



Acid deposition in Asia: Emissions, deposition, and ecosystem effects



Lei Duan ^{a,*}, Qian Yu ^{a,1}, Qiang Zhang ^{b,1}, Zifa Wang ^{c,1}, Yuepeng Pan ^{c,1},
Thorjørn Larssen ^{d,1}, Jie Tang ^{e,1}, Jan Mulder ^{f,1}

^a State Key Joint Laboratory of Environmental Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing, 100084, China

^b Center for Earth System Science, Tsinghua University, Beijing, 100084, China

^c State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, 100029, China

^d Norwegian Institute for Water Research, 0349, Oslo, Norway

^e Center for Atmosphere Watch and Services, Chinese Academy of Meteorological Sciences, Beijing, 100081, China

^f Department of Environmental Sciences, Norwegian University of Life Sciences, 1432, Ås, Norway

HIGHLIGHTS

- Here review more recent studies on acid deposition in Asia, especially in Eastern Asia.
- Surface waters are generally not sensitive to acid deposition in comparison with soils.
- Soil acidification is not very serious because of base cation deposition, N denitrification, and SO_4^{2-} sorption.

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ABSTRACT

We review and synthesize the current state of knowledge regarding acid deposition and its environmental effects across Asia. The extent and magnitude of acid deposition in Asia became apparent only about one decade after this issue was well described in Europe and North America. In addition to the temperate zone, much of eastern and southern Asia is situated in the tropics and subtropics, climate zones hitherto little studied with respect to the effects of high loads of acid deposition. Surface waters across Asia are generally not sensitive to the effects of acid deposition, whereas soils in some regions are sensitive to acidification due to low mineral weathering. However, soil acidification was largely neutralized by such processes as base cation deposition, nitrate (NO_3^-) denitrification, and sulfate (SO_4^{2-}) adsorption. Accompanying the decrease in S deposition in recent years, N deposition is of increasing concern in Asia. The acidifying effect of N deposition may be more important than S deposition in well drained tropical/subtropical soils due to high SO_4^{2-} adsorption. The risk of regional soil acidification is a major threat in Eastern Asia, indicated by critical load exceedance in large areas.

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1. Introduction

Acid deposition became an issue of major concern in Asia in the early 1980s, nearly one decade after widespread acid deposition was recognized in Europe and North America (e.g. Bhatti, 1992). Before the establishment of national monitoring networks for acid deposition in Asia, isolated surveys of acidity level and chemical composition of rainwater in some Asian countries (such as China,

Japan, and India) indicated the occurrence of acid rain (Bhatti, 1992). Specific regions with decreasing pH trends in precipitation included southern China (south of the Yangtze River) (Zhao and Sun, 1986; Galloway et al., 1987), southern (especially along the east coast) and northeastern India (Varma, 1989), and some areas in Japan (Hara, 1997) and Korea (Chung et al., 1996).

Nationwide surveys of acid rain began in the 1980s. In China, these efforts were sponsored by the National Environmental Protection Agency (NEPA) of China from 1982 (Wang and Wang, 1996; Fujita et al., 2000). The Japanese Acid Deposition Survey (JADS) has been conducted since September 1983 by the Japan Environment Agency (Seto et al., 2004; Okuda et al., 2005). These nation-wide monitoring networks provide the longest record of wet

* Corresponding author.

E-mail address: lduan@tsinghua.edu.cn (L. Duan).

¹ All the co-authors contributed equally to this work.

deposition in Asia. To create a common understanding of the state of acidic deposition problems in East Asia, the Acid Deposition Monitoring Network in East Asia (EANET) began regular deposition monitoring activities in January 2001. Currently, this network consists of 54 monitoring sites in thirteen countries, including Cambodia, China, Indonesia, Japan, Lao P.D.R, Malaysia, Mongolia, Myanmar, Philippines, Republic of Korea, Russia, Thailand, and Vietnam (EANET, 2011). Monitoring of wet and dry deposition, together with ecological impacts, has been conducted as part of the activities of this network.

In addition to monitoring, modeling has been carried out to analyze the spatial variations and source-receptor relations of acid deposition. The early modeling studies were carried out not only for the whole of Asia (Kotamarthi and Carmichael, 1990; Arndt and Carmichael, 1995; Arndt et al., 1998), but also more specifically for Eastern Asia (Huang et al., 1995; Kim and Seog, 2003; Park et al., 2005). Recently, the ensemble-mean depositions of sulfur (S) and nitrogen (N) over Eastern Asia was presented based on eight regional chemical models used in a model inter-comparison study for Asia (MICS-Asia; Wang et al., 2008).

Asia is now the global hotspot of S and N deposition (Vet et al., 2014). Since the early 2000s, the global maximum of both S and N deposition is found in East Asia, including regions like eastern China and South Korea. Other areas of high deposition in Asia include sections of Pakistan, India, Bangladesh, Myanmar, Thailand, Laos, North Korea, and Japan (Vet et al., 2014). Both monitoring and modeling indicate that S and N deposition increased in China from the 1980s–2000s, most likely due to increased sulfur dioxide (SO₂) and nitrogen oxides (NO_x) emissions. Recently, some researchers have suggested that S deposition in China started to decrease as early as 2006 (Zhao et al., 2009, 2013).

Anthropogenic inputs of S and N into terrestrial ecosystems impact soil and surface water, causing acidification and eutrophication (Bouwman et al., 2002). Because long-term data on surface water chemistry are limited in the Asian region, the re-measurement of previously surveyed rivers and lakes or the assessment of public data on water quality are among the few options to assess the current situation regarding water acidification and N leaching (Duan et al., 2011). So far, very few areas have shown water acidification. This is even true for areas with acidic soils and high rates of acid deposition (Komai et al., 2001; Chen et al., 2012). In comparison, soil acidification, as indicated by a significant decrease in soil pH and increase in aluminum (Al) mobilization, and increased N leaching (Aber et al., 2003), has been commonly reported in East Asia (Larssen et al., 2011; Asano and Uchida, 2005; Fang et al., 2011). Most of the available Asian data on the impacts of acid deposition originate from Japan and China. Across these nations, the biogeochemical cycles of major solutes like S, N and calcium (Ca) are shown to be different from those in Europe and North America, probably due to the warm and humid climate, different soil and vegetation types, and different deposition characteristics in China (such as high Ca deposition) (Chen and Mulder, 2007; Larssen et al., 2011).

The critical load of acid deposition is defined as “a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge” (Nilsson and Grennfelt, 1988). The critical load concept was developed in the 1980s to support effective acid rain policy. It has been widely used in international negotiations to reduce of SO₂ and NO_x emissions in Europe (Hettelingh et al., 1995a) with the aim of decreasing S and N deposition in excess of critical loads through cost-optimal emission abatement. Critical loads of acid deposition have been determined and mapped in several studies for a number of regions such as southeastern Asia (Hettelingh et al., 1995b) and

northern Asia (Bashkin et al., 1995), and for some countries including China (Duan et al., 2000a; Zhao et al., 2009), Japan (Shindo et al., 1995) and South Korea (Park and Lee, 2001). These studies, mainly focus on acidification, and may support the use of critical loads as a basis for transboundary pollution impact analysis and co-emission reduction negotiation in Asia in the future.

In this paper, we review and synthesize the current state of knowledge regarding acid deposition and its environmental effects across Asia, in particular East Asia. The trends of emissions of acidifying precursors such as SO₂, NO_x, and ammonia (NH₃), and deposition of S and N in recent years are summarized, and the effects of acid deposition on soil and surface water are discussed. We hope the review may be useful for future studies and policy-making.

2. Emission and deposition

2.1. Emission

Driven by a dramatic economic development, Asian anthropogenic emissions of SO₂, NO_x, and NH₃ show increasing proportions in the global budgets since 1970s (e.g., Fig. 1). For all of the three acidifying precursors, more than 35% of the global emissions were contributed by Asia in 2005, mainly contributed by China and India (EC-JRC/PBL, 2011; Smith et al., 2011).

Emission trends of SO₂, NO_x, and NH₃ varied significantly among Asian regions (Fig. 2). China's SO₂ emissions increased continuously before 2006, and declined after 2006 due to wide application of flue-gas desulfurization (FGD) in power plant units since 2005. Decreasing trends were found for SO₂ emissions from Japan and South Korea due to the much earlier implementation of stringent emission control measures. The rapid increase in SO₂ emission from Southeast Asia has slowed since 1997 (Fig. 2).

A rapid increase in NO_x emissions (counted as NO₂) was observed for China between 1990 and 2011 (Fig. 2), due to the increasing energy consumption in industry and transportation sectors, and lack of adequate control measures. NO_x emissions from China began to decrease in 2012 because of the wide applications of selective catalytic reduction (SCR) in coal-fired power plants (Wang et al., 2014). Decreasing emission trends for Japan and South Korea in recent years were driven by the stringent emission standards implemented for vehicles (Wang et al., 2014).

