Soil Acidification in China: Is Controlling SO_2 Emissions Enough?

YU ZHAO,[†,‡] LEI DUAN,[*,†] JIA XING,[†] THORJORN LARSSEN,[§] CHRIS P. NIELSEN,[‡] AND JIMING HAO[†]

Department of Environmental Science and Engineering, Tsinghua University, Beijing 100084, China, School of Engineering and Applied Sciences, Harvard University, 29 Oxford St., Cambridge, Massachusetts 02138, and Norwegian Institute for Water Research, Gaustadalléen 21, 0349 Oslo, Norway

Received May 13, 2009. Revised manuscript received August 17, 2009. Accepted September 3, 2009.

Facing challenges of increased energy consumption and related regional air pollution, China has been aggressively implementing flue gas desulfurization (FGD) and phasing out small inefficient units in the power sector in order to achieve the national goal of 10% reduction in sulfur dioxide (SO_2) emissions from 2005 to 2010. In this paper, the effect of these measures on soil acidification is explored. An integrated methodology is used, combining emission inventory data, emission forecasts, air quality modeling, and ecological sensitivities indicated by critical load. National emissions of SO_2, oxides of nitrogen (NO_X), particulate matter (PM), and ammonia (NH_3) in 2005 were estimated to be 30.7, 19.6, 31.3, and 16.6 Mt, respectively. Implementation of existing policy will lead to reductions in SO_2 and PM emissions, while those of NO_X and NH_3 will continue to rise, even under tentatively proposed control measures. In 2005, the critical load for soil acidification caused by sulfur (S) deposition was exceeded in 28% of the country's territory, mainly in eastern and south-central China. The area in exceedance will decrease to 26% and 20% in 2010 and 2020, respectively, given implementation of current plans for emission reductions. However, the exceedance of the critical load for nitrogen (N, combining effects of eutrophication and acidification) will double from 2005 to 2020 due to increased NO_X and NH_3 emissions. Combining the acidification effects of S and N, the benefits of SO_2 reductions during 2005−2010 will almost be negated by increased N emissions. Therefore abatement of N emissions (NO_X and NH_3) and deposition will be a major challenge to China, requiring policy development and technology investments. To mitigate acidification in the future, China needs a multipollutant control strategy that integrates measures to reduce S, N, and PM.

1. Current Air Pollution Situation and Policy

SO_2 and NO_X emissions, largely from fossil-fuel combustion, are the main cause of acid deposition. Along with rapid economic development, Chinese energy consumption (about 70% of which is supplied by coal combustion) has increased by 10% annually since 2000, compared to annual increases of only 4% for 1980−2000 (*1*). The national SO_2 emission level was reported as ∼26 million tons (Mt) in 2006 (*2*), much larger than total European anthropogenic emissions of ∼21 Mt (*3*) and U.S. emissions of ∼14 Mt (*4*) in the same year. Although no official statistics for NO_X are reported, emissions in China were estimated at ∼19 Mt in 2004 (*5*), close to the figures for Europe at ∼22 Mt and the United States at ∼20 Mt (*4*).

From 2000 to 2005, the area of China suffering acid rain was relatively stable or expanded only slightly in the south (*2, 6*). That is different from Europe and North America, where decreasing trends in acid deposition and related effects (e.g., recovery of acidified surface waters) are clearly observed (*7*). The power and transportation sectors are considered the most important sources of emissions and hence of regional air pollution and acidification in China. From 1980 to 2006, Chinese electricity consumption and number of vehicles increased by 10 and 15 times, respectively, much faster than the rise in total energy use.

Under strong environmental pressure, the Chinese government has targeted the power sector as the most important source of emissions (especially of SO_2) endangering regional atmospheric environment from 2006 to 2010, the period of the "Eleventh Five Year Plan" (11th FYP). The 11th FYP set as targets the reduction of national energy intensity (i.e., energy consumption per unit GDP output) and SO_2 emissions of 20% and 10%, respectively, measured in 2010 against 2005 levels. To achieve the targets, several regulations have been enforced in the power sector: all new thermal power units as well as most existing ones must have flue gas desulfurization (FGD) systems installed, and small units with low energy efficiency, totaling over 50 GW, should be gradually shut down. The measures appear to have had an effect: SO_2 emissions decreased slightly from 2006 to 2007, in contrast with substantial increases every year during the prior decade (*2*). Meanwhile, however, NO_X pollution is still serious, increasing nitrate concentrations and nitrate/sulfate ratios of precipitation in monitored cities (*8*). Furthermore, it is estimated that FGD penetration in the power sector will exceed 70% in 2010, limiting the potential for further SO_2 emission cuts using FGD after 2010. As energy demand is projected to keep rising beyond 2010, the long-term trajectory of acid deposition is still uncertain. Comprehensive analysis is thus necessary to improve understanding of future acidification trends.

