole definition domain, and the number without the symbols means that the variation is not always positive or negative while the parameter changes. As can be seen from the table, the wet deposition coefficient is the most important factor in determining the distribution of SO_2/SO_4^{2-} concentration in China. Although the wet deposition is quite sensitive to the mixing height, the wet deposition coefficient and the chemical conversion rate, the total sulfur deposition, which is the most important output of the model, shows little sensitivity to each parameter (with the deviation resulting from a 25% change in any one parameter being no more than 9.0%).

2.2 Result Intercomparison. The long-range transport and deposition model was evaluated by comparing the wet deposition estimated with the monitoring results from 261 stations in 1992 to 1993 (*14*) (see Figure 1). It shows a reasonable agreement with a correlation coefficient of 0.80.

[†] This project was supported by the National Natural Science Foundation of China.

* Corresponding author. Phone: 86-10-62782195. Fax: 86-10-62773650. E-mail: hjm-den@tsinghua.edu.cn.

10.1021/es001888u CCC: $20.00 © 2001 American Chemical Society
Published on Web 07/07/2001

TABLE 1. Sensitivity of Model Results to Parameter Variation

variation of parameter (%)		deviation of model results[a] (%)								
		conc		dry deposn		wet deposn		sulfur deposn		
		SO_2	SO_4^{2-}	SO_2	SO_4^{2-}	SO_2	SO_4^{2-}	SO_2	SO_4^{2-}	total
V_{d,SO_2}	+25	−5.6	−3.4	+16.7	−3.6	−5.4	−3.7	+7.8	−9.9	1.9
	−25	−6.8	+4.2	−18.8	+4.3	+6.9	+3.8	−9.4	+13.7	2.0
$V_{d,SO_4^{2-}}$	+25	0	−3.5	0	+19.7	0	−3.9	+2.4	−2.3	0.5
	−25	0	+4.4	0	−20.8	0	+4.0	−2.5	+2.9	0.3
K_{w,SO_2}	+25	−18.2	−11.3	−17.8	−10.5	8.2	−11.5	−4.7	3.9	6.6
	−25	+29.5	+16.7	+26.5	+14.9	12.5	+16.2	+5.1	4.5	9.0
$K_{w,SO_4^{2-}}$	+25	0	−20.9	0	−19.2	0	8.5	−2.6	4.1	3.8
	−25	0	+36.7	0	+30.6	0	12.1	+3.9	6.9	5.1
K_c	+25	−4.8	+15.3	−4.6	+14.9	−4.7	+15.0	−7.3	+5.3	1.6
	−25	+5.8	−16.3	+5.1	−15.8	+5.8	−16.6	+9.3	−6.8	1.4
α	+25	−2.7	−2.9	−2.7	−2.9	−2.8	−2.9	2.7	−2.8	−2.9
	−25	+2.8	+3.2	+2.8	+3.3	+2.8	+2.9	2.8	+3.0	+2.7
β	+25	−1.4	+4.4	−1.3	+4.9	−1.4	+4.2	−1.0	0.5	0.4
	−25	+1.4	−4.3	+1.3	−4.3	+1.4	−4.4	+1.0	0.6	0.5
H	+25	−10.5	−10.2	−8.8	−10.2	+13.2	+12.5	−8.5	+13.0	4.8
	−25	+14.3	+12.9	+11.5	+12.7	−16.0	−16.1	+11.5	−15.8	6.4

[a] Maximum absolute value over the study area.

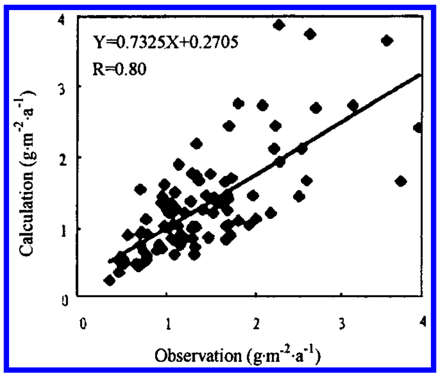

FIGURE 1. Comparison of wet depositions calculated by the model with those observed by the monitoring network for acid rain (1992−1993).

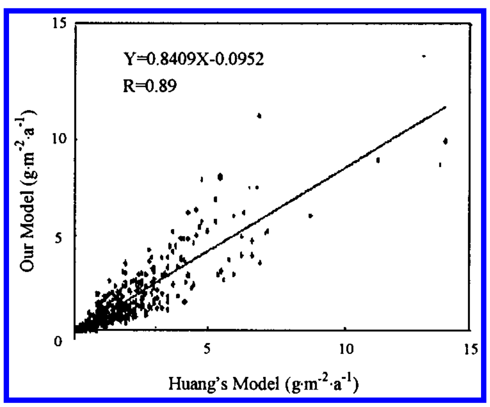

FIGURE 2. Comparison of total depositions calculated by the model with those calculated by the model developed by Huang et al. (1995).

According to the limited availability of emission data, wet deposition in 1990, instead of that in 1992 or 1993, was modeled. (Monitoring had not been widely carried out before 1992.) Since the wet deposition of sulfur estimated is the mean value within each grid, the measured data from several monitoring stations in the same grid should be averaged. The reason the estimated values are a bit lower than those measured may be that emissions increased somewhat from 1990 to 1992−1993, particularly in urban areas where some observation stations are located.