In contrast, NH₃ emission trends were relatively flat among Asian regions, with the exception of China and Southeast Asia (Fig. 2). NH₃ emissions from China increased during 1980–1996, and then decreased (Kang et al., 2016). The decrease in China's NH₃

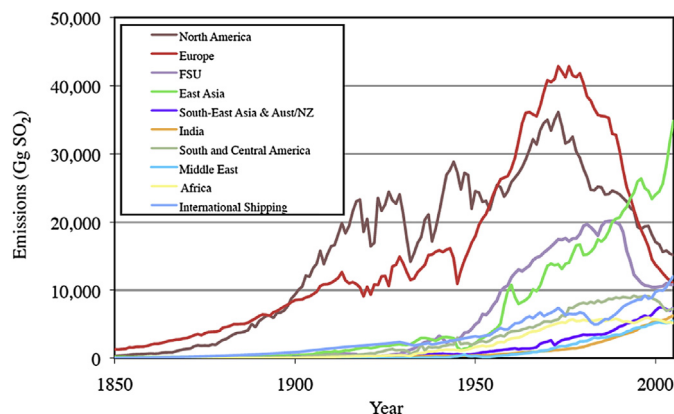


Fig. 1. Global SO₂ emissions by region (North America = USA + Canada; East Asia = Japan + China + South Korea) (Smith et al., 2011).

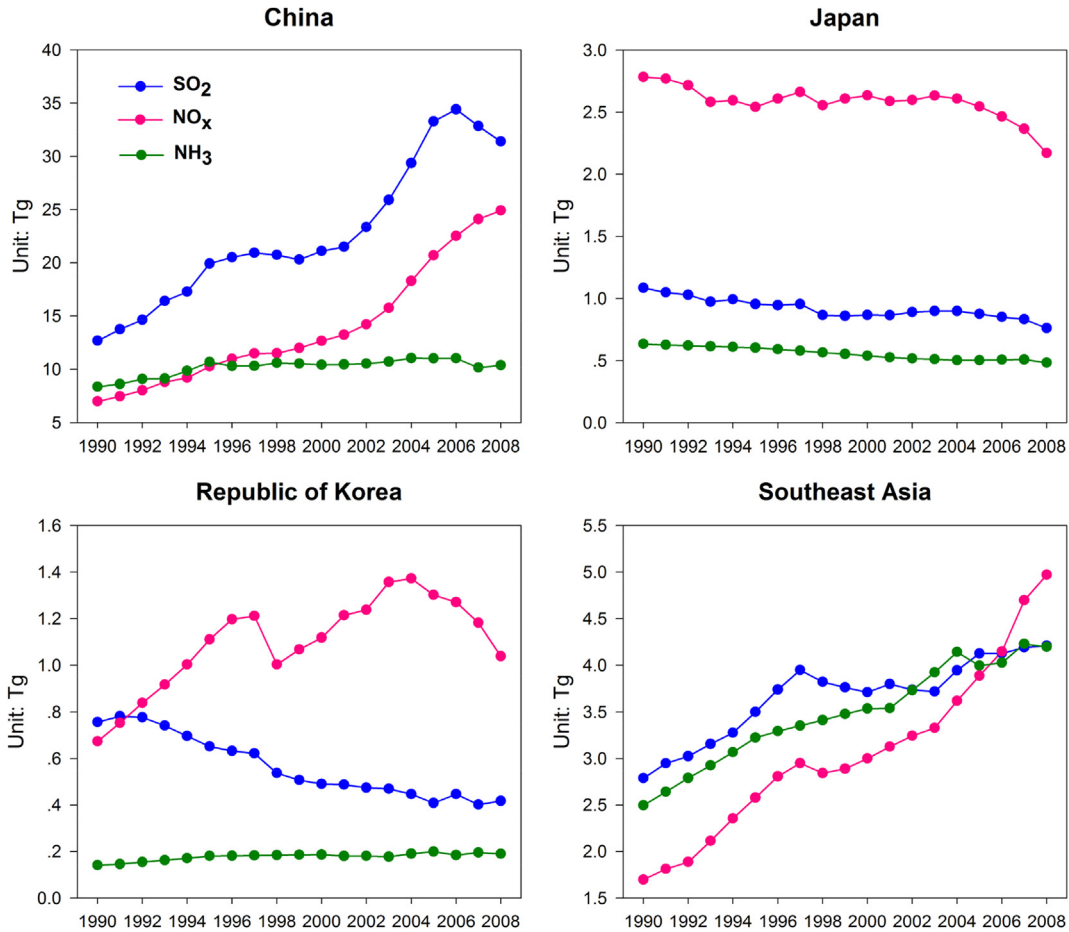


Fig. 2. Emissions of acidifying precursors in several countries/regions in Asia (Data source: MEIC for China, www.meicmodel.org; REAS for other regions, Kurokawa et al., 2013).

emissions was attributed to a decline in ammonium bicarbonate applications as fertilizer. A sharp increase in NH₃ emission from Southeast Asia was mainly driven by fertilizer applications (Kurokawa et al., 2013).

Fig. 3 presents the spatial distribution of Asian SO₂, NO_x, and NH₃ emissions in 2010 (from MIX inventory; Li et al., 2015). Mainly produced during fuel consumption, SO₂ and NO_x emissions were concentrated in populated regions with high energy consumption like East China and India. NH₃ emissions were distributed widely

among regions with intensive agriculture activities, such as China, India and Southeast Asia.

2.2. Deposition

Observational studies on spatial distributions of S and N deposition for the whole of Asia are very limited. The annual bulk S deposition at five forest sites in southern and southwestern China ranged from 24 to 160 kg ha⁻¹ yr⁻¹, which is in the same range as,

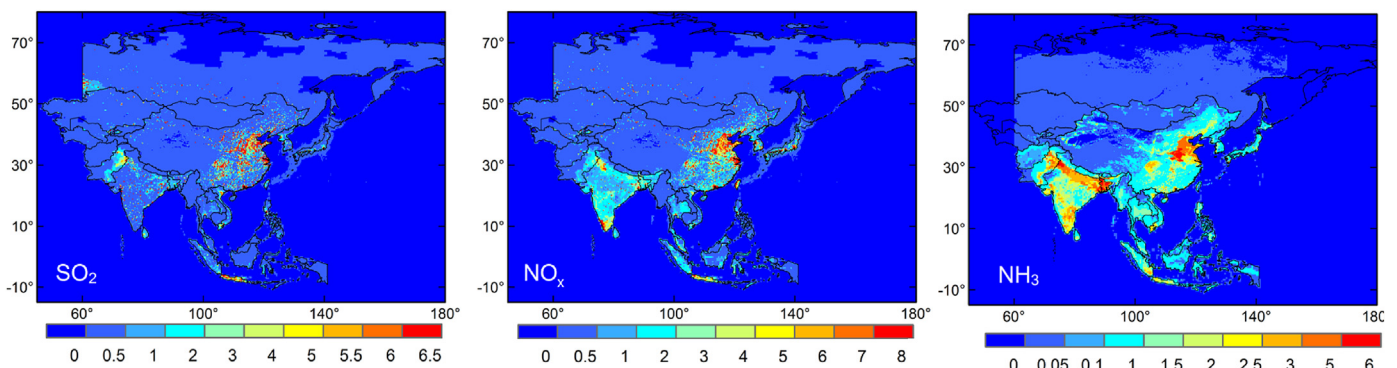


Fig. 3. Emission of acidifying precursors in Asia in 2010 (Unit: Gg/gird; Data source: MIX Asian emission inventory; Li et al., 2015). The map resolution is 0.25° (latitude) by 0.25° (longitude).

or higher than, that seen in most of central Europe in 1980, when acid deposition was at its peak (Larssen et al., 2006). Very high total deposition of S and N also occurred in north China (Pan et al., 2012, 2013). For example, higher S deposition was observed at industrial and urban sites ($50\text{--}100\text{ kg ha}^{-1}\text{ yr}^{-1}$), reflecting a higher contribution of dry deposition (mostly of gaseous SO_2 which accounted for about 70%) than of wet deposition (Pan et al., 2013). In addition, the total N deposition in northern China ($30\text{--}100\text{ kg ha}^{-1}\text{ yr}^{-1}$) was also significantly larger than that in other Asian countries such as Japan because of high rates of wet deposition and gaseous NH_3 dry deposition (Pan et al., 2012). The annual S deposition for six remote EANET sites in Japan during 2000–2004 ranged from 1.9 to $7.5\text{ kg ha}^{-1}\text{ yr}^{-1}$. For the same six sites, the N deposition ranged from 1.5 to $6.4\text{ kg ha}^{-1}\text{ yr}^{-1}$ (about half as NO_3^-), which was lower than values for urban sites in Japan, with average values of $7.8\text{ kg ha}^{-1}\text{ yr}^{-1}$ (Seto et al., 2007).

Most of monitoring data showed a decreasing trend of wet deposition of SO_4^{2-} and thus an increasing trend of precipitation pH in Japan since early 1990s (Seto et al., 2004), in parallel with a decreasing trend in the SO_2 emission from Japan. For example, the acid deposition survey by the Japan Environment Agency showed an increasing trend of pH and a decreasing trend for both S and N deposition from the period 1986–1988 to the period 1989–1993 (Hara, 1997). The pH data collected at the 29 stations in the first period showed a range of annual mean pH from 4.4 to 5.5 (Kitamura et al., 1991). The range changed to 4.5 to 5.8 during the second period (Hara, 1997).