In this study, an integrated methodology is used to illustrate the need for a multipollutant emission control approach in China, i.e., combining measures to reduce S, N, and PM. The approach combines inventorying of emissions, scenario development, atmospheric dispersion modeling, and assessment of ecological sensitivities measured by critical loads (CLs). The impacts on regional soil acidification of the 11th FYP policies as well as preliminary medium-to-long-term policies (through 2020) are quantified. The approach and results serve as direct support for practical policy making on regional air pollution and acidification in China.

2. Atmospheric Emissions and Deposition Modeling

Emission inventory is a fundamental element of modeling atmospheric deposition. To better serve the needs of scientific research and policy making, an integrated emission inventory was developed on the basis of the general methodology of previous studies (*9*−*11*). It relied chiefly on domestic

* Corresponding author phone: (+86-10)62771403; fax: (+86-10)62773650; e-mail: lduan@tsinghua.edu.cn.
† Tsinghua University.
‡ Harvard University.
§ Norwegian Institute for Water Research.

10.1021/es901430n CCC: $40.75
Published on Web 09/21/2009

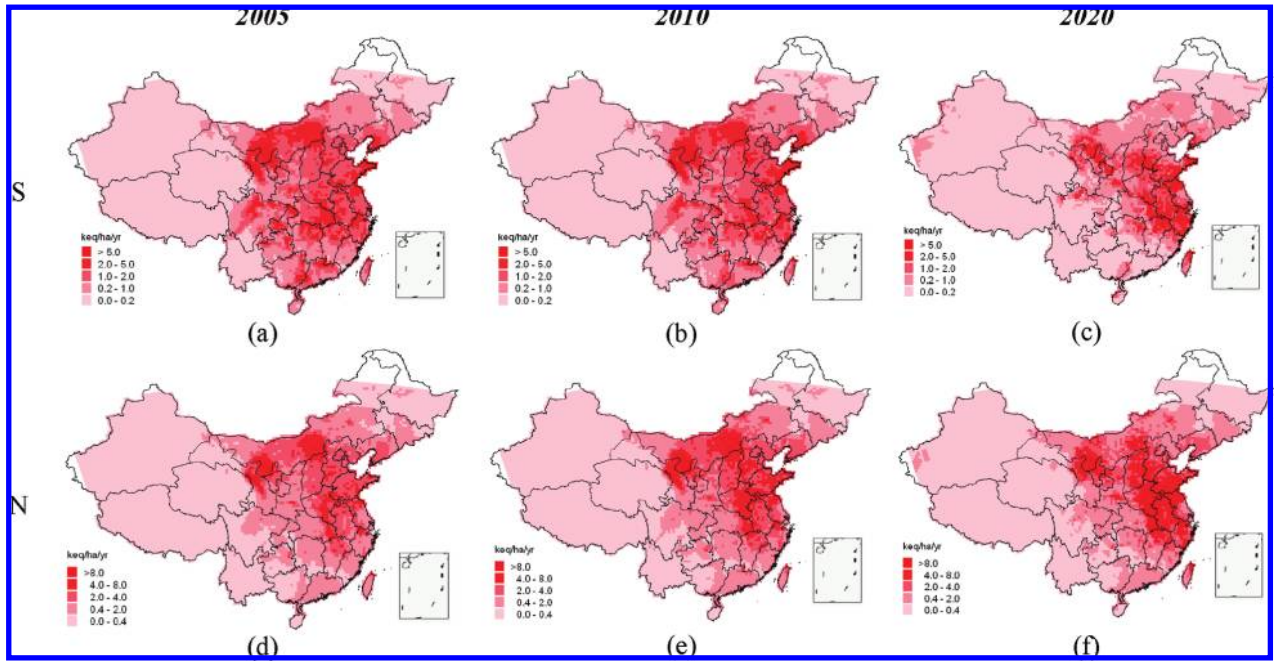

FIGURE 1. Simulated depositions of S (a−c) and N (d−f) for 2005, 2010, and 2020 in China. Because of limitation of the modeling domain, there are no results for small regions in the far northeast and northwest.

information for emission factors and activity levels (see Supporting Information for details). The updated emission estimation reflected the rapid increase of Chinese energy consumption since 2000 and, thus, provided a better basis for analyzing future policies than prior inventories.

Emission sources for SO_2, NO_X, and PM were classified into two categories, combustion (power, industry, transport, and domestic) and noncombustion (mainly industrial processes). The emissions from coal-fired power plants were evaluated with a unit-based methodology (*12*). For other sectors, emissions at the provincial level were calculated as the product of activity levels and corresponding emission factors by region and sector. The activity levels were chiefly obtained from official economic and energy statistics, while emission factors were estimated from diverse sources, including published domestic studies and unpublished field measurements by the authors and collaborators (*13*−*17*). Another important source of acidification, NH_3, is mainly from livestock farming, and fertilizer use. Because of sparse domestic research, NH_3 emission factors were mainly taken from European results (*9, 18*).

