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## ACID RAIN *in* China

Transformation of Acetaminophen by  
Chlorination Produces Toxicants

Accumulation of Contaminants in Fish  
from Wastewater Treatment Wetlands

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# ACID RAIN *in* China



THORJØRN LARSEN

**Rapid industrialization  
has put citizens and  
ecosystems at risk.**

- |                                       |                                      |
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Acid rain emerged as an important environmental problem in China in the late 1970s. Many years of record economic growth have been accompanied by increased energy demand, greater coal combustion, and larger emissions of pollutants. As a result of significant emissions and subsequent deposition of sulfur, widespread acid rain is observed in southern and southwestern China. In fact, the deposition of sulfur is in some places higher than what was reported from the “black triangle” in central Europe in the early 1980s. In addition, nitrogen is emitted from agriculture, power production, and a rapidly increasing number of cars. As a result, considerable deposition of pollutants occurs in forested areas previously thought to be pristine.

Little is known about the effects of acid deposition on terrestrial and aquatic ecosystems in China. In this article, we present the current situation and what to expect in the future, largely on the basis of results from a five-year Chinese–Norwegian cooperative project. In the years ahead, new environmental challenges must be expected if proper countermeasures are not put into place.

### **Acid rain, acidification, and their environmental consequences**

Acid deposition is formed from  $\text{SO}_2$  and  $\text{NO}_x$  emitted to the atmosphere, largely because of fossil-fuel combustion. The most important sources are energy production, especially coal- and oil-fired power plants, and transportation sources, such as vehicles and ships. The air pollutants are transformed in the atmosphere to  $\text{H}_2\text{SO}_4$  and  $\text{HNO}_3$ , transported across distances potentially as far as hundreds of kilometers, and deposited as precipitation (wet deposition) and as gas and particles (dry deposition).

Alkaline dust and  $\text{NH}_3$  are other important components. These compounds act to neutralize the acids. The main source of  $\text{NH}_3$  to the atmosphere is agriculture. Although  $\text{NH}_3$  neutralizes acidity in precipitation, the resulting  $\text{NH}_4^+$  contributes to acidification of soil and surface water through chemical processes in the soil. Alkaline dust in the atmosphere can, for instance, be particles of limestone ( $\text{CaCO}_3$ ) or  $\text{CaO}$ . The sources of alkaline dust are many; some are natural (e.g., windblown dust from deserts) and some anthropogenic (e.g., industrial and construction activities). Such alkaline dust can neutralize much of the acidity from the  $\text{SO}_2$  by forming neutral  $\text{CaSO}_4$ , instead of  $\text{H}_2\text{SO}_4$ , in the atmosphere.

Acid rain has been a well-known environmental problem for decades and can lead to acidification of surface waters and soils. Surface-water acidification has caused widespread loss of fish populations, especially in Scandinavia but also in the U.S., Canada, and the U.K. Severe forest dieback caused by direct  $\text{SO}_2$  damage has been noticed repeatedly in the vicinity of emission sources over the centuries. In the 1980s, forest decline was observed to be widespread and far from emission sources in central Europe. Although other stress factors were present,

the forest losses created concern over the effects of soil acidification, which was hypothesized to damage trees through mechanisms involving aluminum toxicity and nutrient deficiency.

### **Acid rain in China**

In China, concern about the possible effects of acid rain emerged later than in Europe and North America. The first reports in the international literature regarding acid rain in China appeared in the 1980s (1, 2).

A major cause of acid rain in China is the extensive use of coal, which in 2004 accounted for 69% of the energy production (3). Oil generated 23% (Figure 1a). Consumption of coal and oil and subsequent emissions of  $\text{SO}_2$  have increased rapidly since the 1970s (Figure 1b). After a couple of years of apparently decreasing  $\text{SO}_2$  emissions, the rate of increase is currently faster than ever. The recent annual growth in the coal consumption has been ~20% (4). The national average content of sulfur in the coal is 1.1%, but in some heavily industrialized areas in the southwest, the sulfur content can be as high as 4%. Oil is also a substantial source of sulfur to the atmosphere. The total emissions of  $\text{SO}_2$  to the atmosphere in China were ~22 million metric tons (t) in 2003 (4). This is more than the total anthropogenic European emissions of 17 million t in 2002 (exclud-