Although the monitoring of S and N deposition was very limited in China, both in space and time, long-term records of precipitation pH are available. The overall precipitation pH showed three stages (Fig. 4). In the period 1992–1999, the precipitation pH showed an increasing trend. During 2000–2006, however, a decreasing trend of the precipitation pH was observed in North China, Central China, East China, and South China. The precipitation over North China, Central China, and South China became more acidified during 1992–2006, with more pronounced trends in North China and the north of Central China. A slight increase in the precipitation pH was found in Southwest China, an area characterized by the most severe acid rain for about two decades since the early 1980s. Consequently, the center of the most severe acid rain area, south of the Yangtze River, moved eastwards (Tang et al., 2010). Accompanying the rapid reduction of SO_2 emission after 2006 (Fig. 2), precipitation pH began to increase (Fig. 4).

Based on the EANET dataset of precipitation chemistry, yearly wet deposition of SO_4^{2-} and NO_3^- was averaged for all sites in each country (Fig. 5). The annual SO_4^{2-} deposition was significantly larger in China than in South Korea and Japan, whereas the NO_3^- deposition was similar in these countries. However, the pH of rainfall in China was higher due to the high buffering of precipitation acidity by emissions of basic particulate matter (PM), including soil dust (Larssen and Carmichael, 2000), anthropogenic dust (Zhu et al., 2004; Lei et al., 2011), and NH_3 (mainly from agricultural activities; Kang et al., 2016). For example, the Ca^{2+} concentration in rainwater in northern China is much higher than in southern China or in the United States and other industrial nations (Wang and Wang, 1996; Wang et al., 2012; Cao et al., 2013). It is estimated that if the soil-derived bases (particularly CaCO_3) were eliminated from the atmosphere, the precipitation pH in these northern sites would average approximately 3.5 (Galloway et al., 1987).

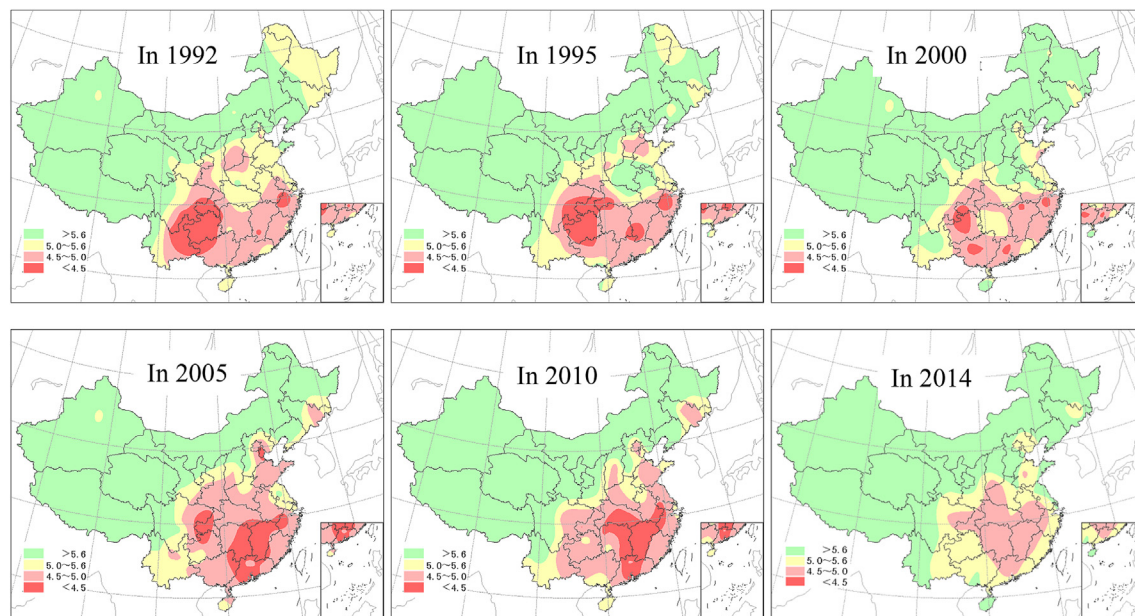
Long-range transport of acidic pollutants has been suggested to occur from the Asian continent to Japan and Korea. This may be one reason why the trend of S and N deposition in Korea and Japan (Fig. 5) did not coincide well with the SO_2 and NO_x emission (Fig. 2). For example, high atmospheric non-sea salt SO_4^{2-} concentration observed at the Japan Sea coast during southwesterly wind, and the

concentration of anthropogenic sulfate and nitrate aerosol at the central North Pacific site almost doubled from 1981 to the mid-1990s, parallel to increased SO_2 emission from China (Prospero et al., 2003; Hideaki et al., 2008). Anthropogenic SO_2 and NO_x emission from China contributed to the increase in SO_4^{2-} and NO_3^- concentration in the wet-only samples from 2000 to 2007 in coastal areas of Korea (Park et al., 2015). Higher S and N deposition occurred on the western coasts of Korea than on the eastern coasts (Kang et al., 2004). During 2000–2007, when SO_2 emissions from China increased by 53%, there was a longitudinal gradient in urban SO_2 concentration in Japan, with values decreasing the further the site was from the Asian continent. This result demonstrates that, in spite of the relatively short tropospheric lifetime of SO_2 , the transport of increasing SO_2 from the Asian continent can partially counteract the local reduction of SO_2 emission downwind, and even override it in some southwestern areas of Japan (Lu et al., 2010b). After SO_2 emission from China began to decrease after 2006, ambient air SO_2 concentration and SO_4^{2-} concentrations in precipitation also started to decrease, while rain pH increased in the whole of East Asia (Lu et al., 2010b).

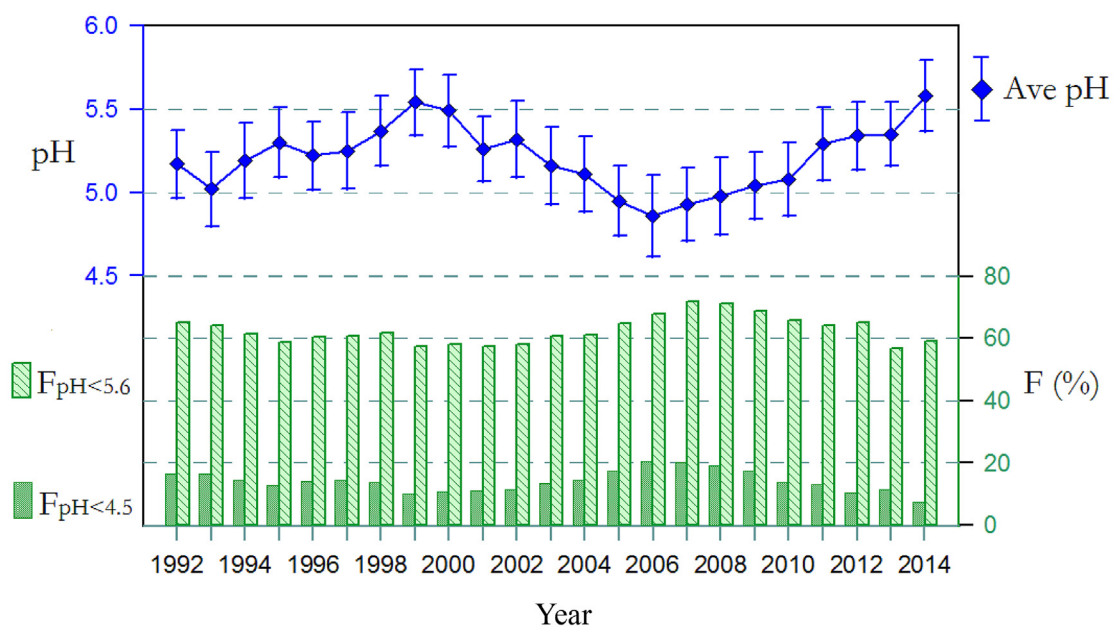
Unlike S, atmospheric N deposition rates in eastern Asia have dramatically increased during recent decades due to the emissions of NO_x and NH_3 from combustion processes and agricultural activities, respectively, not only from China (Liu et al., 2011), but also from Japan (Kannari et al., 2001). For example, average bulk N deposition of all available monitoring sites throughout China increased from $13.2\text{ kg ha}^{-1}\text{ yr}^{-1}$ in 1980s to $21.1\text{ kg ha}^{-1}\text{ yr}^{-1}$ in 2000s (Liu et al., 2013). It has also been reported that forests in Central Japan revealed a high level of N deposition and NO_3^- in stream water (signal for N saturation) (Baba et al., 1995; Baba and Okazaki, 1998; Ohruji and Mitchell, 1997, 1998; Matano et al., 2001; Ham et al., 2010). Wet deposition of N varied between 5.7 and $16.7\text{ kg ha}^{-1}\text{ yr}^{-1}$ (with similar contribution of NH_4^+ and NO_3^-) from rural sites to urban areas during 1999–2002 (Paramee et al., 2005). A literature review on N deposition from 69 forest ecosystems at 50 sites throughout China indicated that the wet deposition of N ranged from 2.6 to $48.2\text{ kg ha}^{-1}\text{ yr}^{-1}$, with an average of $16.6\text{ kg ha}^{-1}\text{ yr}^{-1}$ (Fang et al., 2011). Ammonium was the dominant form of N at most sites, accounting for, on average, 63% of total inorganic N deposition (Fang et al., 2011).