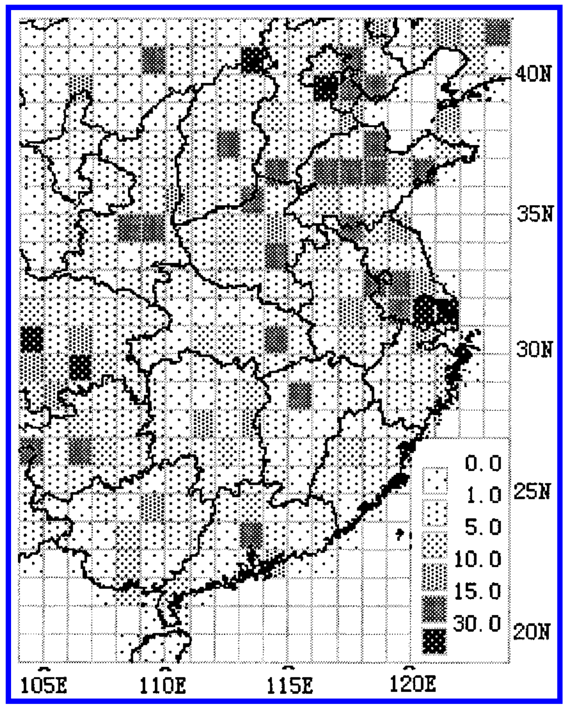

FIGURE 3. SO_2 emission intensity in east China in 1995 (t km^{-2} a^{-1}).

A comparison was also carried out between different models. A scatterplot diagram for sulfur deposition calculated by the regional model, and a more complex model developed by Huang et al. (*8*) is shown in Figure 2. As can be seen, the results of the two models were in good agreement, with a correlation coefficient of 0.89. On the basis of the comparison to observed data and the more complex model, the performance of the regional model seems to be satisfying.

3. Modeling Sulfur Deposition in East China

3.1 Input Data. The modeling region contains 19 province/municipalities including Beijing, Tianjin, Hebei, Shanxi, Shaanxi, Henan, Shandong, Anhui, Jiangsu, Shanghai, Zhejiang, Fujian, Jiangxi, Hubei, Hunan, Guangdong, Guangxi, Sichuan, and Guizhou. The emission intensity of SO_2 in 1990 and 1995 (data for 1990 only for model test, see 2.4) was extracted from the database of emission sources established by the Chinese Research Academy of Environmental Sciences.

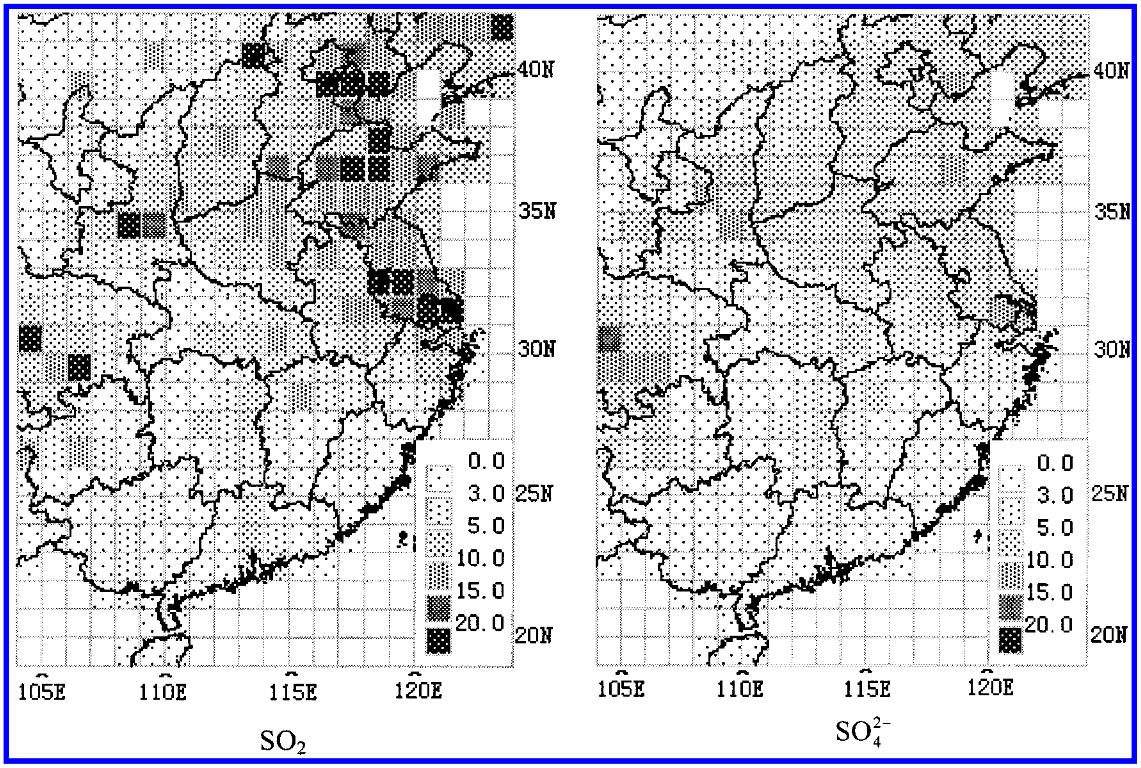

FIGURE 4. Distribution of calculated annual average concentration in east China in 1995 (μg m^{-3}).