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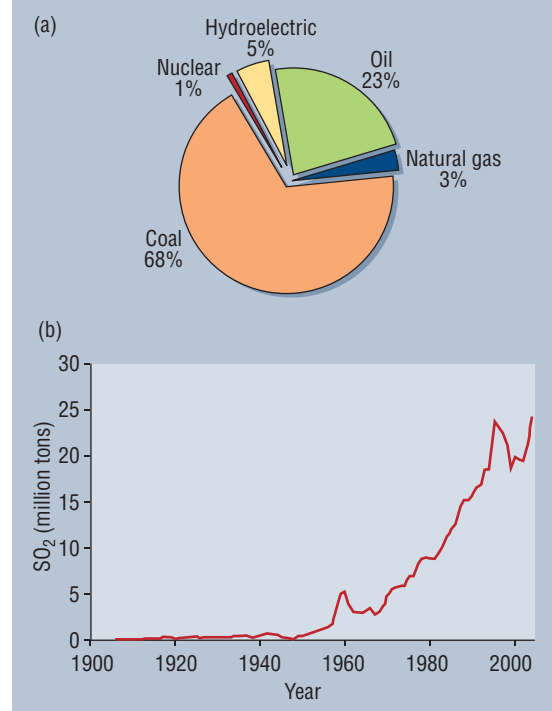
ing ships; 5) and 2× the U.S. emissions of 10 million t in 2002 (6).

The extensive coal combustion also results in considerable emissions of  $\text{NO}_x$  to the atmosphere. In addition, the number of motor vehicles has increased dramatically in recent years, from 6.2 million in 1990 to 36.0 million in 2003 (7); a continued rapid increase is expected.  $\text{NO}_x$  emissions in 2003 were estimated at 12 million t (counted as  $\text{NO}_2$ ; 8), which is just more than half of the total U.S. emissions (9) and ~2× the total European emissions (5).

**FIGURE 1**

**(a) Primary Chinese energy sources in 2003 and (b) the historical growth of  $\text{SO}_2$  emissions**

Data in (b) for 1990–2003 from Ref. 4; estimates for before 1990 from Ref. 11.



However, China's ammonia emissions are still >3× larger than its  $\text{NO}_x$  emissions, because agricultural activities are widespread and intensive (10).

Many Chinese cities have high concentrations of particulates and  $\text{SO}_2$  in the air. However, local air quality has improved substantially in recent years because of measures such as restrictions on the use of coal-fired household stoves, the termination or relocation of heavily polluting industries in urban areas to the countryside, and the installation of scrubbers on coal-fired boilers. These measures have reduced air pollution and lessened health effects considerably, although there still is a long way to go (Figure 1b).

The effects of air pollution on the natural environment may have a different geographical distribution than the impacts on human health. Although considerable emissions of acidifying compounds

occur in most parts of the country (except in the huge mountain and desert areas in the western and northwestern regions), acid deposition is mainly a problem in southern and southwestern China. In northern China, alkaline dust from the desert areas largely neutralizes the acids in the deposition. In southern China, however, the influence of the desert dust is much less (14). On the other hand, alkaline dust from coal combustion, cement production, and construction activities is important in this region.

**Monitoring is needed!**

Experience in Europe and North America has clearly shown that monitoring air pollution and its effects is necessary to document distribution and trends, to check whether measures are working as anticipated, and to provide the basis for testing and calibrating models. As the acid rain problem in China has developed, it has become increasingly clear that appropriate environmental monitoring is necessary.

In China, national monitoring programs generally focus on urban air quality and precipitation pH. Little other information exists in China or in other countries with similar subtropical and monsoonal climate conditions and ecosystems. In order to supplement the existing monitoring and gather new information on Chinese systems, a set of integrated monitoring sites was established through a Chinese–Norwegian cooperative project, the Integrated Monitoring Program on Acidification of Chinese Terrestrial Systems (IMPACTS; 15). Air pollution; precipitation composition; and soil, water, and vegetative effects are being intensively studied at five forested sites (Figure 2).