A decreasing trend of wet deposition has been observed in Southeast Asia for SO_4^{2-} but not for NO_3^- (EANET, 2006, 2011), which coincides with the trend of SO_2 and NO_x emissions (Fig. 2). The wet deposition of SO_4^{2-} also decreased by about 5% between 2000–2002 and 2005–2007 in northeast India, one of the areas with highest S deposition in India ($11.0\text{ kg ha}^{-1}\text{ yr}^{-1}$ in 2005–2007) (Vet et al., 2014). However, the wet deposition of N increased by more than 30% in this area, which measured among the highest wet N deposition rates in the world with values of about $20\text{ kg ha}^{-1}\text{ yr}^{-1}$ (Vet et al., 2014).

A recent study using the Nested Air Quality Prediction Modeling System (NAQPMS), that coupled the cloud-process and aqueous chemistry module from the Community Multi-scale Air Quality (CMAQ) modeling system, indicated that a very high wet deposition of both S and N (in the range of $16\text{--}25\text{ kg ha}^{-1}\text{ yr}^{-1}$ and $21\text{--}32\text{ kg ha}^{-1}\text{ yr}^{-1}$, respectively) occurred in northern, southwestern, and eastern China in 2007 (Ge et al., 2014). The total deposition of S and N was even higher than $70\text{ kg ha}^{-1}\text{ yr}^{-1}$ and $50\text{ kg ha}^{-1}\text{ yr}^{-1}$ respectively in these areas (Fig. 6). Strong neutralization of precipitation by soil aerosols over northeastern Asia was also estimated, with the increase in annual mean pH by 0.8–2.5 in northern China and Korea, while less than 0.1 in southern China and Japan (Wang et al., 2002). In comparison with natural sources such as desert, which mainly contributes to high Ca^{2+} deposition in northern and northwestern China, the anthropogenic sources of Ca^{2+} -content particulate matter such as cement



(a)



(b)

Fig. 4. Spatial and temporal distribution of precipitation pH in China: (a) Maps of average annual precipitation pH; (b) Trend of nationally average precipitation pH and acid rain frequency (F) during 1992–2014 (Data from Acid Rain Monitoring Network; CMA, 2014).

production and coal combustion result in high Ca^{2+} deposition in eastern and southern China (Fig. 7). The highest Ca^{2+} deposition modeled is even comparable with the S deposition on an equivalent basis, indicating significant neutralization of precipitation acidity by base cations.

3. Effects

Here only surface water and soil acidification/eutrophication were focused on, although acid deposition has other impacts on agriculture, human health, and infrastructure.

3.1. Water acidification

Surface water acidification, causing severe ecological damage in Scandinavia and to some extent also elsewhere in northern Europe and North America, used to be the main concern of damage caused by acid deposition. Although extremely high deposition rates for SO_4^{2-} occur in East Asia, especially in China and Japan, only very few streams in forested catchments in central Japan and on islands have suffered from acidification (Nakano et al., 2001; Kurita and Ueda, 2006; Yamada et al., 2007; Matsubara et al., 2009; Nakahara et al., 2010). A large buffering capacity of the soil and high

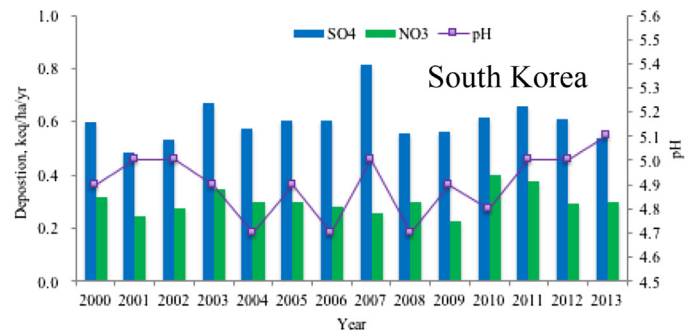
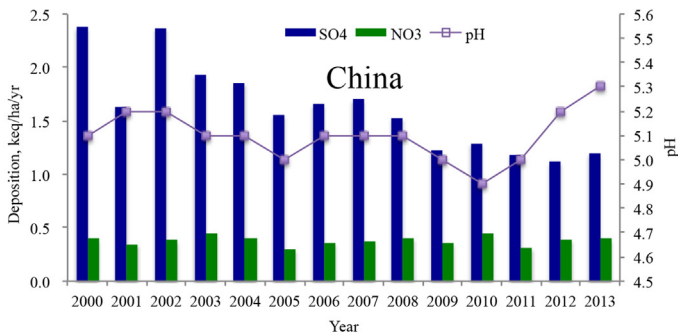
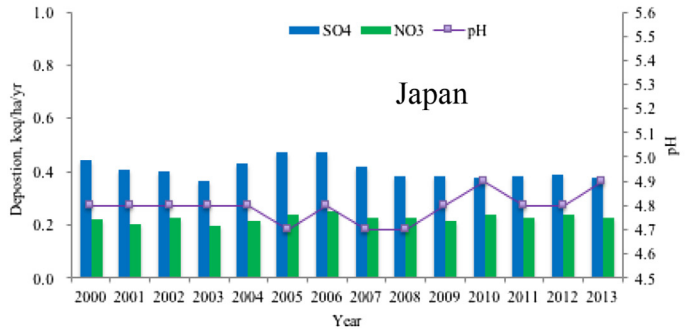
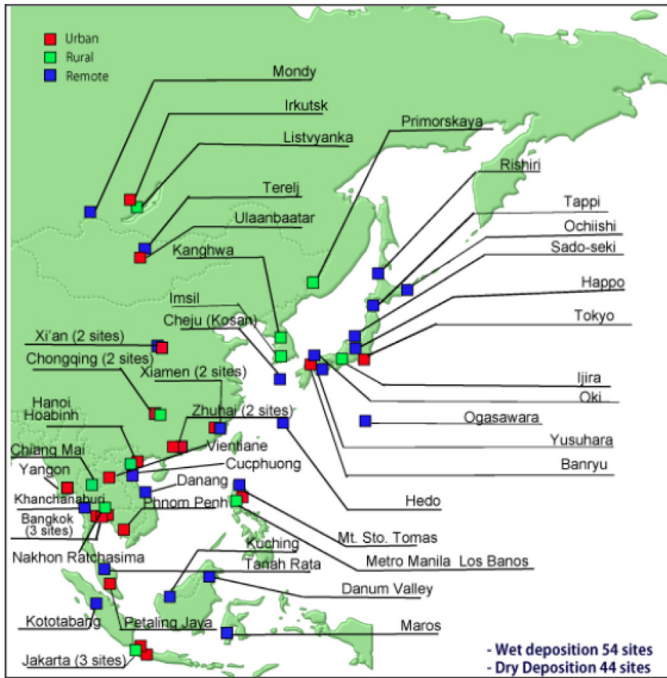


Fig. 5. Monitoring sites of EANET (Acid Deposition Monitoring Network in East Asia) and trend of average wet deposition in China, Japan, and South Korea (Data source: <http://www.eanet.asia/product/index.html>).

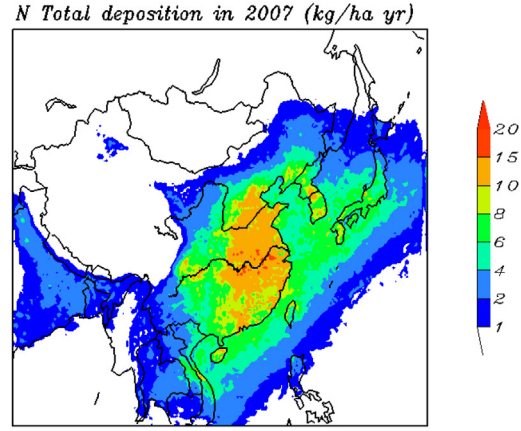
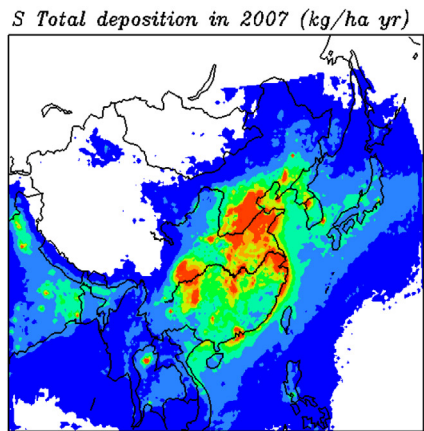


Fig. 6. S and N deposition in Asia modeled by the Nested Air Quality Prediction Modeling System (NAQPMS) (Ge et al., 2014).

