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Will PM control undermine China's efforts to reduce soil acidification?

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ABSTRACT

China's strategies to control acidifying pollutants and particulate matter (PM) may be in conflict for soil acidification abatement. Acidifying pollutant emissions are estimated for 2005 and 2020 with anticipated control policies. PM emissions including base cations (BCs) are evaluated with two scenarios, a base case applying existing policy to 2020, and a control case including anticipated tightened measures. Depositions of sulfur (S), nitrogen (N) and BCs are simulated and their acidification risks are evaluated with critical load (CL). In 2005, the area exceeding CL covered 15.6% of mainland China, with total exceedance of 2.2 Mt S. These values decrease in the base scenario 2020, implying partial recovery from acidification. Under more realistic PM control, the respective estimates are 17.9% and 2.4 Mt S, indicating increased acidification risks due to abatement of acid-neutralizing BCs. China's anthropogenic PM abatement will have potentially stronger chemical implications for acidification than developed countries.

1. Introduction

Acidification in China is generally attributed to intensive coal combustion and hence high emissions of acid precursors (Larssen et al., 2006). Parts of China now have the highest observed acidity of precipitation in the world, with continuously worsening conditions in recent years (Larssen et al., 2006; Wang and Xu, 2009; Tang et al., 2010). To reverse growth in SO_2 emissions, China's government has enacted stringent requirements in the power sector: flue gas desulfurization (FGD) is mandated at all newly-built and many pre-existing units and over 50 GW of inefficient small units are being replaced by larger, efficient ones (Zhao et al., 2008). The results are encouraging, with national SO_2 emissions reported officially to decrease by 13% from 2005 to 2009. The benefits of sulfur (S) abatement to reduced soil acidification during 2005–2010, however, have likely been negated by concurrent increased emissions of NO_x and NH_3 (Zhao et al., 2009). Given this rising nitrogen (N) pollution, policies for NO_x control are currently under consideration for implementation after 2010. In the latest emission standards for power plants proposed (but not published) by the Ministry of Environmental Protection, the limits of NO_x concentrations in flue gas are 11%–87% lower than in current standards, depending on unit age and fuel quality.

Other species influencing acidification are base cations (BCs), including Ca^{2+}, Mg^{2+}, K^{+}, and Na^{+}, which mitigate the problem. A series of studies has confirmed the acid-neutralizing effects of BCs at varied sites in Europe (Hedin et al., 1994; Rogora et al., 2004; Watmough et al., 2005), North America (Driscoll et al., 1989; Hedin et al., 1994; Likens et al., 1996, 1998; Gbondo-Tugbawa and Driscoll, 2003; Watmough et al., 2005; Wu and Driscoll, 2010), and China (Larssen et al., 2006; Zhao et al., 2007a, b). Most of them, however, focus on sub-national regions or individual catchments, limiting their value to national policy-making. Some apply hypothetical scenarios for BCs (e.g., a simple percentage reduction of deposition assumed uniformly across a domain) without consideration of spatial distributions and other source-specific factors influencing such changes in actual conditions (Larssen and Carmichael, 2000).

Critically, moreover, the emissions and depositions of BCs of anthropogenic origin have not been well estimated to quantify the effects on acidification, and large uncertainties still remain. A series of studies found that the reduction in deposition of BCs at a U.S. forest catchment contributed considerably to the depletion in the soil Ca^{2+} exchange pool and to the decreases in stream water concentrations of BCs (Driscoll et al., 1989; Likens et al., 1996, 1998; Gbondo-Tugbawa and Driscoll, 2003). Lacking detailed information on sources of BCs to the atmosphere or emission inventory of BCs, the reduction was speculated to be associated with anthropogenic activities such as emission control on power plants and industrial boilers; this was based on the regression relationships between observed depositions of BCs and U.S. national emissions of particulate matter (PM) (Gbondo-Tugbawa and Driscoll, 2003). Regarding

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acidification estimates at national or continental scales, previous studies are either limited to depositions of natural dust (Sanderson et al., 2006; Hicks et al., 2008) or hypothesize constant fractions of total depositions of BCs that are of anthropogenic origin (Larssen and Carmichael, 2000). The true size of anthropogenic dust emissions and hence the anthropogenic contribution to the total deposition is described as largely unknown (Larssen and Carmichael, 2000). To better understand the role of anthropogenic PM in neutralizing soil acidification, more information on the emissions of BCs from anthropogenic sources, the range of atmospheric transport, and the true future trends in depositions of BCs are thus required (Driscoll et al., 1989; Hicks et al., 2008).