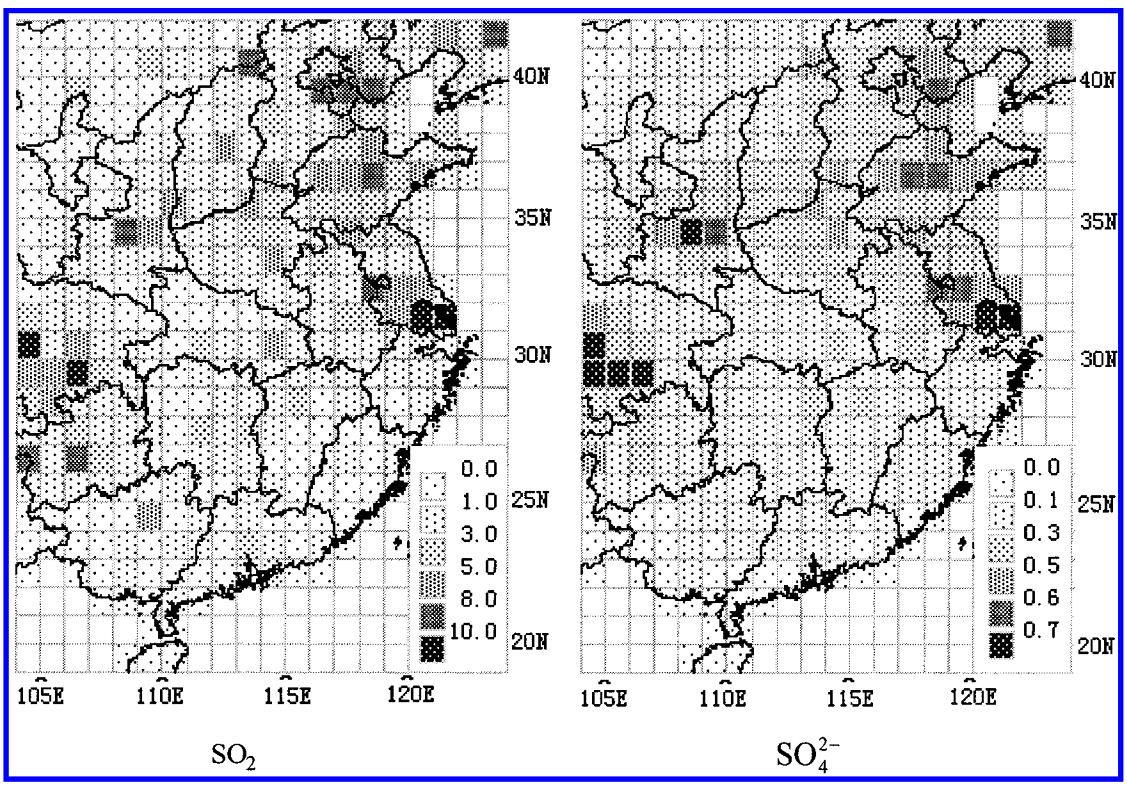

FIGURE 5. Distribution of calculated dry deposition in east China in 1995 (g m^{-2} a^{-1}).

According to the national program of economical development and assumed energy consumption, SO_2 emissions in 2000 were predicted (*15*).

To derive the emission intensity for each 1° × 1° grid, point sources, which are mainly large power plants, were separated from the area ones. Emissions of each area source are allotted to every cell based on how much of the source is within the grid, while the emission from a point source is directly added to the cells in which the point sources are located. The emission intensity in the modeling region estimated for 1995 is shown in Figure 3. As can be seen, a high intensity of SO_2 emission, i.e., over 15 t km^{-2} a^{-1}, occurs in coastal areas near the Bo Sea and the Yellow Sea, and in the grids where large cities such as Wuhan, Chongqing, Shanghai, and Guiyang are located. The total SO_2 emission in the modeled region is about 20 million tons in 1995, about 85% of the estimated total emission for the whole country (23.7 million tons).

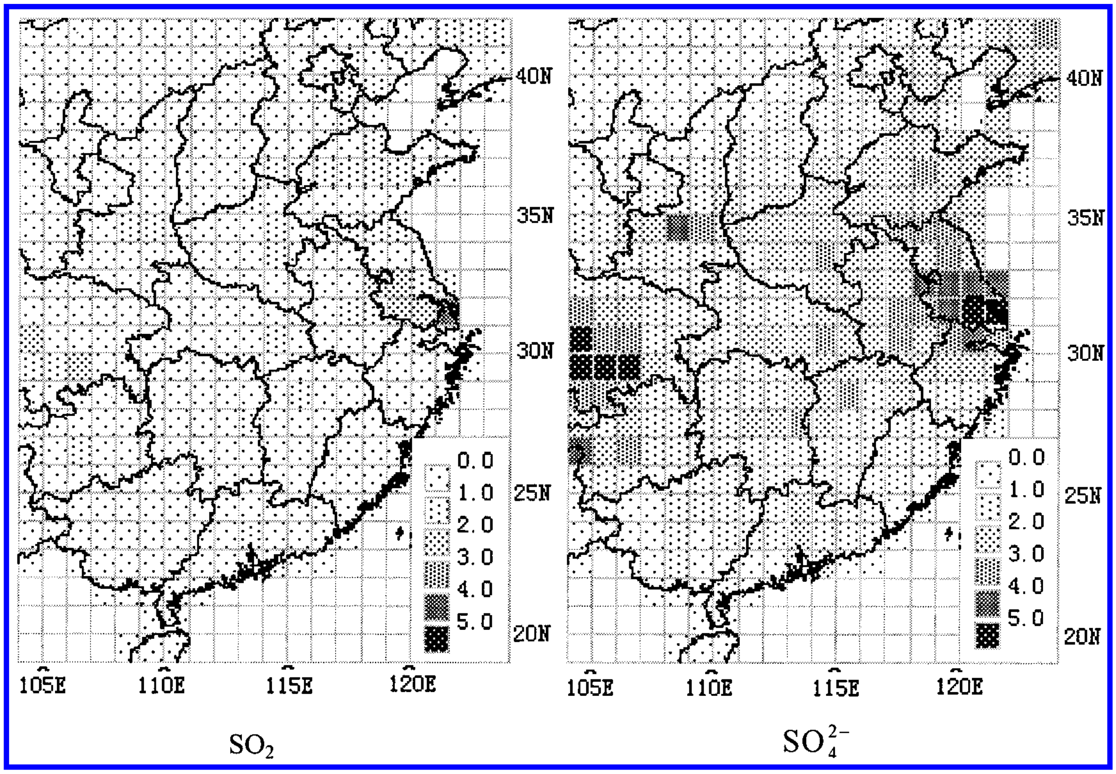

FIGURE 6. Distribution of calculated wet deposition in east China in 1995 (g m^{-2} a^{-1}).