**High emissions produce high deposition**

The five monitoring sites represent acid-sensitive forested ecosystems in southern and southwestern China that are considered to be exposed to acid deposition. The annual sulfur deposition at the sites ranges from ~2 to 16 g-S/m<sup>2</sup> (Figure 3a), which is in the same range as, or higher than, that seen within most of central Europe in ~1980, when acid depo-

**The number of motor vehicles [in China] has increased dramatically in recent years, from 6.2 million in 1990 to 36.0 million in 2003.**

sition was at its peak. The highest deposition was observed at the Tie Shan Ping (TSP) monitoring site (Figure 2) in a forest reserve outside Chongqing, one of the most heavily polluted areas of China. The lowest deposition was at the Lei Gong Shan (LGS) mountain reserve site in the Guizhou province, which is quite remote and is not near any large local emission sources.



Dry deposition is generally high in China. The data from the integrated monitoring sites reported here are for estimated total deposition, in which dry and wet deposition are integrated via the collection of throughfall under the tree canopies. Dry deposition at these sites is typically  $\geq 50\%$  of the total deposition flux. Wet or bulk deposition values thus greatly underestimate total deposition. Most monitoring currently carried out in China measures only bulk deposition. The use of throughfall as total deposition may be problematic because of leaching from the tree canopies. However, at high deposition fluxes, the contribution from the canopies is less important.

The total nitrogen deposition at the 5 sites ranges from 0.6 to 4.4 g-N/m<sup>2</sup> in 2003 (Figure 3b), which is in the same range as that observed in Europe and North America, although somewhat lower than the highest level measured in Europe (in The Netherlands) during the 1980s. Deposition of NH<sub>4</sub><sup>+</sup> is typically 2× the amount of NO<sub>3</sub><sup>-</sup> deposition; this reflects the importance of NH<sub>4</sub><sup>+</sup> emissions from agricultural sources for the total nitrogen load.

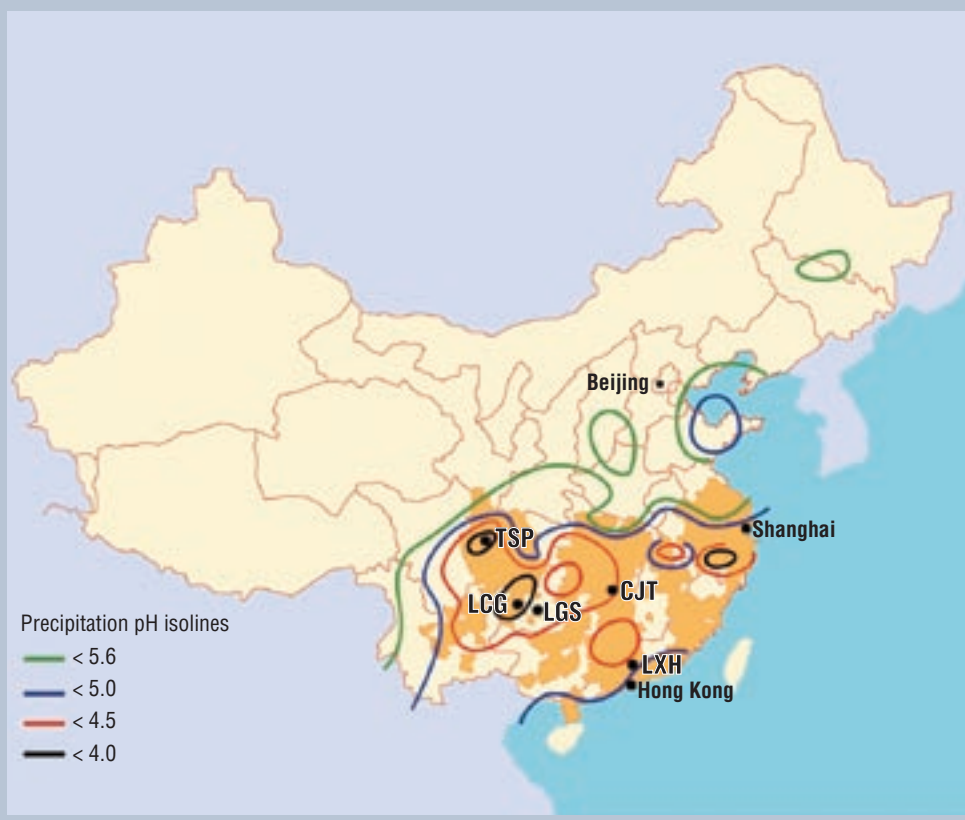
The considerable deposition of NH<sub>4</sub><sup>+</sup>, with its potential to contribute to acidification, implies that pH alone is not a good indicator of acid rain. This needs to be taken into account when the target area for acid-rain control is chosen. Because NH<sub>4</sub><sup>+</sup> and NO<sub>3</sub><sup>-</sup> are also important in terms of eutrophication of terrestrial and aquatic ecosystems, monitoring of these compounds and their environmental effects will also serve purposes other than acidification studies.