While PM emissions of anthropogenic origin have been gradually controlled and BCs from natural sources may dominate deposition in developed countries, China still has much higher emissions of BC-containing PM of anthropogenic origin. The emission of total primary PM for China was estimated at 34 million metric tons (Mt) in 2005 (Zhao et al., 2011), in contrast to 5 Mt for Europe (CEIP, 2010) in the same year. In this respect the underlying atmospheric composition differs from that evaluated in most prior studies, and abatement of anthropogenic PM in countries like China could have much stronger chemical implications for the environment. Experiencing fast industrialization, China's government is attempting to shift its development mode from one dependent on massive fossil energy inputs and high emissions to a more resource-efficient and environment-friendly mode. As the urban air pollutant most damaging to public health, emissions of PM (and hence those of BCs) are expected to decline in the future due to enhanced controls. These PM control measures are being developed without consideration of the poorly recognized role of anthropogenic BCs on soil acidification. If these effects are shown to be significant – that decreasing PM emissions will prevent recovery of soil acidification even as S is aggressively abated and growth in N is reduced – it will indicate that soil acidification cannot be combated efficiently or even effectively independent of other air quality challenges. Instead, a multipollutant perspective must be taken that considers variations of emissions and interactions of all these species simultaneously.

In this study, anthropogenic emissions of SO_2, NO_x, NH_3, and BCs are estimated in 2005 and 2020 incorporating the latest emission factors for China (Zhao et al., 2008; Zhang et al., 2009). After gridding the emissions spatially, atmospheric depositions are simulated with chemistry and transport models and then analyzed using an extended framework of critical load (CL) that combines the acidifying effects of S, N and BCs (Zhao et al., 2007b). Soil acidification risks are indicated by exceedances of CL, providing a basis for considering the problem within a comprehensive, multipollutant control strategy.

2. Control of acidifying pollutants during 2005–2020

National emissions of SO_2, NO_x and NH_3 in 2005 are updated to 31.1, 18.8 and 16.6 Mt respectively based on previous studies (Zhao et al., 2009; Zhang et al., 2009). Anticipated control measures and their effects on emissions of those species through 2020 are discussed below and summarized in Table 1.

The power sector is the primary focus of SO_2 pollution control (Zhao et al., 2008). Due to FGD mandates, we estimate that the penetration rate of FGD in the sector will increase from 14% in 2005 to 80% in 2010. The actual control effects, however, are less than designed because the FGD are not always operated as intended. The average removal efficiency of FGD has recently been estimated to be around 70% by an unpublished government survey. In this study we assume that the average removal efficiency will reach 80% in 2020 due to improved implementation. In addition, we assume that FGD systems will be required at iron & steel plants by 2020. We further estimate that SO_2 emission factors for coal-fired boilers in industrial and residential sectors will decline by 15%, due mainly to implementation of national emission standards. In transportation, we assume a large, 80% reduction of the emission factor, due to an anticipated shift to lower-S fuels.

Increasing N pollution in China is raising attention to the need for NO_x control. Based on the latest proposed standard for power plants, we assume that all power units in "key areas" will be required after 2010 to install denitrogenation technologies, e.g., selective catalyst reduction (SCR). Key areas are those with high densities of emissions: north-central, south-central, and eastern China (see the Supporting information for description of regions). Based on observed control effects of SCR (Zhao et al., 2010), we assume that the average removal efficiency in 2020 will be 40% and 50% for units built before and after 2010, respectively. In transportation, new on-road vehicles have been required since 2007 to meet China's stage III standard (equivalent to Euro III). In this study, we assume that stage IV and V standards will be required for on-road vehicles after 2010 and 2012, respectively, and stage II and III standards for non-road sources in the same years. Based on a projection of fleet composition (Wang et al., 2006), the average emission factors for on-road and non-road sources are estimated to decline 60% and 30%, respectively, in 2020 compared to 2005. For NH_3, since there is little technology available in China to capture emissions from livestock and fertilizer applications, we assume that the emission factors will remain unchanged from 2005 to 2020.