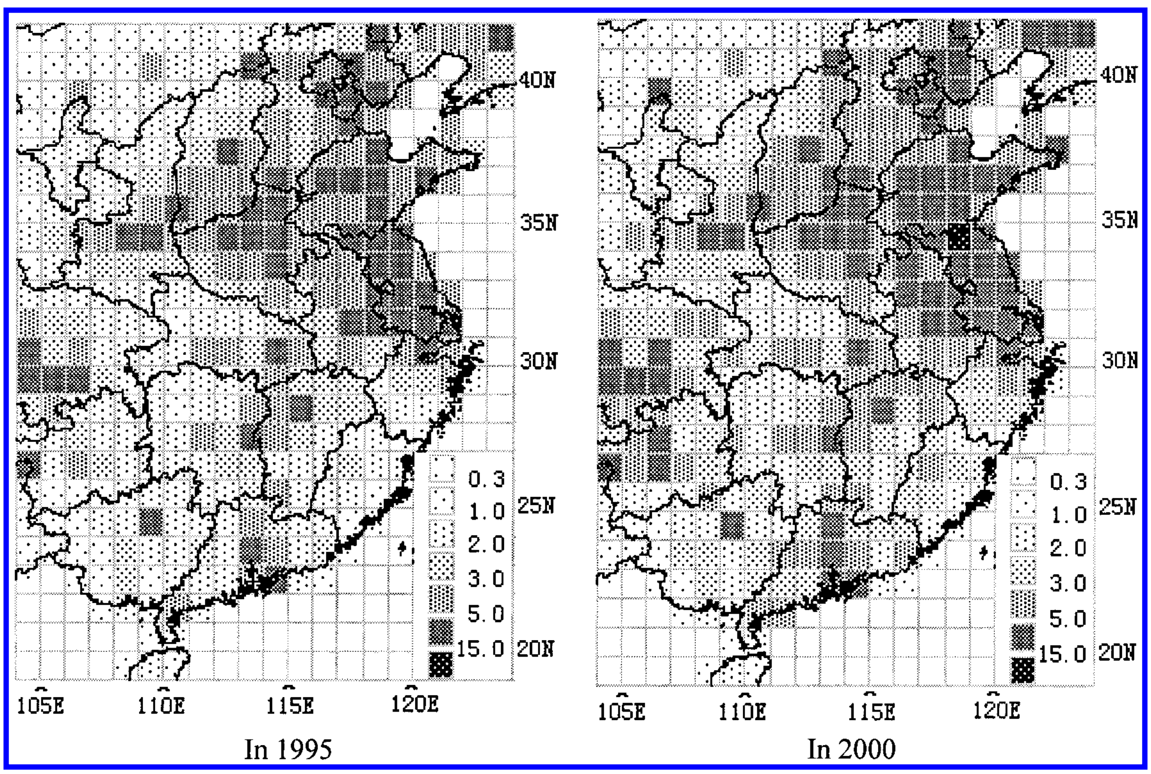

FIGURE 7. Calculated sulfur deposition in east China in 1995 and 2000 (g m^{-2} a^{-1}).

Meteorological data were obtained from two sources. The precipitation data were obtained from the China Meteorological Administration and other model inputs (wind fields and humidity) from the ECMWF (European Center for Middle-range Weather Forecasts). The original data were converted to the necessities of model input by approximate interpolation. To reduce the influence of yearly variation of meteorological condition on model results, average values in five years (from 1991 to 1995) were applied to predict sulfur deposition in 2000.

3.2 Results and Discussion. *3.2.1 Concentration.* Figure 4 shows the estimated distribution of the annual average concentration of SO_2 and SO_4^{2-}. As can be seen, high concentrations of SO_2 occur in grid cells where large cities, such as Shanghai, Beijing, Chongqing, Xi'an, and Qingdao are located, and the highest predicted value even reaches 26

TABLE 2. Transfer Matrix between the Modeling Provinces[a] Indicating the Partion of the Sulfur Emission from the Source Region that Deposits in the Receptor Area[b]

R/S	GD	GX	FJ	JX	HuN	GZ	ZJ	SH	JS	AH	HuB	HeN	SD	HeB	BJ	TJ	SX1	SX2
GD	619	30	56	31	31	0	2	0	0	1	4	0	0	0	0	0	0	0
GX	137	480	3	7	40	40	0	0	0	0	3	0	0	0	0	0	0	0
FJ	26	0	588	36	5	0	38	5	6	10	2	0	0	0	0	0	0	0
JX	62	4	116	529	107	0	40	5	10	40	65	6	3	1	0	0	0	0
HuN	72	61	10	105	635	11	3	0	2	6	67	9	0	0	0	0	1	2
GZ	4	127	0	3	37	639	0	0	0	0	5	0	0	0	0	0	0	2
ZJ	2	0	59	15	3	0	516	81	58	55	8	2	4	1	0	1	0	0
SH	0	0	0	1	0	0	24	186	27	6	0	0	1	0	0	0	0	0
JS	0	0	5	9	1	0	68	295	461	178	9	26	72	14	9	13	10	4
AH	2	0	20	93	9	0	65	28	104	476	50	51	32	9	5	6	8	5
HuB	5	4	2	76	92	2	5	4	10	42	653	87	5	5	1	1	11	32
HeN	0	0	0	13	9	0	1	3	19	71	83	524	33	38	2	5	67	62
SD	0	0	0	4	1	0	4	8	74	54	7	72	521	102	55	87	55	10
HeB	0	0	0	1	0	0	0	0	5	8	4	35	59	424	333	281	184	9
BJ	0	0	0	0	0	0	0	0	0	0	0	2	3	34	186	15	16	0
TJ	0	0	0	0	0	0	0	0	0	1	0	2	8	25	44	214	10	0
SX1	0	0	0	0	1	0	0	0	2	4	9	112	13	59	7	5	426	77
SX2	0	0	0	0	2	2	0	0	1	3	11	60	3	5	0	0	45	578
Total	929	707	858	923	972	695	764	613	778	955	977	988	756	718	645	629	832	780