The importance of the alkaline dust is reflected in the high calcium deposition at the IMPACTS sites, ranging from 2 to 12 g-Ca/m<sup>2</sup> in 2003 (Figure 3c). This is much higher than calcium deposition in typical acidified regions in North America and Europe. Despite the large deposition of alkaline dust, precipitation can be very acidic in southern China, with pH values of ~4 (Figure 3d). Evidently, the alkaline

**FIGURE 2**

## Map of China showing isolines for precipitation pH, area of the official acid-rain control zone, and the five IMPACTS sites

The acid-rain control zone is highlighted in orange. At the IMPACTS sites, which are marked on the map with a three-letter acronym, air quality and acid deposition are monitored, as well as their effects on soil, soil water, forest vitality, and biodiversity of ground vegetation. The five IMPACTS sites are Cai Jia Tang (CJT), Liu Chong Guan (LCG), Lei Gong Shan (LGS), Liu Xi He (LXH), and Tie Shan Ping (TSP). TSP and LCG are located near big cities, whereas CJT, LXH, and LGS are more regionally representative. LGS is a rural reference site, probably the only rural site in China with such a complete measurement program. The pH isolines are redrawn from a map from the China Meteorological Administration, based on data from 86 monitoring stations (12). The precision and accuracy of the isolines may be low because of the large-scale map and limited sets of data from different types of stations. The overlapping acid-rain control zone and its connection with the acid deposition areas is clearly illustrated. The acid-rain control zone is redrawn from Ref. 13.



dust levels are not high enough to neutralize all the sulfuric and nitric acids. For instance, at the high-deposition TSP site, the SO<sub>4</sub><sup>2-</sup> concentration in the deposition is so high that the annual average pH of deposition would have been ~3 without the calcium and other base cations.

What will happen with alkaline dust in the future? Because particles can cause serious health effects and are fairly easy to remove, at least from large point sources, it is likely that measures for reducing their emission will come before reductions in SO<sub>2</sub> emissions. This may lead to greatly increased acidity of the deposition at the same sulfur concentrations.