alkalinity of the inland waters inhibit acidification of the inland water ecosystem in Japan under current levels of acidic precipitation (Suzuki, 2003). The survey of rain and head water chemistry between 1991 and 1997 revealed that the watershed ecosystem did

not show any direct evidence of water acidification in spite of decades of elevated H^+ deposition (estimated at $0.43 \text{ keq ha}^{-1} \text{ yr}^{-1}$ or more since 1960s; Ikeda and Hmada, 2001). Many monitoring studies conducted during the 1990s showed that stream waters in

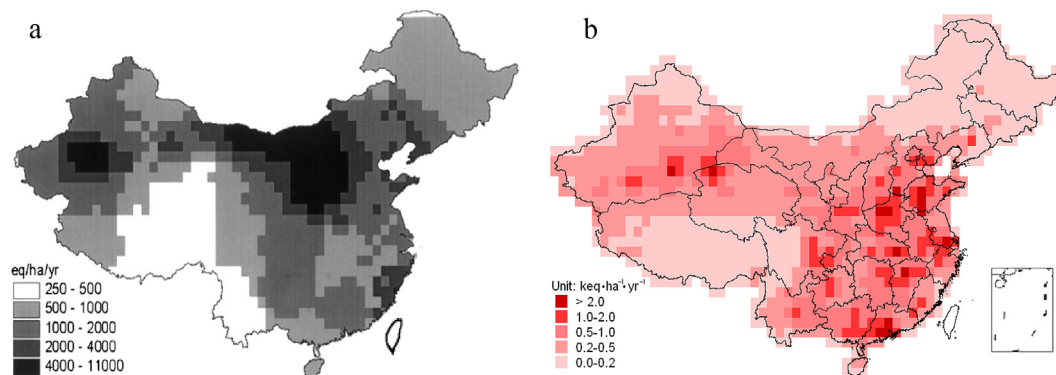


Fig. 7. Ca^{2+} deposition estimated with origin in the desert areas (a; Larssen and Carmichael, 2000) and anthropogenic emission in 2005 (b; Zhao et al., 2011) in China.

Japan (e.g., Baba and Okazaki, 1998; Komai et al., 2001; Ohte et al., 2001) and China (Duan et al., 2000b; Hao et al., 2001a; Ye et al., 2002) were well buffered (with high ANC) and there were no signs of a long-term trend of pH decline.

Although some aquatic ecosystems with low alkalinity, considered to be sensitive to acidification, are present in China, especially in the south and northeast, until recently detailed surveys of those headwater streams were scarce. A recent survey of headwater streams showed enhanced concentration of SO_4^{2-} and NO_3^- (higher than 0.5 meq/L and 0.1 meq/L, respectively) in stream waters in southwestern China under very high atmospheric deposition of S and N (Fig. 8). Much lower concentration of SO_4^{2-} and NO_3^- occurred in stream waters in Japan and South Korea (Fig. 8).

As can be seen from Fig. 8, there may be a big difference between China and Japan in the acidifying potential of N, which is mainly due to the difference in N input (relatively small in Japan as in the US, and much higher in China as in Northwest Europe; Figs. 5 and 6). In Japan even with N deposition below $10 \text{ kg ha}^{-1} \text{ yr}^{-1}$ ($1.0 \text{ g m}^{-2} \text{ yr}^{-1}$), a generally used threshold above which nitrate leaching to streams is predicted (Dise et al., 1998), there was evidence of elevated NO_3^- concentration of stream water in a larch forest (Nakahara et al., 2003). The enhanced NO_3^- discharge may accelerate surface water acidification in central Japan. One example was the Lake Ijira catchment, which is one of the forested catchments with wet N depositions ($19 \text{ kg ha}^{-1} \text{ yr}^{-1}$) among the highest levels observed in Japan (Nakahara et al., 2010). In contrast, considerable NO_3^- leaching to stream waters are not likely to occur in China, unless N deposition exceeds $25 \text{ kg ha}^{-1} \text{ yr}^{-1}$ ($2.5 \text{ g m}^{-2} \text{ yr}^{-1}$, or even more in some region) (Fig. 8).

Typical for much of southern China, where soil and forest types (Haplic Acrisol and subtropical evergreen coniferous forests, respectively) are similar as in SW2, and N deposition (both $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^+\text{-N}$) is commonly high, is that forested catchments act as large N sinks, shown by relatively low NO_3^- concentration (about a few mg/L) in stream water (Larssen et al., 2011). Most of the NO_3^- is lost from solution upon its transport through the groundwater discharge zone (GDZ) before reaching the stream, due to denitrification (Zhu et al., 2013a). This is confirmed by studies of natural abundance, where enrichment of ^{15}N and ^{18}O in the GDZ were attributed to the denitrification process (Yu et al., 2016). Denitrification is an acid neutralizing process, and contributes to the higher soil pH in the GDZ than on the well-drained hill slopes. Denitrification results in not only the production of N_2 gas, but also emission of nitrous oxide (N_2O), a potent greenhouse gas. In wet years, N_2O emission is accounted for close to 10% of the total N sink at the Tieshanping site in southwestern China (Zhu et al., 2013b). Thus, the groundwater discharge zones of N-saturated forests in southern

China are hot-spots of denitrification, thus protecting stream water from excessive acidification due to NO_3^- leaching. In addition, relative low leaching of SO_4^{2-} (in comparison with S deposition) occurred in many stream waters in Japan and southwestern China even with high S deposition (Fig. 8), indicating considerable S sink in the forested catchments, mainly due to SO_4^{2-} adsorption by soils (Vogt et al., 2007; Huang et al., 2015).

3.2. Soil acidification

Most of streams studied in Japan and China are not acidified, although hillslopes receive acidic precipitation similar to or higher than acid-sensitive areas of eastern North America and northern Europe, and soil water in the root zone is often found to be acidified (Miyanaga and Ikeda, 1994; Sato and Takahashi, 1996; Toda et al., 2000; Asano and Uchida, 2005; Iizumi et al., 2005; Ebise and Nagafuchi, 2002; Larssen et al., 2011).

Acidification of soils has been widely observed in China. In the 1980s, soil pH decreased by 0.1–0.5 units on Lushan Mountain in southern China (Pan et al., 1993). On Hengshan Mountain in southern China, surface soil pH decreased by 0.5–1.1 for different soil types from 1983 to 2001 (Wu et al., 2005). A decreasing trend in surface soil pH during 1980–2009 was also found on Taishan Mountain in northern China (Zhang and Li, 2010). Regionally, significant soil acidification across major forest ecosystems and grasslands was found in China during 1980s–2000s (Yang et al., 2012, 2015). Maximum decrease in soil pH was found in evergreen forests (on average from 5.4 in the 1980s to 4.8 in the 2000s) in southern China (Yang et al., 2012). Soil pH in the surface layer declined significantly over the last two decades across grassland in northern China, with an overall decrease of 0.63 units (Yang et al., 2012). The decrease in soil pH in China may be partly attributed to acid deposition. However, soil pH was also decreased by 0.5 units due to overuse of N fertilizers rather than acid deposition (although the mechanism of soil acidification is the same) (Guo et al., 2010).

In Japan and Korea, dominant soils are developed from granite bedrock, and they are characterized by a low acid buffering capacity (Yagasaki et al., 2001). Soil acidification was also observed in Japan. For example, the average pH of the surface mineral soils in a forested catchment in central Japan decreased from 4.5 in 1990 to 3.9 in 2003, with the rate of decrease of 0.07 pH units per year noticeably higher than many previously reported values in Europe and North America (Nakahara et al., 2010). Surface soils of Andosols also showed decreases in soil pH, the dissolution of aluminum, and the formation of precipitates, such as aluminum hydroxysulfate and basic iron sulfate (Onodera et al., 2002; Takahashi and Higashi, 2013).