[a] Abbreviated: R(Receptors), S(Sources), GD(Guangdong), GX(Guangxi), FJ(Fujian), JX(Jiangxi), HuN(Hunan), GZ(Guizhou), ZJ(Zhejiang), SH(Shanghai), JS(Jiangsu), AH(Anhui), HuB(Hubei), HeN(Henan), SD (Shandong), HeB(Hebei), BJ(Beijing), TJ(Tianjin), SX1(Shanxi), SX2(Shaanxi). [b] Out of a total emission of 1000, i.e., fraction × 1000.

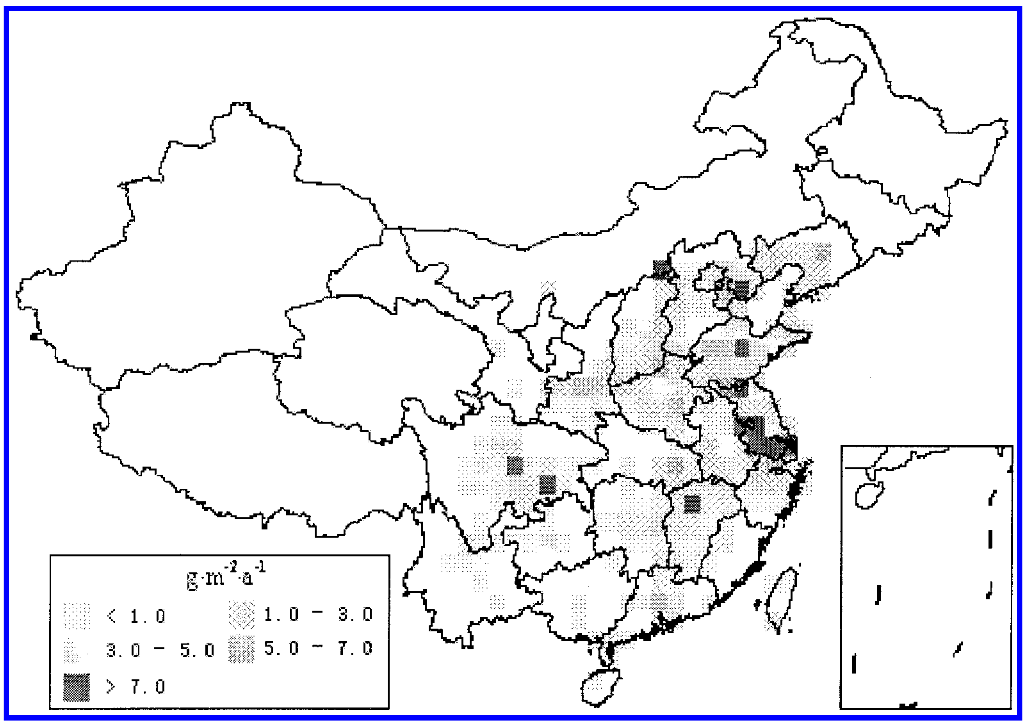

FIGURE 8. Critical load exceedance map of sulfur deposition in China in 1995. Empty cells indicate no exceedance.

μg m^{-3} in Shanghai. As a whole, the annual average concentration of SO_2 in the north is higher than that in the south, which conforms to the fact that extra coal is consumed for heating in the winter in north China.

In contrast to SO_2, the spatial difference of SO_4^{2-} concentration is much less. High SO_4^{2-} concentration always appears tens of kilometers away from large sources and to the leeward of them, because most SO_4^{2-} comes into being through chemical processes during the course of advection. The annual average concentration of SO_4^{2-} in the south is relatively lower than that in the north, too, probably because of the abundant rainfall that washes sulfate out from the atmosphere in the south.

3.2.2 Deposition. Figure 5 and Figure 6 show the geographical distribution of dry deposition and wet deposition, respectively. It can be seen from Figure 5 that dry deposition of SO_2 in most areas is greater than 1.0 g m^{-2} a^{-1}, and the highest even reaches 15.0 g m^{-2} a^{-1}. As compared with dry deposition of SO_2, dry deposition of SO_4^{2-} is much smaller, with the highest value of 0.75 g m^{-2} a^{-1}. Similar to the characteristics of the concentration distributions, the distribution of SO_2 dry deposition is closely related to the distribution of emission sources, while the distribution of SO_4^{2-} dry deposition is significantly affected by prevailing winds.

As indicated in Figure 6, wet deposition of SO_2 is about one-fifth to one-fourth that of dry deposition, but wet deposition of SO_4^{2-} is greater than dry deposition. This indicates that SO_2 in the atmosphere is mainly removed through dry deposition, but SO_4^{2-} is mostly scavenged by