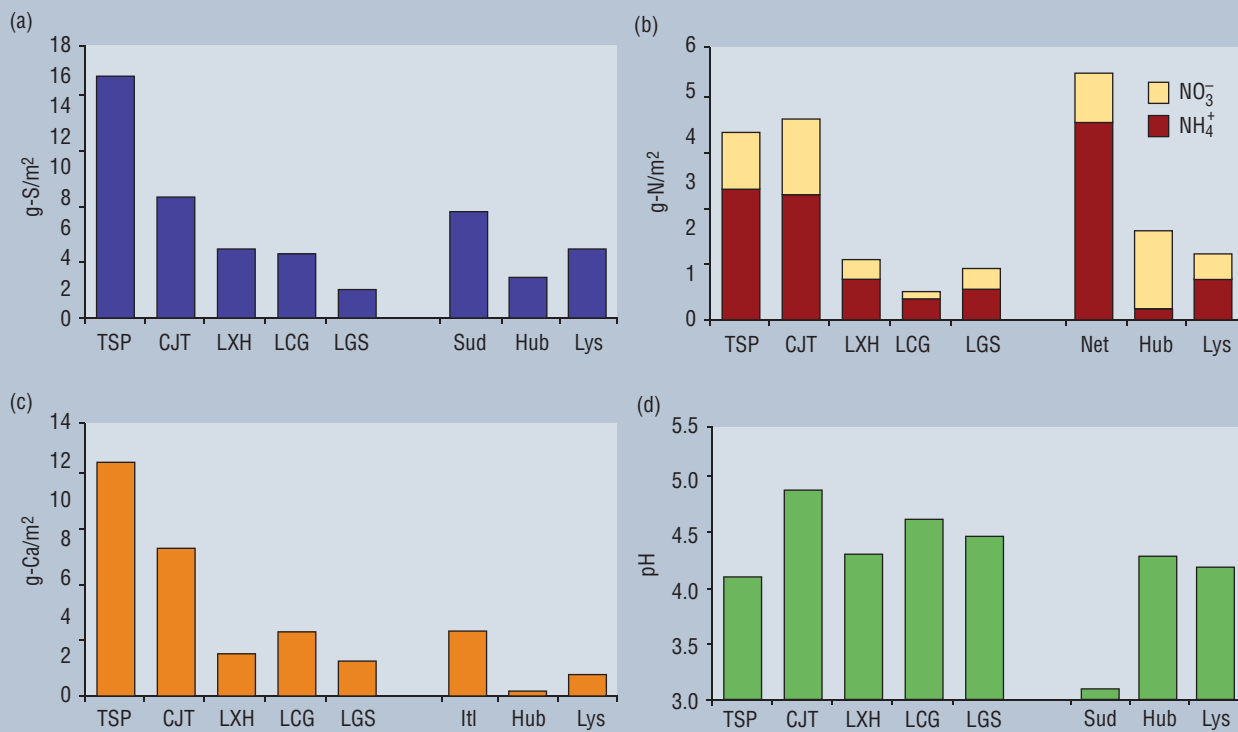
### So, are effects on forest health to be expected?

Given the experiences in central Europe, severe forest damage could be expected from direct effects of air pollutants in the most polluted areas. Forest de-

**FIGURE 3**

## Total deposition of (a) $\text{SO}_4^{2-}$ , (b) nitrogen ( $\text{NH}_4^+$ and $\text{NO}_3^-$ ), (c) calcium, and (d) precipitation pH at the IMPACTS sites in 2003 (15)

Charts include selected data from North America and Europe for comparison. (a–d) Deposition at Hubbard Brook (Hub), N.H., is for 1979–1983 (16). (a–d) The Lysina (Lys) site in the Czech Republic is an example of the most sulfur-polluted part of Europe in ~1980 (17). (a, d) Sulfur deposition and pH data in heavily industrialized Sudbury (Sud), Ontario (Canada) are from 1970; Sudbury serves as an example of high sulfur deposition in North America (18). (b) The 1990 nitrogen deposition in The Netherlands (Net) illustrates a case of high nitrogen deposition in Europe (19). (c) Calcium deposition at an Italian (Itl) monitoring site in Sicily is an example of high inputs of dust from 1995 to 1999 (20). Locations for the five IMPACTS sites listed in this figure are shown in Figure 2.



cline has occurred in some areas of China because of the direct effects of  $\text{SO}_2$ , extremely acidic mist or rain events, and other pollutants (21). One example is on the outskirts of Chongqing, with damage caused by extremely high concentrations of gaseous  $\text{SO}_2$  and/or HF (22). Effects due to soil acidification on forests are much less certain, because few reports are made about widespread damage in more remote areas. However, scientists have stated that soil acidification is likely to have negative effects on forest growth in China (21).

Severe defoliation is observed at two of the five IMPACTS sites—TSP and Liu Chong Guan (LCG). At the TSP site, the needle loss from the dominant masson pine (*Pinus massoniana*) has been considerable (40–50%) and is accompanied by high tree mortality (15). At the LCG site, the needle loss from dominant masson pine increased during the period 2000–2003, followed by a slight decrease in 2004. This defoliation has not been fully diagnosed, because insect attacks (by *Diprion pini* and others) play a major role and indications of climatic stress are seen. Predisposing effects of acid rain cannot be ruled out as an additional cause, but they are difficult to diagnose.