As listed in Table 1, the changes of activity levels by sector during 2005–2020, expressed either in energy consumption or industrial production, are taken from different studies. Specifically, there is currently little projection on livestock and fertilizer amount for NH_3 emission estimate. According to official statistics (NBS, 2009), the

Table 1
Changes of emission factors and activity levels for 2005–2020. All estimates are national averages for sectors. Those without a reference are original to this study. Indicated changes of activity levels concern production of brick and lime, and energy consumption in all other sectors.

	Emission factor					Activity level
	SO_2	NO_x	PM-base	PM-control	NH_3	
Power plant	−72%	−39%	−72%	−75%	–	+110%[a]
Cement	−50%[b]	+14%[b]	0%	−82%[b]	–	+60%[c]
Iron & steel	−80%	0%	−74%	−80%	–	+117%[c]
Other industry	−15%	0%	0%	−20%	–	+40%[d]; including −37% for brick making and −10% for lime production[e]
On-road transportation	−80%	−60%	−85%	−85%	–	+161%[f]
Non-road transportation	−80%	−30%	−56%	−56%	–	+27%[g]
Residential (biofuel)	0%	0%	0%	0%	–	−20%[d]
Residential (coal)	−15%	0%	0%	−20%	–	−5%[g]
Biomass open burning	0%	0%	0%	0%	–	−20%[d]
Livestock	–	–	–	–	0%	+29%
Nitrogenous fertilizer	–	–	–	–	0%	+16%
Compound fertilizer	–	–	–	–	0%	+179%

[a] From Zhao et al. (2008).
[b] From Lei et al. (2011).
[c] From CAEP and SIC (2009).
[d] From IEA (2007).
[e] From Zhou et al. (2003).
[f] From Wang et al. (2006).
[g] From Zhang et al. (2008).

annual increasing rates for livestock and nitrogenous fertilizer after 2005 are small, around 2% and 1% respectively, while the use of compound fertilizer, which has much lower emission factors of NH_3 than nitrogenous fertilizer, increased much faster at 7% per year. In this study, such increasing rates are assumed unchanged till 2020. By combining activity levels and emission factors, the emissions of SO_2, NO_x, and NH_3 in 2020 are derived. With emission spatially gridded, deposition is simulated using the Models-3/Community Multiscale Air Quality (CMAQ) system. The model settings are described in a previous study (Zhao et al., 2009) and the simulated results compared well to both ground and satellite observations (Zhao et al., 2009; Xing et al., submitted for publication).

National anthropogenic emissions of SO_2, NO_x, and NH_3 in 2020 are estimated at 23.0, 23.4, and 22.0 Mt, respectively. The projected SO_2 emission is generally lower than other recent estimates (Ohara et al., 2007; Klimont et al., 2009) except the "Policy Success Case" of Ohara et al. (2007), due mainly to the improved removal efficiency by FGD systems assumed in this study. In particular, emissions from the power sector are projected to decline by more than 40% from 2005 to 2020, and its corresponding share of total emissions from 55% to 42%. In contrast, however, NO_x emission is estimated to keep rising and is generally higher than other studies, except the "Policy Failure Case" of Ohara et al. (2007). This is attributed largely to a conservative assumption of removal efficiency by SCR systems and the rising emission factors in cement production.