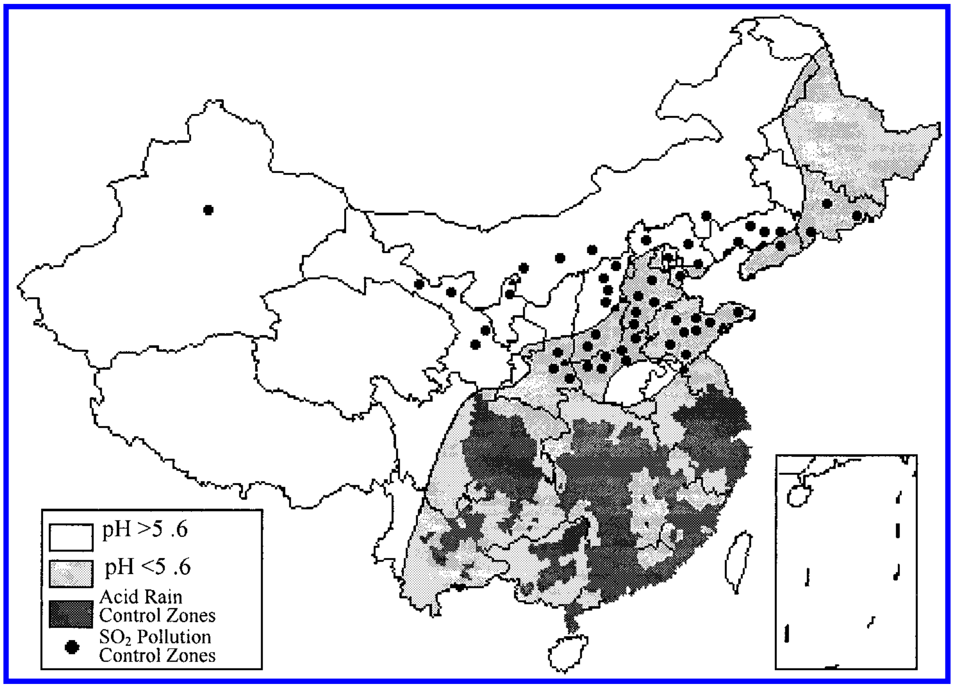

FIGURE 9. Sketch map of the acid rain control zones and the SO_2 pollution control zones in China (Hao et al., 1998).

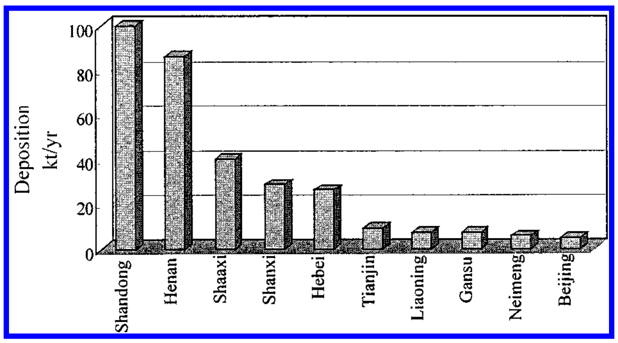

FIGURE 10. Sulfur deposition in the acid rain control zones calculated as coming from provinces outside.

wet deposition. In the modeling region, the calculated wet deposition of SO_4^{2-} is smaller in the north and larger in the south, probably because precipitation increases gradually from the northwest to the southeast. In summary, the distribution of SO_4^{2-} wet deposition depends primarily on the precipitation, while wet deposition of SO_2 is jointly affected by local emission sources and the precipitation.

Combining the dry deposition and wet deposition both for SO_2 and SO_4^{2-} gives the total predicted sulfur deposition in east China (see Figure 7). It can be seen that sulfur deposition is about 1.0−3.0 g m^{-2} a^{-1} in most areas, although there are some grids with high deposition over 10.0 g m^{-2} a^{-1}. High deposition of sulfur occurs in most area of North China, in the lower reaches of the Changjiang (Yangtze) River, and around Chongqing and Guiyang in southwest China.

To predict the future trend of sulfur deposition in China, we also calculate the sulfur deposition in east China in 2000 based on the predicted emission intensity. These results are shown in Figure 7. As can be seen, sulfur deposition in China may continue to increase, and the area suffering from sulfur deposition will expand northward and westward from the east of China to the northeast and northwest, respectively.

3.2.3 Source-Receptor Relationship. In this study, source-receptor relationships for sulfur were also derived (see Table 2). The first row of the table represents the provinces/municipalities as sources, and the first column represents the receptors. As can be seen, except for three municipalities, i.e., Beijing, Tianjin, and Shanghai, about half of the emission from each model province is deposited within its own area, and about 30% is deposited in the adjacent provinces. Since the area of each municipality is relatively small (less than two grid cells), a large part of the sulfur emission (about 50%) is deposited in other provinces, and only about 20% is deposited in its own area.

Table 2 show that nearly all emissions from inland provinces such as Anhui, Hunan, and Hubei are deposited in the modeling region. About 70−80% of the emission from east coastal provinces, such as Jiangsu, Zhejiang etc., is predicted to be deposited in the region. In summary, about 48% of the total emission from the whole region (4.83 Mt sulfur) is deposited within the region as dry deposition, 38% (3.82 Mt) as wet deposition, and 14% (1.35 Mt) is transported out of the region. Among the outflow sulfur, a large part (about 89%) passes through the eastern border of the modeled region. Although most of the sulfur blown out is deposited into the Pacific Ocean, sulfur emission from this region may affect other countries such as Japan and Korea. The contribution of Chinese sources to Japan's deposition has been investigated by some researchers (*7, 8, 16, 17*), but the results are very different. The variations may be partly due to the differences in the removal rates and chemical conversion rates in the different models. Low removal rates result in greater transport away from source locations and thus higher transboundary pollution. Huang et al. (*8*) and Yang et al. (*17*) estimated that China accounts for only 3.5 and 10.0% of Japan's annual sulfur deposition, respectively. Since the transport flux of sulfur out of China is estimated in this model to be higher than the Huang et al. (*8*) model, but much lower than Yang et al. (*17*) (about a third), this study seems to indicate an intermediate contribution of Chinese sources to Japan's overall deposition (i.e., between 3.5 and 10%, but closer to the former), still indicating a small contribution relative to Japanese sources.