Aluminum concentrations in soil solution are very high. For instance, at the TSP site, average concentrations are >10 mg/L and peak concentrations are >20 mg/L. This is considerably higher than what is considered toxic for tree roots (2 mg/L; 23) and higher than levels observed in most places in Europe. Calcium and magnesium are believed to play an important role in modifying aluminum toxicity (24), and the very high calcium deposition is therefore likely to counteract the toxicity from aluminum under the current conditions. Whether the ratio of calcium to aluminum is sufficiently high to avoid negative long-term effects is uncertain.

During the severe dieback of forest in some heavily polluted areas in central Europe in the mid-1980s,  $\text{SO}_2$  was considered to be the main cause, in combination with frosts and other stress factors. For instance, the annual average  $\text{SO}_2$  concentrations were reported at >100  $\mu\text{g}/\text{m}^3$  in the northwest Czech Republic (25). Although such high concentrations are commonly reported in Chinese industrial cities, the concentrations in the rural forested areas are lower. In 2003, the annual average  $\text{SO}_2$  concentration was ~80  $\mu\text{g}/\text{m}^3$  at the LCG forest site and ~40  $\mu\text{g}/\text{m}^3$  at the TSP site. The critical concentration for

negative effects used in assessments in Europe is  $20 \mu\text{g}/\text{m}^3$  (23).

### What about effects on ground vegetation, biodiversity, and water?

Experience from other parts of the world shows that ground vegetation contains good indicators of the long-term effects of airborne pollutants. This has led to questions about the effects on China's ground vegetation and biodiversity in areas receiving high loads of airborne deposition. So far, monitoring data for ground vegetation exist only for the five IMPACTS sites. The data series are still too short to reveal any dependencies with the pollution pressure; long-term monitoring data are needed to assess the extent and rate of vegetation change.

## Air pollution and acid rain are now considered high-priority areas for the Chinese environmental authorities.

In northern Europe, particularly in Scandinavia, as well as in parts of eastern North America, the main effect of acid deposition has been the acidification of lakes and streams. In Norway, for instance, fish populations have been wiped out in thousands of lakes. Surface-water acidification is not considered a large-scale problem in China, although heavily acidified first-order streams are found in acid-sensitive areas (26). Relatively few investigations on surface-water acidification have been conducted; more data are needed before surface-water acidification can be regarded as unimportant.

### The government recognizes the problem

Air pollution and acid rain are now considered high-priority areas for the Chinese environmental authorities. As a step to curb the problem, China has developed the concept of an acid-rain control zone as the main framework for setting priorities in the acid-rain reduction policy (13; Figure 2).

Various attempts have been made to estimate the societal costs of air pollution and acid rain in China. The Chinese State Environmental Protection Administration (SEPA) has set the costs of acid rain at U.S.\$13 billion. The World Bank estimates the human health costs at U.S.\$11–32 billion, depending on the method used in valuation. The World Bank calculates another U.S.\$5 billion for effects on forests and agriculture (27), whereas a third report suggests slightly less than U.S.\$1 billion (28). Thus, cost estimates of the damage cover a wide range, because the calculations are necessarily based on many uncertain assumptions, such as those involving dose-response functions and how monetary values are ascribed to health and environment effects. Despite the large variations in estimates, all the figures are high; this illustrates the severity of the problem.

### Development is rapidly increasing

The growing demand for electricity is an important feature of China's rapid development. Economic growth is greatest in the eastern and southeastern (i.e., coastal) parts of the country, and these regions constantly need more electricity. These facts, in combination with the desire for further economic development in the western, interior provinces, are the basis for a major national development program called the West to East Electricity Transfer Project (29). For instance, in the Guizhou province, one of the poorest in China, many new coal-fired power plants will be built in the near future. The province already suffers from substantial environmental degradation due to air pollution. Unless strict measures are taken, the ongoing campaign for increasing electricity production will worsen the environmental problems (29).