In comparison to soil, soil water in the root zone is more

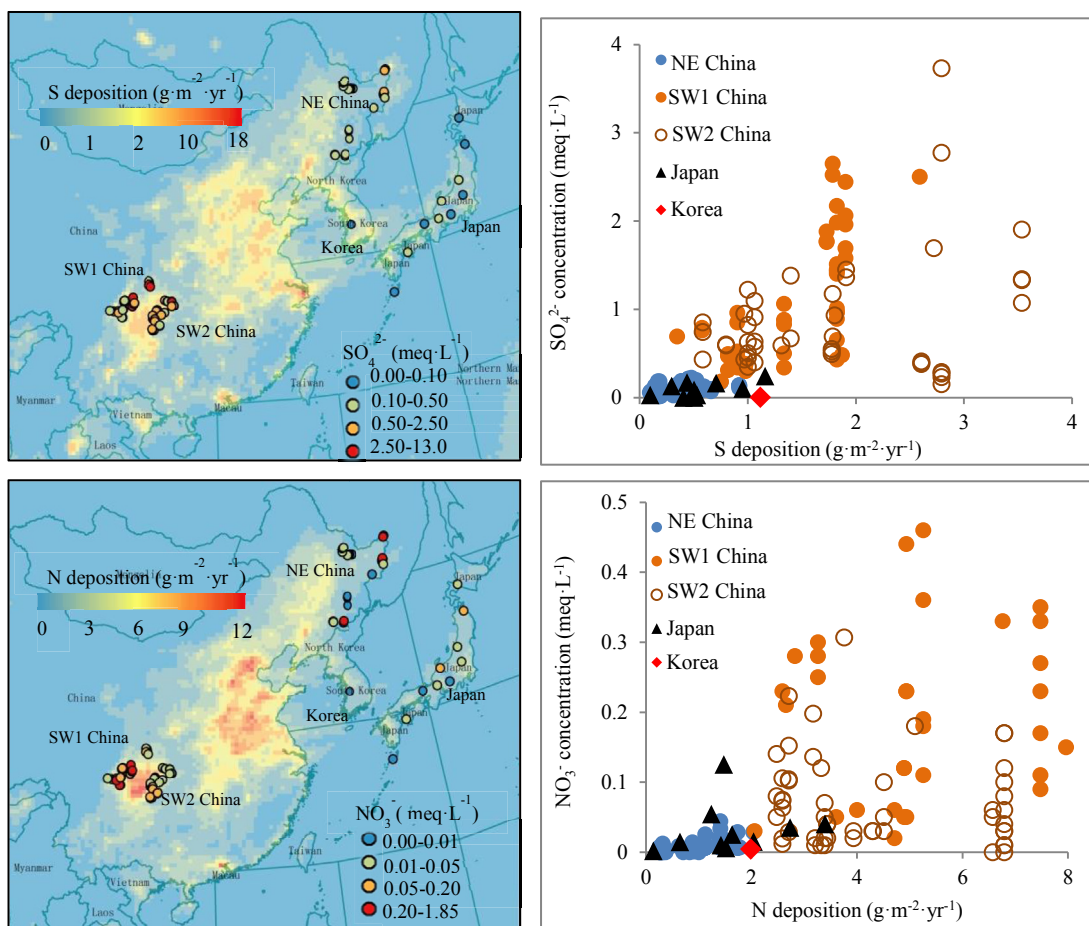


Fig. 8. SO_4^{2-} and NO_3^- concentrations in stream waters in Eastern Asia. Data from three regions of China: northeast (NE), west of southwest (SW1), and east of southwest (SW2) (Chen et al., 2012; Xu et al., 2013). Data from Japan (Baba et al., 2001; Ebise and Nagafuchi, 2002; Farah et al., 2006; Iizumi et al., 2005; Kawakami et al., 2001; Kobayashi et al., 2013; Komai et al., 2001; Nakahara et al., 2003; Nakahara et al., 2010; Sato and Takahashi, 1996). Data from Korea (Jeon and Nakano, 2001). Left figures show S and N deposition modeled by CMAQ (Zhao et al., 2013).

dynamic and responds more quickly to acid deposition. In order to supplement the existing monitoring data 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; Larssen et al., 2006). Precipitation composition, as well as soil, water, and vegetation effects were being intensively studied at five forested sites, which represented acid-sensitive forested ecosystems in southern and southwestern China. All sites were exposed to ambient acid deposition. Representative for areas with high S and N deposition, the Tieshanping site in southwestern China showed significant soil acidification through significantly higher leaching of strong acid anions (SO_4^{2-} and NO_3^-) in soil solution (SS) than base cations (especially Ca^{2+}) (Fig. 9; Larssen et al., 2011). Elevated H^+ fluxes and associated high inorganic monomeric aluminum (Al_i) fluxes were observed in soil solution (Fig. 9). It should be noted that the Ca^{2+} input fluxes (in throughfall) are high compared to Ca deposition in most other countries (due to elevated atmospheric transport of Ca^{2+}). Without this input of Ca^{2+} , the flux of H^+ and Al_i in soil solution would be substantially greater.

Although forest dieback in Eastern Asia associated with acid deposition has not been widespread, several phenomena such as abnormal defoliation have been reported, not only in Japan (Izuta, 1998; Nakahara et al., 2010) but also in China (Larssen et al.,

2006; Wang et al., 2007) and Korea (Lee et al., 2005). For example, severe defoliation is observed at two of the five IMPACTS sites, Tieshanping and Luchongguan (near Guiyang in Guizhou province), with 40–50% of defoliation and maximum 6% tree death of Masson pine (*Pinus massoniana*) at the Tieshanping site (Larssen et al., 2006; Wang et al., 2007). This was attributed to air pollution and soil acidification, although other stress factors such as insect attacks and summer drought may have been important as well (Wang et al., 2007). A literature review of the effect of acidification on forests in Japan indicated that the most important indicator for soil acidification is the Ca/Al molar ratio of soil water in the root zone (Hirano et al., 2007). Japanese coniferous tree species such as Japanese cedar and red pine are relatively sensitive to a reduction in $(\text{Ca} + \text{Mg} + \text{K})/\text{Al}$ molar ratio in soil solution (Izuta, 1998). These results were similar as other earlier studies in Europe and North America (e.g., Sverdrup and Warfvinge, 1993). Recently, soil acidification, with nutrient imbalance and low $(\text{Ca} + \text{Mg} + \text{K})/\text{Al}$ molar ratios (<10), was found to hamper the sound growth of both Japanese cedar and Japanese cypress, and is one of the most likely causes of the decline of temple and shrine forests in Kyoto (Ito et al., 2011). The $(\text{Ca} + \text{Mg} + \text{K})/\text{Al}$ molar ratio of the soil water at Tieshanping, southwest China was lower than 2.0 (Huang et al., 2014), the critical limit widely accepted for Masson pines (Gao et al., 1992), which are widely distributed in the subtropical areas of southern and southwestern China. This species seems very sensitive to soil

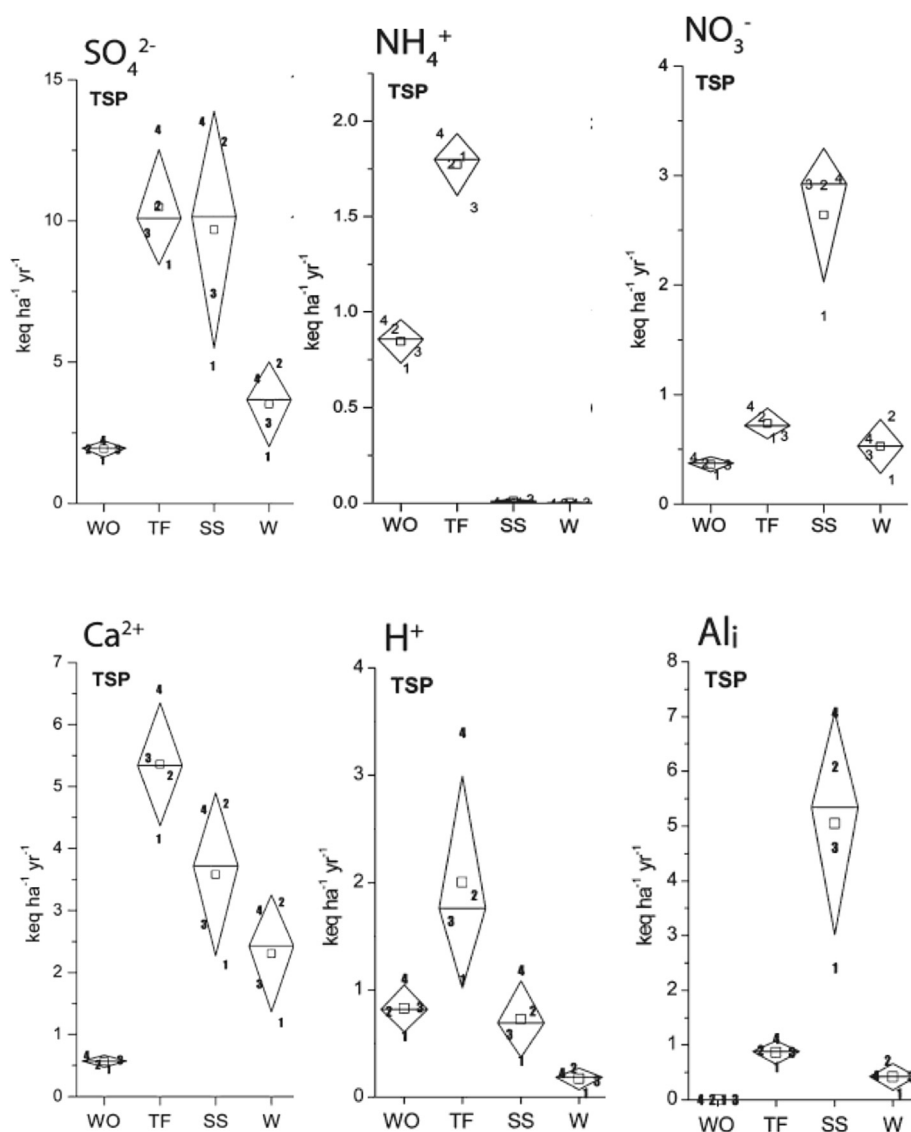


Fig. 9. Fluxes of SO_4^{2-} , Ca^{2+} , NH_4^+ , NO_3^- , H^+ , and inorganic monomeric aluminum (Al_i) in wet deposition (marked WO), throughfall (TF), soil-water (SS) and streamwater (W) in Tieshanping (TSP), a subtropical forested catchment in Chongqing, southwest China (Larssen et al., 2011). Assume that throughfall fluxes represent a reasonable estimate of total deposition. The horizontal lines in each diamond box show the median, the range of the boxes is SD and the square inside each box shows the average. The numbers indicate fluxes for the individual years (1 represents 2001, 2 represents 2002, etc.). The size (height) of the diamond illustrate the variation between plots and between years; the numbers show the variation between years only.

acidification in comparison with some species in Europe and North America, where 1.0 is widely used as a critical limit of $(\text{Ca} + \text{Mg} + \text{K})/\text{Al}$ (Sverdrup and Warfvinge, 1993).