Fig. 1 shows S and N depositions in 2020 simulated by CMAQ (see also our previous study (Zhao et al., 2009) for simulated depositions in 2005). From 2005 to 2020, 26 out of 31 provinces in China are estimated to experience decreased S deposition. In particular the S depositions in north-central and eastern China are estimated to decline by 28% and 19%, respectively. This is larger than the national decline of 17%, implying successful achievement of S pollution control in those target areas. In contrast to S, however, N deposition will rise by at least 10% in all provinces except Tibet. Even with wide application of denitrogenation technologies in the power sector, N deposition in some provinces of south-central and eastern China will grow by more than 34%, the national average increase.

3. Emission and deposition of base cations: 2005 and 2020 scenarios

As indicated by eq (1), anthropogenic emissions of BCs are generally calculated as the product of PM emissions and BC mass fractions of PM:

$$E_{i,j,k} = E_{PM,j,k} \times w_{i,j,k}\% \quad (1)$$

where i, j, and k stand for the species of base cations, regions, and sectors, respectively; E is the emission level; and $w\%$ is the percentage of BCs to total PM emission by mass.

Seven dominant sector sources are included: power, cement, iron & steel, lime production, brick making, other industrial sources, and residential coal combustion. PM emissions from coal-fired power plants are evaluated using a unit-based methodology (Zhao et al., 2008). For cement (Lei et al., 2011) and iron & steel sectors, emission factor database was developed based on investigation of major plants in the country, capturing the emission characteristics by technology. The location, capacity, and technology for each plant in these three sectors are available, and thus they are analyzed as large point sources. PM emissions of the other four sectors are calculated at the provincial level as the product of activity levels and emission factors. The activity levels are chiefly obtained from national industrial and energy statistics, while emission factors are either from domestic measurements (SEPA, 1996) or otherwise from international research (USEPA, 2010a). Mass fractions of Ca^{2+} and Mg^{2+} in total PM emissions by region and sector are obtained from a national investigation (Zhu et al., 2004). Due to their tiny shares of both PM emissions and precipitation, K^+ and Na^+ are not included.

Regarding PM emissions in 2020, two scenarios are set to evaluate the effects of emission control. As summarized in Table 1, PM emission factors in the base scenario are assumed to remain at the current national average levels for most sectors that generate BCs. However, the ancillary effects of SO_2 emission control measures, which are not specifically designed for PM, are also included in the scenario. There is a 72% reduction of the emission factor for the power sector, due mainly to the ongoing replacement of small units by large ones with electrostatic precipitators and wet-FGD systems (Zhao et al., 2008). Similarly, the PM emission factor for iron & steel production will be reduced by 74% due to the penetration of wet-FGD systems. In the control scenario, the emission control measures currently mandated mainly in the power sector are broadened to other sectors. PM emission factors are assumed to decline further, due to technology innovation in the power and cement sectors and to tightened emission standards in other industrial and residential sectors. Most notably, the PM emission factor of cement production declines by 82% compared to 2005, due mainly to anticipated widespread deployment of fabric filter systems (Lei et al., 2011). Noting the recent successes of emission control policy implementation at power plants in China, the control scenario is considered more realistic.

Since CMAQ does not include the module of simulating depositions of BCs, a multi-layer Eulerian model for long-range transport of BCs is used to simulate their depositions (Duan et al., 2007).