The years 2010 and 2020 were chosen for emission projections. The FGD deployments and small boiler retirements in the power sector will continue and also spread into industrial sectors after 2010 (*12*). NO_X control policies have not been promulgated and thus were projected, specifically: inclusion of denitrogenation technologies (e.g., selective catalytic reduction, SCR) in new power plants in eastern, north-central, and south-central China; installation of low-NO_X burners (LNB) in industrial boilers and cement plants in those areas; and a requirement of Euro V emission standards for new vehicles starting in 2012. NH_3 emission factors were assumed to be unchanged from 2005 onward, and the annual rates of increase in livestock numbers and fertilizer use were assumed to continue at the reported rates for 2000−2005 (4.0% and 2.5%, respectively). Other assumptions were taken from a recent study of Chinese energy and emission trends (*19*).

On the basis of the emission inventory, sulfur and nitrogen depositions were simulated with the Models-3/Community Multiscale Air Quality (CMAQ) system (V4.4) (*20*). This model was previously modified for China and has proven suitable for regional and urban-scale air quality simulations (*21, 22*). The modeling domain covered most of East Asia. Provincial emissions were distributed on a 36 km × 36 km grid system using geographic information such as locations of power plants, population density, and road networks (see Supporting Information for the gridded emissions of SO_2 and NO_X). Emissions in the domain but outside of China were taken from the NASA TRACE-P inventory (*10*). Monthly variations of emissions in different sectors were from previous studies (*5, 10*). The driving meteorological inputs were provided by the fifth-generation NCAR/Penn State Mesoscale Model (MM5). Because the work focused on annual estimates, one month representing each season (January, April, July, and October) were chosen as simulation periods and then averaged (see Supporting Information for discussion of model performance).

National anthropogenic emissions of SO_2, NO_X, PM, and NH_3 in 2005 were estimated at 30.7, 19.6, 31.3, and 16.6 Mt, respectively. If current policy is fully implemented, national SO_2 emissions will decrease by 8.3% between 2005 and 2010, close to the national target of 10%. By 2020, the emissions will be further reduced by around 12% compared to 2010, largely through measures implemented in nonpower sectors. A similar trend was also found for PM. In contrast, NO_X and NH_3 emissions were estimated to increase by 30% and 57%, respectively, from 2005 to 2020.

As shown in Figure 1, north-central and eastern China were estimated to receive the most S and N depositions by the CMAQ simulations, attributed mainly to high densities of energy consumption and emissions. From 2005 to 2020, although S deposition will increase in part of eastern China, it will significantly decrease in most other areas, with abatements greater than 30% in some provinces of south-central and southwestern China. In contrast to S, however, N deposition will rise across most of China. Depositions in some provinces of south-central and eastern China will increase by more than 40%.

3. Mapping of Critical Load for Soil Acidification

As an indicator of ecosystem sensitivity, the critical load for acidification, defined as the deposition of acidifying com-

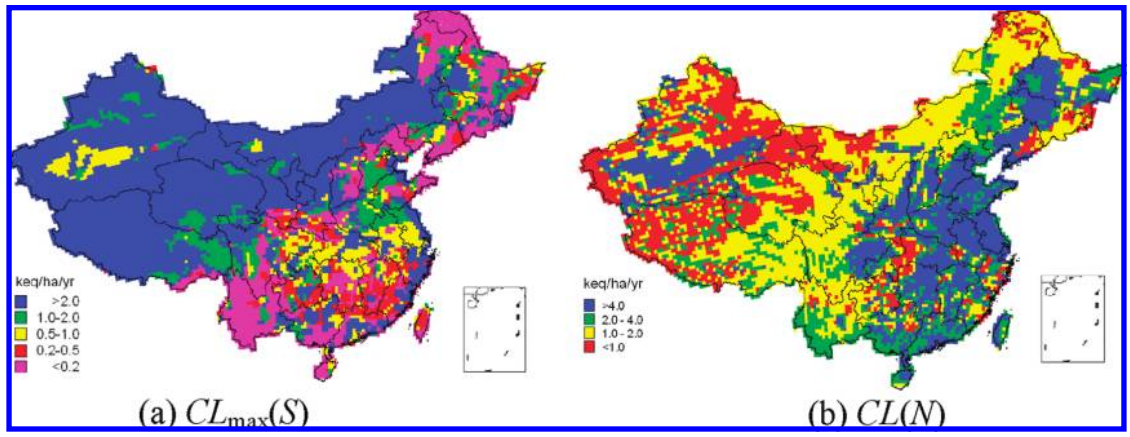

FIGURE 2. Critical loads for S and N in China, calculated using eqs 1 and 2, respectively. Spatial resolution is 36 km × 36 km, the same as the atmospheric dispersion outputs.

pounds below which harmful ecological effects do not occur, has been used as the basis of emission control and acid rain mitigation in Europe as well as China (*23*−*25*). Among different methods of calculating critical loads is the Steady State Mass Balance (SSMB) model (*26*). It assumes that the processes of formation and depletion of acidity in soil are time-independent, thus simplifying the dynamic simulation of soil chemistry and facilitating the mapping of critical loads when ecosystem data are limited (*27*). An extended SSMB method has been developed for areas with high and highly unstable base cation (BC) emissions such as China (*28*). Employing that approach, we updated S and N critical loads and used them as the criteria of acidification in this work. As response criteria, linking the modeled chemical parameters to biological impacts, a combination of alkalinity (for acidification) and nitrogen content (for eutrophication) were used.