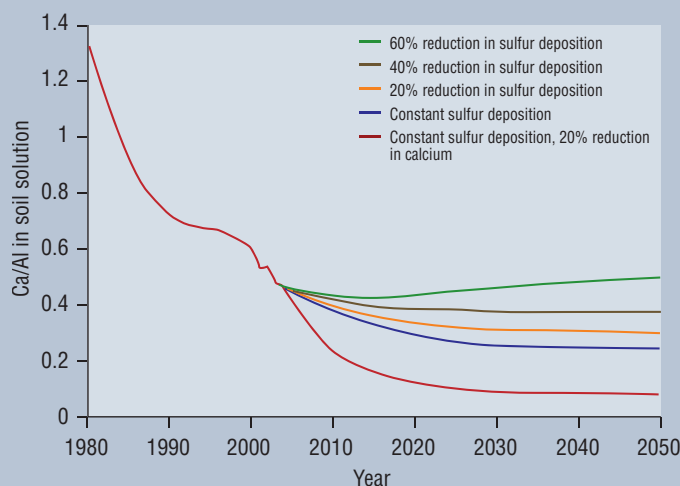
### Can more impacts be expected in the future?

The pollution problems connected to growing energy demand pose a formidable challenge. Unfortunately, a scenario of rising pollution levels with increasing long-range transport cannot be discarded. Several attempts have been made to model the future changes in acidification in China's forest ecosystems. Here, we present one illustrative model calculation with different forecast scenarios for sulfur deposition. Model results of this kind still are quite uncertain for Chinese sites, because of a lack of data. However, the results strongly suggest that considerable reduction in the emission of sulfur is needed to avoid the harmful effects from acidification in the future (Figure 4).

FIGURE 4

### Predictions of the calcium/aluminum molar ratio at IMPACTS site TSP under different scenarios for sulfur deposition

This ratio is used as an indicator for potential long-term forest damage. The current legislation mandates a 20% reduction in sulfur deposition from 2000 to 2010. Calculations were done with the dynamic acidification model MAGIC (30).



The current policy for acid-rain mitigation uses a flat emission reduction goal of 20% from 1995 to 2010 within the acid-rain control zone. This is a reasonable first-generation environmental policy approach that focuses on the right geographical regions. However, the actual impacts on and sensitivities of the ecosystems are not sufficiently taken into account. It is possible to reach the 20% reduction target within the control zone even with increasing deposition in the most sensitive regions. A not-unlikely scenario could be that the emissions in the less-developed interior increase as those along the richer and fast-developing coast decrease.

**Although efforts have been made in recent years to improve the knowledge on acid rain and related effects in China, a great need still exists for better monitoring of the environmental situation.**

A more cost-efficient, effects-based emission reduction policy can be developed by using information on environmental sensitivity, deposition, and emissions. By combining this information on a regional scale, the targets for emission reduction can be made, with the most sensitive environments given priority. If the "critical-load approach" is used, in which the aim is to achieve similar reductions in the gap between the deposition and what the ecosystem can tolerate at all sites, large savings are possible. The critical-load approach was developed in Europe in the 1980s and 1990s and was successfully used to set emission quotas under the recent protocols of the UN Economic Commission for Europe Convention on Long-Range Transboundary Air Pollutants.

The methodology for a similar approach in Asia, including China, has been established (31). As a result of increased efforts at monitoring and capacity building as well as the recognized need for and willingness to establish environmental policies, the time may be right for a more detailed and nationally focused effects-based analysis. Such an approach would enable negotiations among Chinese provinces where emissions and effects are linked. An integrated treatment of air pollutants is important not only for understanding environmental effects but also for reducing emissions of several other pollutants. To find the best options, all important effects—natural environment, human health, and climate—must be considered.

Although efforts have been made in recent years to improve the knowledge on acid rain and related effects in China, a great need still exists for better monitoring of the environmental situation in China to support policy development and follow-up on emission reduction measures. In particular, more

monitoring stations in rural and remote areas are required. In addition, more studies are needed on the impacts on forests and ecosystems.

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