4. Model Application

4.1 Designation of the Two Control Zones. Nilsson and Grennfelt (*18*) state that the resistibility of ecosystems to acid deposition, which varies from site to site, can be quantified by critical loads. Acid deposition exceeding the

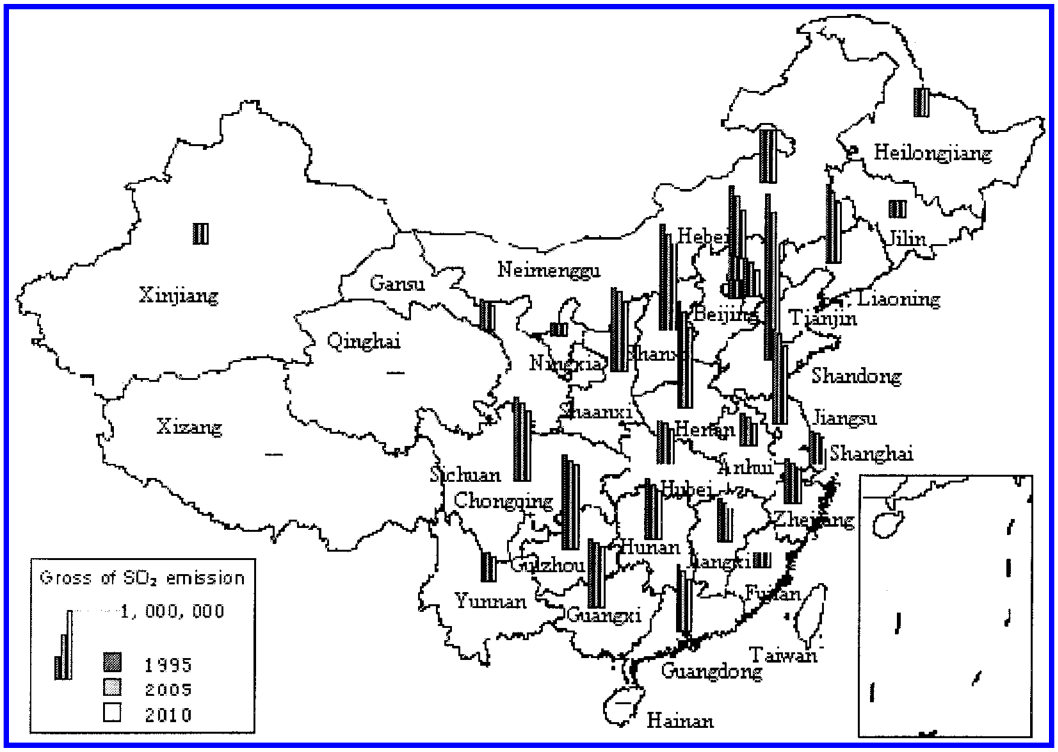

FIGURE 11. Control objective of gross SO_2 emission in each province in 2005 and 2010.

critical loads may cause long-term harmful effects on ecosystems. In China, sulfur deposition should be preferentially controlled, because the major acidifying precursor is sulfur dioxide. The most important principle for designating an acid rain control zone is that the critical load is exceeded by the sulfur deposition. In the study on designating acid rain control zones, critical loads of acid deposition in China were calculated (*19*). Comparing the sulfur deposition estimated with the critical loads, we became aware of where sulfur deposition should be controlled. The predicted critical load exceedance map of China is shown in Figure 8.

On the basis of the results of supporting research, the designation scheme for two control zones was finished in 1996 and was finally ratified by the State Council in early 1998. Considering the sensitivity of ecosystems to acidification and the regional difference of economic and technical conditions, those areas with precipitation pH monitored lower than 4.5 and sulfur deposition higher than their critical loads, except the national poverty counties, were designated as the acid rain control zones. The cities outside of the acid rain control Zones with an annual average concentration of ambient SO_2 higher than the second class of the national ambient air quality standard (60 $\mu g/m^3$) and the daily average concentration exceeding the third class (100 $\mu g/m^3$), except the national poverty counties, constitute the SO_2 pollution control zones. As shown in Figure 9, the range of the acid rain control zones and the SO_2 pollution control zones is about 1.09 million km^2 and occupies 11.4% of the whole area in China (*1*). The acid rain control zones involves 14 provinces south to the Yangtze River, with an area of 0.806 million km^2, while the SO_2 pollution control zones includes 63 cities north to the Yangtze River, with a total area of 0.29 million km^2. The acid rain control zones and the SO_2 pollution control zones share 8.4 and 3% of the Chinese land, respectively. In 1995, the SO_2 emission was about 23.7 million tons in China, while the emission was 14 million tons and nearly 60% of the total in the two control zones. Thus, the acid rain and SO_2 pollution in China should not be deteriorated if the SO_2 emission is well controlled in the acid rain control zones and the SO_2 pollution control zones.

4.2 Planing of Regional Acid Rain Control. After the two control zones were designated in 1998, a national program of comprehensive control of acid rain and SO_2 pollution in China was formulated to fulfill the objective of the two control zones, and should be carried out together with the plan of national economic and social development in the tenth five-year plan (for the period 2001 to 2005). On the basis of the emission−deposition relationship for each province (see Table 2), the effect of provinces outside of the acid rain control zones on the acid rain control zone provinces was first analyzed. As can be seen from Figure 10, the provinces outside of the acid rain control zones with high contribution to the sulfur deposition in the acid rain control zone provinces include Shangdong, Henan, Shaaxi, Shanxi, and Hebei (deposition higher than 20 kt/yr). However, SO_2 emitted from local sources, rather than from long-range transport, is predicted by our model to be the major cause of acid rain. Therefore, regional gross control of SO_2 emission should be carried out preferentially within the acid rain control zones.