The critical load of S and N can be independently calculated by eqs 1 and 2 (see Supporting Information for details and definitions of terms). Relevant parameters were mainly taken from previous studies (*27*−*33*).

$$CL_{max}(S) = BC_D + BC_W - BC_U - ANC_{L,crit} \quad (1)$$

$$CL(N) = N_I + N_U + \frac{\min(CL_{max}(S), N_{L,crit})}{1 - f_{DE}} \quad (2)$$

When combining the effects of S and N, the critical load of S can be recalculated with eq 3, which means that the threshold of S deposition might be lower than $CL_{max}(S)$ under a given N deposition level. This is the basis for integrating analyses of S and N in this assessment (see Supporting Information for details).

$$CL(S) = \begin{cases} CL_{max}(S) & (N_D \le N_I + N_U) \\ CL_{max}(S) - (1 - f_{DE}) \times [N_D - (N_I + N_U)] & (N_D > N_I + N_U) \end{cases} \quad (3)$$

Maps of $CL_{max}(S)$ and CL(N) over the country were developed and are shown in Figure 2. Because of different criteria of effects used for S and N, the distributions of sensitive regions were different for the two compounds. Because of high weathering rates and natural deposition of base cations, the S critical loads in north-central and northwestern China were generally higher than 2.0 keq/ha/yr, while the values could be lower than 0.2 keq/ha/yr in the northeast where temperature and weathering rates were low, and in the south, where scarcity of weatherable minerals and high vegetation uptake of base cations occurred. In contrast, the N critical load (often driven by the N leaching criterion) was relatively low in the northwest, with poor vegetation uptake of N, and high in the south and east, with considerable soil denitrification.

4. Assessment of National Soil Acidification

Soil acidification as well as soil eutrophication by N was evaluated by directly comparing the simulated S and N deposition and critical load maps. The distributions of critical load exceedance were estimated for 2005, 2010, and 2020 (Figure 3) and are summarized at the national level in Table 1.

In 2005, the areas exceeding $CL_{max}(S)$ covered approximately 28% of the mainland territory, mainly in eastern, south-central, and part of southwestern China (Figure 3a). The total exceedance for the country was estimated at 4.1 Mt S, equaling 27% of national SO_2 emissions. As noted above, considerable measures are being implemented during 2005−2010 in the power sector, the largest emitter of SO_2. As a result, it is estimated that the percentage of areas exceeding $CL_{max}(S)$ will decrease to 26% in 2010, and that the total exceedance will decrease by 9% (Figure 3b). Following the SO_2 control strategy, further improvement will occur during 2010−2020. In 2020, even though a small region of eastern China may have a somewhat larger exceedance, the total area exceeding $CL_{max}(S)$ will decline to 20% of the mainland territory (Figure 3c). Generally, looking at S separately, the implementation of SO_2 emission control measures will gradually alleviate acidification.

In contrast to S, the CL(N) exceedance is expected to keep rising from 2005 to 2020, along with increased NO_X and NH_3 emissions (Figure 3d−f). In 2005, the percentage of China's territory exceeding CL(N) was 12%, and the total quantity was estimated at 2.8 Mt N. The values will increase to 15% and 4.2 Mt N in 2010. The larger increase in total tons compared to the total area indicates worsening conditions in regions already in exceedance. From 2010 to 2020, CL(N) exceedances will largely increase in the east, indicating that emission and deposition controls are urgently needed in this region. Because $N_{L,crit}$ was generally lower than $CL_{max}(S)$ and thus binding under eq 2 for most ecosystems, the CL(N) exceedances are more often driven by ecosystem eutrophication than acidification.