Generally, chemical weathering of minerals is the only long-term sustainable source of alkalinity neutralizing acid input in North America and Northern Europe. Because of the distribution of soils (such as Haplic Podzol and Albic Luvisol developed from granite) with lower weathering rates in northeast China (Duan et al., 2002), where the natural conditions such as the soil and vegetation types are quite similar to those in North America and Northern Europe, the soil cannot provide a strong acidity buffering capacity. Although a much higher soil weathering rate occurred for Haplic Luvisol in the west part of southwestern China (indicated as SW1 in Fig. 8), the soil weathering rates of Haplic Acrisol in the east part of southwestern China (indicated as SW2 in Fig. 8) and large region in southern China were similar to those in northeastern China (Duan et al., 2002). More significant acidity buffering

capacity is therefore produced by several other processes including high Ca^{2+} deposition.

The overall catchment budget (i.e., the difference between throughfall deposition flux and stream water output) indicated considerable retention of N and S in the five forested catchments in southern and southwestern China (Fig. 9; Larssen et al., 2011). As for N, several studies based on the mass balance approach indicate that after its deposition, NH_4^+ nitrifies to NO_3^- (Larssen et al., 2011; Huang et al., 2015). Since relatively little NO_3^- uptake is reported in N saturated forests due to limitation of other nutrients (Huang et al., 2014), significant NO_3^- leaching occurs, which is associated with strong acidification in the rooting zone of the soil (Chen et al., 2004). The acidifying effects of N deposition were also found in field studies at Dinghushan in the tropics of China (Lu et al., 2014), where the increased Al and decreased base cation concentrations were attributed to the loss in biodiversity (Lu et al., 2011). A literature review on N leaching from 69 forest ecosystems at 50 sites

throughout China indicated that overall 22% of the N input via throughfall was leached from soil, which is lower than the 50–59% observed for European forests (Fig. 10; Fang et al., 2011). Note that there may be large differences in NO_3^- fluxes between soil water (Fig. 10) and stream water (Fig. 8). In China, elevated N leaching by soil water (e.g., NO_3^- -N concentrations exceeding 1.0 mg L^{-1}) occurs in forest ecosystems when they receive N deposition of more than $5 \text{ kg ha}^{-1} \text{ yr}^{-1}$ (Fang et al., 2011), while N deposition needs to be above $25 \text{ kg ha}^{-1} \text{ yr}^{-1}$ for considerable N leaching into stream waters (Fig. 8). The big gap between the two thresholds may attribute to the active denitrification occurring in the groundwater discharge zones, indicated by much lower NO_3^- fluxes in stream water than in soil water (Fig. 9; Larssen et al., 2011; Zhu et al., 2013a). The N input thresholds for elevated NO_3^- leaching in soil water seems lower than those found in Europe and North America (at approximately $10 \text{ kg ha}^{-1} \text{ yr}^{-1}$; Dise et al., 1998), where the denitrification rate in head water catchments may be relatively limited.

The main cause of substantial net loss of SO_4^{2-} within the catchment is likely the adsorption of SO_4^{2-} in soil layers (Vogt et al., 2007; Duan et al., 2013; Huang et al., 2015), which produces OH^- and can neutralize soil acidification. It coincides with very low leaching of SO_4^{2-} by stream waters in southern China and Japan with high S deposition (Fig. 8). The anion exchange capacity is high in these soils with low pH and a high content of Al oxides (Vogt et al., 2007). By contrast, soil nitrification produces H^+ to acidify soil, while denitrification mainly neutralizes surface water acidification. It seems that the acidifying effect of N deposition may be more important than S deposition in the well-drained tropical/subtropical soils. For some catchments where SO_4^{2-} saturation may occur, with the SO_4^{2-} flux in soil water similar as that in throughfall (Fig. 9), the SO_4^{2-} reduction to sulfides, as implied by high groundwater table and effective denitrification, may be another SO_4^{2-} sink in the sub-soil or even more likely in the groundwater discharge zones.

In summary, elevated Ca^{2+} deposition and significant sinks of N (denitrification) and S (sorption, including reduction, adsorption and precipitation) explain the issue of why there is little surface water acidification in China. Other processes like SO_4^{2-} adsorption are only temporarily important (until approaching SO_4^{2-}

saturation). Denitrification and SO_4^{2-} reduction may be more permanent sinks of acidity but depend on soil N status and S status. In addition, SO_4^{2-} sorption is most likely reversible, implying that SO_4^{2-} desorption may delay the increase in soil water pH after a decrease in S deposition. Moreover, modeling results by MAGIC indicated that the current regulation of SO_2 emission abatement could not significantly increase soil water pH values, the $(\text{Ca} + \text{Mg} + \text{K})/\text{Al}$ molar ratio, or soil base saturation to the level of 2000 before 2050, and the emission reduction of particulate matter would offset the benefits of SO_2 reduction by greatly decreasing the deposition of base cations, particularly Ca^{2+} (Duan et al., 2013). Continuous droughts in southwestern China in the future might also delay acidification recovery (Duan et al., 2013).

3.3. Eutrophication

Excess nitrogen deposition has not only led to acidification, but also resulted in ecosystem eutrophication in Eastern Asia, shown as changes in N dynamics, plant growth, or biodiversity. Atmospheric N deposition could stimulate enzyme activities and accelerate N transformation and cycling processes (Kim and Kang, 2011). For example, N addition increased rates of net N mineralization and nitrification, regulating organic matter decomposition (Mo et al., 2006, 2007, 2008a; Mochizuki et al., 2012). Examination of six forests in southern China and Japan indicated that in addition to leaching, denitrification losses of NO_3^- significantly increased with increasing N deposition (Fang et al., 2015), which increased soil N_2O emissions (Zhong et al., 2008).

Although N deposition could improve soil N availability and result in an increased photosynthetic capacity and stimulation of plant growth in N-limited ecosystems (Fan et al., 2007; Xia et al., 2009; Bai et al., 2010), excess N input led to restriction to plant growth or even damage to plants due to change in soil N status (Fang et al., 2009; Lu et al., 2009; Xu et al., 2009), nutrient imbalance (Yang et al., 2009), or reduction in net photosynthesis (Mo et al., 2008b; Guo et al., 2014).

Biodiversity could also be significantly affected by N deposition, with the level depending on soil N status, vegetation composition, dose and duration of N addition, and N requirements by different species (Bai et al., 2010). Excessive N deposition normally reduced biodiversity, including forest understory species (Lu et al., 2008, 2010a), grasses and forbs (Bai et al., 2010), and soil fauna (Xu et al., 2006).

4. Critical loads

In Southeast Asia, comprising China, Korea, Japan, The Philippines, Indo-China, Indonesia and the Indian subcontinent, critical loads were first computed and mapped as part of the impact module of the Asian version of the Regional Air pollution INformation and Simulation model (RAINS-Asia) (Hettelingh et al., 1995b). RAINS-Asia is used to assess abatement strategies for S emissions through the application of the critical loads concept (Streets et al., 1999). According to that study, low critical loads (subject to high risk of acidification) are found in southeastern Asia, parts of the Himalayan range and the Tibetan plateau, parts of the boreal forest in northern China, and the rain forest strip in southwestern India, while the dry regions in most of India and northwestern China show relatively high critical loads (Hettelingh et al., 1995b). To improve the spatial resolution, critical loads were also studied in many Asian countries such as Japan (Shindo et al., 1995; Shindo and Fumoto, 1998; Hayashi and Okazaki, 2001), Russia (Bashkin et al., 1995; Semenov et al., 2001), South Korea (Park and Lee, 2001; Park and Shim, 2002; Park and Bashkin, 2001), India (Gautam et al., 2010; Satsangi et al., 1995, 1998), and China (Zhao

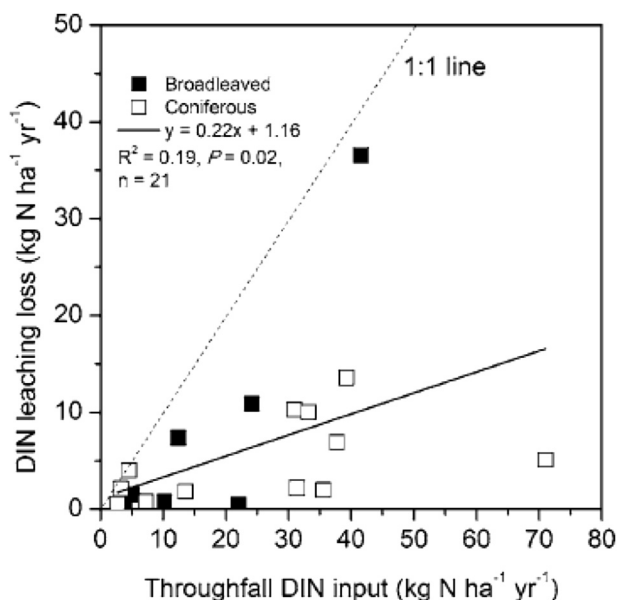


Fig. 10. Throughfall N input versus dissolved inorganic nitrogen (DIN) leaching by soil water in China (Fang et al., 2011).

and Seip, 1991; Duan et al., 2000a).