One of the most important purposes of the regional acid rain control program plan is to prescribe the control objectives of gross SO_2 emission from each province in different phases. The years of 2000, 2005, and 2010 were considered by the programming group as the time limit of the short-, middle-, and long-range planning, respectively. As critical loads present thresholds to environmental damage from acid deposition, the ultimate policy target is to maintain deposition below critical loads in all areas. However, the goal is not attainable soon due to the limited technical and economic conditions and resources available in China. Therefore, the selection of intermediate targets based on critical loads was adopted because it can offer the potential for deriving a strategy with an improved environmental performance, e.g., a lower amount and a smaller area of critical load exceedance under the same level of emission abatement, than the uniform reduction approach. According to the short-range objective that the total SO_2 emission should be controlled within 24.6 billion tons before 2000, two methods of defining the target loads of sulfur deposition (i) critical load exceedance minimization and (ii) reducing

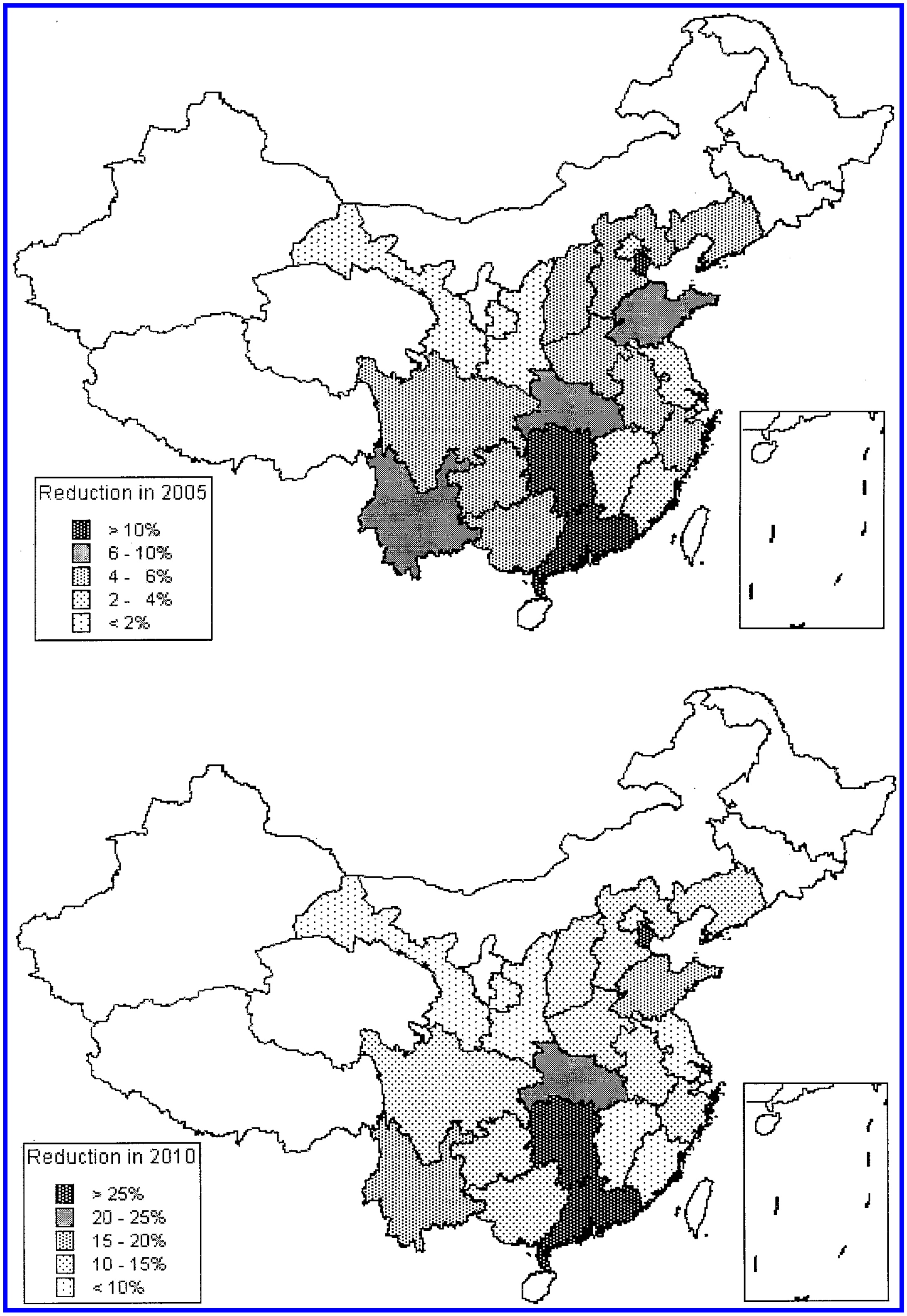

FIGURE 12. Percentage reduction of SO_2 emission required in each province in 2005 and 2010.

critical load exceedance by a fixed percentage were applied to calculate the required percentage reduction for each province (*20*). In the regional acid rain control program, strategies of reducing critical load exceedance by 25 and 40% were recommended for the middle- and long-range planning, respectively. Taking the year of 1995 as the basic year and the emission−deposition transfer matrix as an important input, an optimization procedure was solved for emission reduction minimization. The results are shown in Figure 11 and Figure 12. As can be seen from these figures, SO_2 emission should be reduced by a large amount in Shangdong, Guangdong, Henan, Hunan, Hebei, Guizhou, Sichuan, and Shanxi province to fulfill the middle- and long-range objective of acid deposition control in China, while the reduction ratio should be high for such provinces as Tianjin, Hunan, Guangdong, Hubei, Shandong, Chongqin, Yunan, and Anhui.

Acknowledgments

The authors are grateful to the National Natural Science Foundation of China for its financial support to carry out this study.

Supporting Information Available

The policy-oriented, two-dimensional, Eulerian statistical model for long-range transport and deposition of sulfur. This material is available free of charge via the Internet at http://pubs.acs.org.

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Received for review November 20, 2000. Revised manuscript received May 3, 2001. Accepted May 17, 2001.

ES001888U