If the acidification effect of N is combined with that of S, however, the importance of N on future acidification risks is clearly revealed. Maps g−i of Figure 3 show the critical load exceedance based on CL(S) calculated with eq 3. Taking N deposition into account, the exceedance of CL(S) in 2005 (Figure 3g) was 4.9 Mt S, 18% higher than that of $CL_{max}(S)$ (Figure 3a). This discrepancy will reach 33% in 2010 (Figure 3h versus Figure 3b) and 31% in 2020 (Figure 3i versus Figure

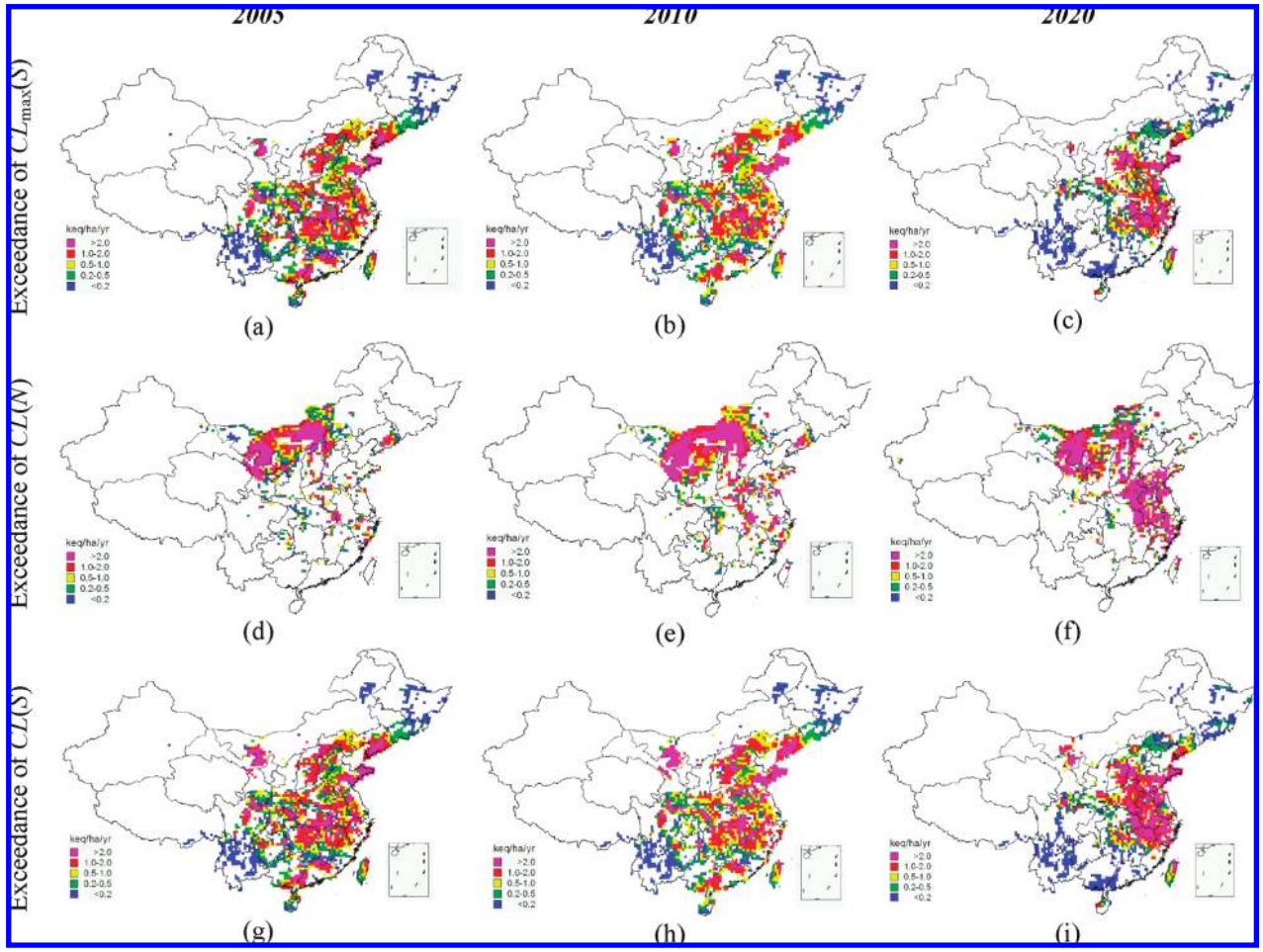

FIGURE 3. Exceedances of critical loads for acidification and nutrient nitrogen in 2005, 2010, and 2020. Depositions simulated by CMAQ were compared with selected criteria. CL_{max}(S), calculated by eq 1, and CL(N), calculated by eq 2, were used as criteria for maps a−c and d−f, respectively. CL(S), which was calculated by eq 3 combining the effect of simulated N deposition, was used as the criterion for maps g−i.

TABLE 1. Summary of Critical Load Exceedance in 2005, 2010, and 2020

	million tons of S/N			area percentage of the country (%)		
	2005	2010	2020	2005	2010	2020
exceedance of CL_{max}(S)	4.14	3.77	3.29	27.7	26.2	20.4
exceedance of CL(N)	2.76	4.19	5.84	12.1	15.2	16.1
exceedance of CL(S)	4.89	5.05	4.29	28.2	28.1	22.3

3c) as N deposition rises. It will particularly exacerbate conditions in the east, where N deposition and CL(N) exceedances increase substantially.