In most of the above studies, similar methods were applied in Asia as in Europe, with some minor modifications (Duan et al., 2000a; Posch et al., 2015). One of the most important modifications of the widely used steady state mass balance (SSMB) method was the consideration of base cation (BC) deposition (Zhao et al., 2007a, b). As mentioned above, BC deposition, with a considerable fraction of anthropogenic origin, becomes the most important source of ANC instead of weathering rate in China. Thus BC deposition is considered variable instead of constant in the critical load equation, which leads to the extended S-N-BC critical load function (Zhao et al., 2007a). Both the maximum critical load of S ($CL_{max}(S)$) and the maximum critical load of N ($CL_{max}(N)$) would decrease with the reduction of BC deposition (Fig. 11). If alkaline dust emissions are controlled in the future, more efforts will be required to prevent soil acidification and ecosystem damage. This illustrates the potential of future acidification induced by reduced BC deposition, if S and N deposition are not reduced correspondingly.

Based on the extended SSMB method, a map of critical loads for S and N for China was developed under current BC deposition (Zhao et al., 2009). It shows that the S critical loads in the northern and northwestern China were generally higher than $30 \text{ kg ha}^{-1} \text{ yr}^{-1}$, due to high weathering rates and natural deposition of base cations, while the values could be lower than $0.3 \text{ kg ha}^{-1} \text{ yr}^{-1}$ in north-eastern China, with low temperatures and thus low weathering rates, and in southern China, where both low weathering rates (due to low content of weatherable minerals) and high vegetation uptake of base cations occurs. Such results have been applied in policy-making in China, such as the designation of the Acid Rain Control Zones (Hao et al., 2001b) and the total emission control planning. Under the current high BC deposition, the area exceeding the $CL_{max}(S)$ covered about 15.6% of mainland China (Zhao et al., 2011). Unanticipated side effects of the control of primary PM and thus BCs, particularly from the anthropogenic sources, may wholly counteract the benefits to regional acidification of reduced

emissions of acid precursors, including large-scale abatement of SO_2 achieved since 2006 (Fig. 12). This suggests that policy-makers may have little choice but to pursue even more stringent SO_2 and NO_x controls.

5. Summary

Associated with rapid economic development, acid deposition has become a major issue in Asia, especially in East Asia. Generally, surface waters in Asia are not as sensitive to acid deposition in comparison with soil. This is even true in acid forest soils in tropical and subtropical regions, which are characterized by low mineral weathering. East Asia is different from North America and North-west Europe as the acidification potential of atmospheric deposition is less than expected due to high base cation deposition, particularly Ca^{2+} deposition, derived from soil dust and particulate matter from cement production and fossil fuel combustion. Therefore, more attention should be paid to the trend of base cation emission (both natural and anthropogenic) and deposition in Asia. In addition, NO_3^- denitrification and SO_4^{2-} adsorption are processes that play a more prominent role in acid neutralization in soils of East Asia than in Europe and North America.

Nitrogen deposition, especially of NH_4^+ , is of increasing concern in Asia due to nitrification and nitrate leaching in N-saturated ecosystems causing acidification of soils and water. Enhanced NO_3^- leaching has been observed in China and Japan. Although further studies are needed, the acidifying effect of N deposition may be more important than S deposition in well drained tropical/sub-tropical soils due to high SO_4^{2-} adsorption.

As the biggest contributor of S and N emissions, China's emissions have begun to decrease in recent years, following Japan, South Korea and some other countries. This has led to a decrease in S and N deposition, and beginning of recovery from soil acidification in these countries. However, the large stores of adsorbed SO_4^{2-} are expected to be desorbed, a process which delays the recovery of the

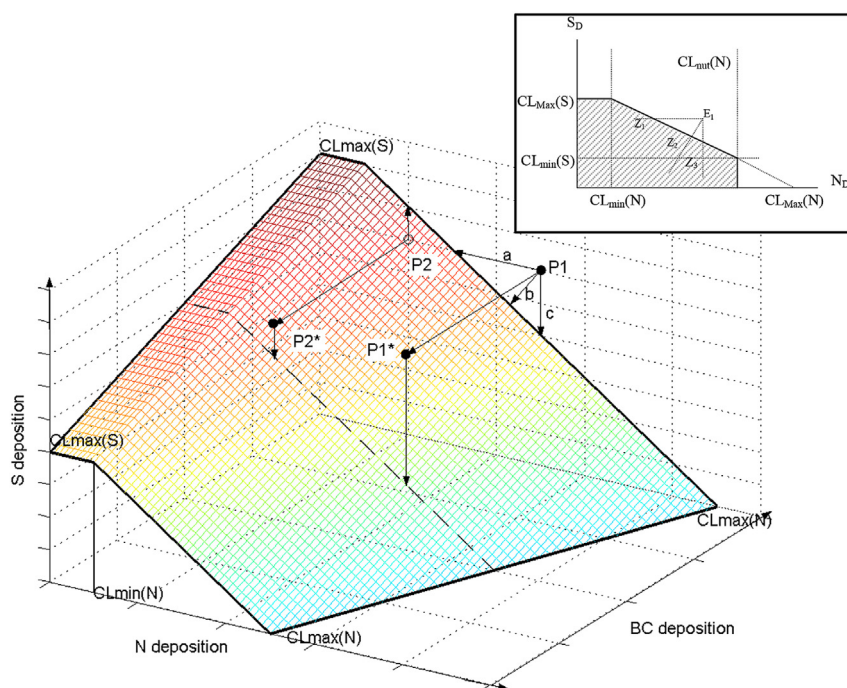


Fig. 11. Extended S-N-BC critical load function (Zhao et al., 2007a). To avoid exceedance, depositions of S, N, and BC should be limited below the shaded surface. More effort to reduce S and/or N deposition should be taken to avoid acidification as BC deposition is reduced, regardless of whether the critical load is currently exceeded (route P1 to P1*) or not (route P2 to P2*). Small diagram shows the traditional S-N critical load function (Posch et al., 1995).

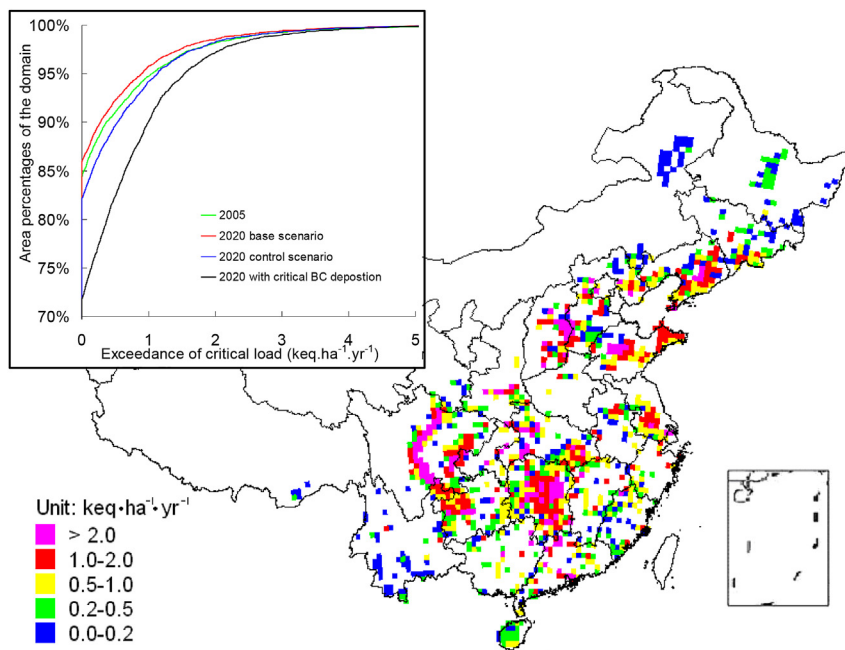


Fig. 12. Critical loads exceedances in 2005 in China (Zhao et al., 2011). Small diagram shows the cumulative distributions of the critical load exceedances in different future scenarios. The area exceeding the $CL_{max}(S)$ covered 15.6%, 13.5%, 17.9%, and 27.5% of mainland China under the current BC deposition in 2005, and under the base scenario (without stricter PM emission abatement), the control scenario (with more realistic PM control), and the critical BC deposition (without all anthropogenic BC sources) in 2020, respectively. Higher curves indicate relatively lower acidification risk at the national level.

soil from acidification. Thus, how quickly soils respond to decreased deposition is uncertain. Risk of regional soil acidification still exists, as can be seen from critical load exceedance in large areas of East Asia. Further studies on the effect of acid deposition in Asia are therefore needed, not only for improving our understanding, but also for supporting future policy-making.

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