Cumulative distributions of S exceedances for 2005 and 2010 are illustrated in Figure 4. Disregarding N, S acidification at the national level during those five years clearly will be reduced (from the red curve to the green one). However, that benefit will be counteracted largely, or almost totally by increased N deposition (from the blue curve to the black curve). In other words, the potential risk of soil acidification will probably grow more serious even with an 8% decline in SO_2 emissions during 2005−2010. That trend can also be confirmed by the comparison of critical load exceedances given in Table 1. If there are no improvements in N control during those five years (which is likely to be the case), even more abatement of S deposition, mainly in eastern and northern China, would be required to prevent worsening acidification as shown in Figure 5. The total quantity of

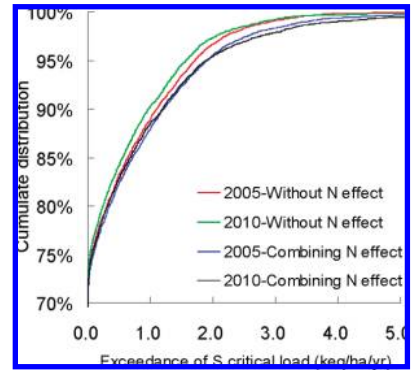

FIGURE 4. Cumulative distributions of exceedances of S critical loads in 2005 and 2010 with and without the additional effects of nitrogen. Higher curves indicate relatively lower acidification risks at the national level.

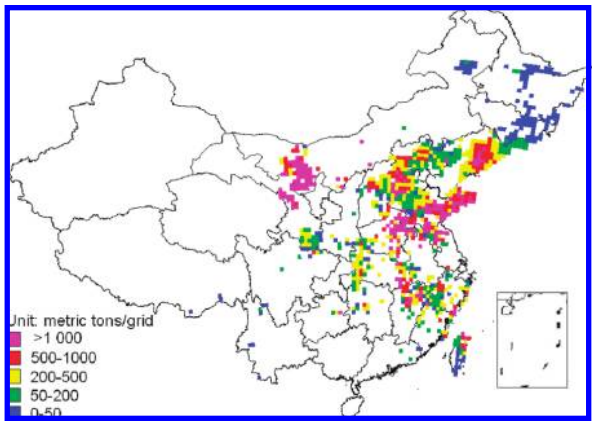

FIGURE 5. Extra S abatement required in 2010 to prevent exacerbation of acidification compared to the 2005 level, taking increased N deposition into consideration.

additional S abatement required to counteract effects of increased N emissions is estimated at 0.7 Mt, equaling 5% of the SO_2 emissions in 2010.

5. Discussion and Policy Implications

On the basis of the results, it can be concluded that the growth of NO_X and NH_3 emissions in China will probably negate the gains from SO_2 control on mitigation of soil acidification, and that expanded N pollution control is required in the future. Currently, the electric power and industry sectors are the main sources of NO_X emissions in China. Although LNBs are now generally built in new power plants, their efficiency of removing NO_X averages around 30%, which is poor compared to more advanced technologies (*12*). In the industrial sector, there is currently no NO_X emission standard for coal combustion boilers, and emission control measures are rarely taken. Given rising NO_X emissions, power plants increasingly install NO_X reduction technology such as SCR systems. In contrast to FGD, however, NO_X emission control in China is in its infancy and still far from generally applied. At present, the penetration rate of SCR in the thermal power sector is less than 10% (personal communication from a Ministry of Environmental Protection officer). According to domestic measurements (by the authors, as yet unpublished), the removal efficiency of SCR systems in China ranged from 40% to 70%, generally lower than that of FGD (which can reach 90%). Furthermore, another major NO_X source, the transportation sector, is expected to have a much greater impact in the future because of an inevitable increase in vehicle numbers. For these reasons, NO_X control could be more difficult than SO_2 control. In this study, NO_X emissions were projected to continue rising to 2020 even if tentatively proposed control measures are implemented, indicating the need for a more stringent policy than previously recognized.

Regarding NH_3, another species contributing to N pollution, to date there are no emission standards or control regulations in China. In this study, the NH_3 emissions are expected to exceed SO_2 and NO_X by 2020 (in metric tons), raising additional concern. As most NH_3 is emitted from scattered agricultural activities, it must be controlled not by measures focused on large point sources but by long-term changes in farming practices, a more difficult and time-consuming requirement. With little available information on relevant processes in China, however, emission factors and activity levels regarding NH_3 are uncertain, and further research, notably field measurement, is thus needed.

Emission and deposition of alkaline dust play an important role counteracting acidification, more so in China than in Europe or North America (*6, 28*). To reduce the uncertainty from alkaline dust variation, simulated critical depositions of base cations (from natural sources and thus considered constant) were used to estimate critical loads in this work (*32*). That approach identified a long-term potential of soil acidification. To better evaluate the temporal trend, the variation of anthropogenic base cations, which are mainly from the PM emissions of industrial processes like cement and limestone production, should be included in future research. Because of large health risks, such anthropogenic PM emissions (and thereby base cations) will be reduced in the future by specific policies and side effects of SO_2 controls (e.g., wet FGD). In other words, not only increased emissions of NO_X and NH_3 but also decreased PM emissions could raise pressure to limit acidification through further SO_2 abatement. This will become more difficult after the current implementation of controls in power plants, the largest sector source of SO_2. Therefore, decision makers will benefit by taking a multipollutant perspective, in terms of emission scenarios for the future and design of possible control policies that combine scientific, economic, and practical feasibility of S, N (including NO_X, and NH_3), and PM abatement simultaneously.

Acknowledgments

The authors are grateful for the financial support of the National Basic Research Program of China (No. 2005 CB422206) and the U.S. National Science Foundation (Grant ATM-0635548). We would also like to thank three anonymous referees for useful comments on this work.

Supporting Information Available

Methodology of emission inventory estimate, methods and results of spatial allocation of emissions, comparisons of CMAQ simulations and observations of depositions, and methods of critic load calculation. This material is available free of charge via the Internet at http://pubs.acs.org.

Literature Cited

(1) *National Bureau of Statistics. China Energy Statistical Yearbook*; China Statistics Press: Beijing, 2006, in Chinese.
(2) Chinese Environment Communique 2001−2007. Ministry of Environmental Protection, The People's Republic of China. http://www.sepa.gov.cn.
(3) Emission Data. Centre on Emission Inventories and Projections. http://www.ceip.at.
(4) National Emissions Inventory (NEI) Air Pollutant Emissions Trends Data. U.S. Environmental Protection Agency. http://www.epa.gov/ttn/chief/trends/index.html.
(5) Zhang, Q.; Streets, D. G.; He, K. B.; Wang, Y. X.; Richter, A.; Burrows, J. P.; Uno, I.; Jang, C. J.; Chen, D.; Yao, Z.L. NO_X emission trends for China, 1995−2004: The view from the ground and the view from space. *J. Geo. Res.* **2007**, *112*, (D22), Art. No. D22306.
(6) Larssen, T.; Lydersen, E.; Tang, D. G.; He, Y.; Gao, J. X.; Liu, H. Y.; Duan, L.; Seip, H. M.; Vogt, R. D.; Mulder, J.; et al. Acid rain in China. *Environ. Sci. Technol.* **2006**, *40* (2), 418–425.
(7) Stoddard, J. L.; Jeffries, D. S.; Lukewille, A.; Clair, T. A.; Dillon, P. J.; Driscoll, C. T.; Forsius, M.; Johannessen, M.; Kahl, J. S.; Kellogg, J. H.; et al. Regional trends in aquatic recovery from acidification in North America and Europe. *Nature.* **1999**, *401* (6753), 575–578.
(8) Zou, S. M.; Wang, J. N.; Hong, Y. X. *Research Report on National Environmental Protection Planning of 11th FYP*; Chinese Environmental Science Press: Beijing, 2006, in Chinese.
(9) Klimont, Z.; Cofala, J.; Schopp, W.; Amann, M.; Streets, D. G.; Ichikawa, Y.; Fujita, S. Projections of SO_2, NO_X, NH_3, and VOC emissions in East Asia to 2030. *Water, Air, Soil Pollut.* **2001**, *130* (1−4), 193–198.
(10) Streets, D. G.; Bond, T. C.; Carmichael, G. R.; Fernandes, S. D.; Fu, Q.; He, D.; Klimont, K.; Nelson, S. M.; Tsai, N. Y.; Wang, M.Q. An inventory of gaseous and primary aerosol emissions in Asia in the year 2000. *J. Geo.Res.* **2003**, 108 (D21), Art. No. 8809.
(11) Ohara, T.; Akimoto, H.; Kurokawa, K.; Horii, N.; Yamaji, K.; Yan, X.; Hayasaka, T. An Asian emission inventory of anthropogenic emission sources for the period 1980−2020. *Atmos. Chem. Phys.* **2007**, *7* (16), 4419–4444.

(12) Zhao, Y.; Wang, S. X.; Duan, L.; Lei, Y.; Cao, P. F.; Hao, J. M. Primary air pollutant emissions of coal-fired power plants in China: current status and future prediction. *Atmos. Environ.* **2008**, *42* (36), 8442–8452.
(13) *State Environmental Protection Administration. Handbook of Industrial Pollution Emission Rates*; China Environmental Science Press: Beijing, 1996, in Chinese.
(14) Hao, J. M.; Tian, H. Z.; Lu, Y. Q. Emission inventories of NO_X from commercial energy consumption in China, 1995−1998. *Environ. Sci. Technol.* **2002**, *36* (4), 552–560.
(15) Zhang, Q. Study on regional fine PM emissions and modeling in China. Ph.D. Thesis, Tsinghua University: Beijing, 2005, in Chinese.
(16) Compilation of Air Pollutant Emission Factors. U.S. Environmental Protection Agency. http://www.epa.gov/ttn/chief/.
(17) EMEP/CORINAIR Atmospheric Emission Inventory Guidebook 2005. European Environment Agency. http://reports.eea.europa.eu/EMEPCORINAIR4/.
(18) Bouwman, A. F.; Lee, D. S.; Asman, W. A. H.; Dentener, F. J.; Vanderhoek, K. W.; Olivier, J. G. J. A global high-resolution emission inventory for ammonia. *Global Biogeochem. Cycles.* **1997**, *11* (4), 561–587.
(19) Zhang, C. Y.; Wang, S. X.; Xing, J.; Zhao, Y.; Hao, J. M. Current status and future projections of NO_X emissions from energy related industries in China. *Acta Sci. Circumstantiae.* **2008**, *12*, 2470–2479, in Chinese.
(20) Byun, D. W.; Ching, J. K. S. Science algorithms of the EPA Models-3 Community Multiscale Air Quality Model (CMAQ) modeling system; Report EPA/600/R-99/030; U.S. Environmental Protection Agency: Research Triangle Park, NC, 1999.
(21) Streets, D. G.; Fu, J. H. S.; Jang, C. J.; Hao, J. M.; He, K. B.; Tang, X. Y.; Zhang, Y. H.; Wang, Z. F.; Li, Z. P.; Zhang, Q.; et al. Air quality during the 2008 Beijing Olympic Games. *Atmos. Environ.* **2007**, *41* (3), 480–492.
(22) Wang, L. T.; Hao, J. M.; He, K. B.; Wang, S. X.; Li, J. H.; Zhang, Q.; Streets, D. G.; Fu, J. S.; Jang, C. J.; Takekawa, H.; et al. A modeling study of coarse particulate matter pollution in Beijing: regional source contributions and control implications for the 2008 Summer Olympics. *J. Air Waste Manage. Assoc.* **2008**, *58* (8), 1057–1069.
(23) *Manual on Methodologies and Criteria for Modelling and Mapping Critical Loads and Levels and Air Pollution Effects, Risks, and Trends*; Umweltbundesamt: Berlin, 2004.
(24) *The 1999 Protocol to Abate Acidification, Eutrophication, and Ground-Level Ozone*; United Nations Economic Commission for Europe: New York and Geneva, 1999.
(25) Hao, J. M.; Wang, S. X.; Liu, B. J.; He, K. B. Designation of acid rain and SO_2 pollution control zones and control policies in China. *J. Environ. Sci. Health, Part A.* **2000**, *35* (10), 1901–1914.
(26) Sverdrup, H.; de Vries, W. Calculating critical loads for acidity with the Simple Mass-Balance method. *Water, Air, Soil Pollut.* **1994**, *72* (1−4), 143–162.
(27) Duan, L.; Xie, S. D.; Zhou, Z. P.; Ye, X. M.; Hao, J. M. Calculation and mapping of critical loads for S, N, and acidity in China. *Water, Air, Soil Pollut.* **2001**, *130* (1−4), 1199–1204.
(28) Zhao, Y.; Duan, L.; Larssen, T.; Hu, L. H.; Hao, J. M. Simultaneous assessment of deposition effects of base cations, sulfur, and nitrogen using an extended critical load function for acidification. *Environ. Sci. Technol.* **2007**, *41* (6), 1815–1820.
(29) Duan, L.; Huang, Y. M.; Hao, J. M.; Xie, S. D.; Hou, M. Vegetation uptake of nitrogen and base cations in China and its role in soil acidification. *Sci. Total Environ.* **2004**, *330* (1−3), 187–198.
(30) Duan, L.; Hao, J. M.; Xie, S. D.; Zhou, Z. P.; Ye, X. M. Determining weathering rates of soils in China. *Geoderma.* **2002**, *110* (3−4), 205–225.
(31) *Integrated Monitoring Program on Acidification of Chinese Terrestrial Systems-IMPACTS. Annual Report 2003*; Larssen, T., Tang, D. G., He, Y., Eds.; Norwegian Institute for Water Research: Oslo, Norway, 2004.
(32) Duan, L.; Lin, Y.; Zhu, X. Y.; Tang, G. G.; Gao, D. F.; Hao, J. M. Modeling atmosheric transport and depostion of calcium in China. *J. Tsinghua Univ. (Sci. Technol.)* **2007**, *47* (9), 1462–1465, in Chinese.
(33) Hao, J. M.; Qi, C. L.; Duan, L.; Zhou, Z. P. Evaluating critical loads of nutrient nitrogen on soils in China using the SMB method. *J. Tsinghua Univ. (Sci. Technol.)* **2003**, *43* (6), 849–853, in Chinese.

ES901430N