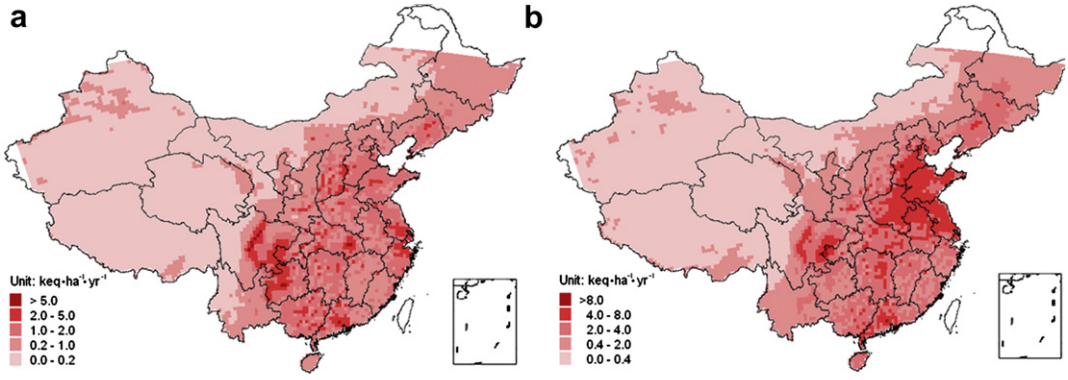

Fig. 1. Simulated deposition of (a) S and (b) N in 2020 using the emission inventory with control policies. The spatial resolution is 36 × 36 km. Due to a limitation of the CMAQ modeling domain, there are no results for small regions in the far northeast and northwest.

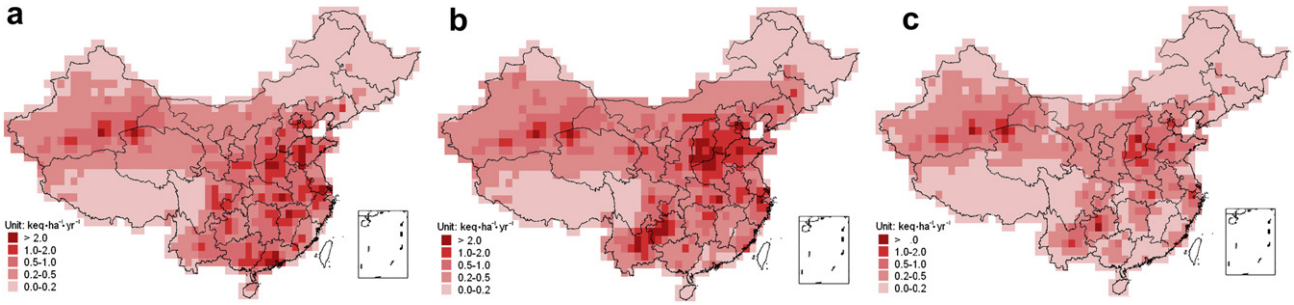

Fig. 2. Simulated deposition of BCs (Ca^{2+} + Mg^{2+}) for (a) 2005; (b) 2020 base scenario; and (c) 2020 control scenario. The spatial resolution is 1°×1°.

Provincial emissions of BCs are distributed on a 1°×1° grid system covering the entire country. Besides anthropogenic emissions, estimates of natural dust entrainment in northern China are taken from a previous study and assumed to be unchanged (Zhu et al., 2004) (see Supporting information for the model description and spatial allocation of emissions).

The total anthropogenic Ca^{2+} emissions in 2005 and in the 2020 base and control scenarios are estimated at 5970, 6882, and 3035 kilo metric tons (kt), respectively (see Supporting information for emissions by provinces). The estimates are much higher than those for Europe of 750–800 kt in 1990 (Lee and Pacyna, 1999), highlighting the limits of applying Europe's experience to countries like China, where BCs may have a much larger role in atmospheric chemistry. Cement and lime production are the leading sources of Ca^{2+}, responsible for 88% of total emissions in 2005. With rising production, emission from cement plants will increase from 2604 kt in 2005 to 3991 kt in the 2020 base scenario, while it will decrease to 718 kt in the control scenario due to the aforementioned fabric filters. For lime, Ca^{2+} emissions will decrease from 2621 kt in 2005 to 2269 kt in the 2020 base scenario due to a decline in production, and to 1815 kt in the control scenario due to stronger regulations. Anthropogenic Mg^{2+} emissions in 2005 and the 2020 base and control scenarios are estimated at 236, 331, and 203 kt, respectively, much less than Ca^{2+}.

The simulated depositions of BCs are found to differ somewhat from observations (see the Supporting information for performance and uncertainty of the model). Without better alternatives, however, current simulations capture the spatial patterns of deposition and should be acceptable for policy purposes in China. Fig. 2 shows simulated deposition of BCs over the country. The total national deposition will increase by 16% from 2005 to the base scenario 2020, but decrease by 32% in the control scenario. A consistently large

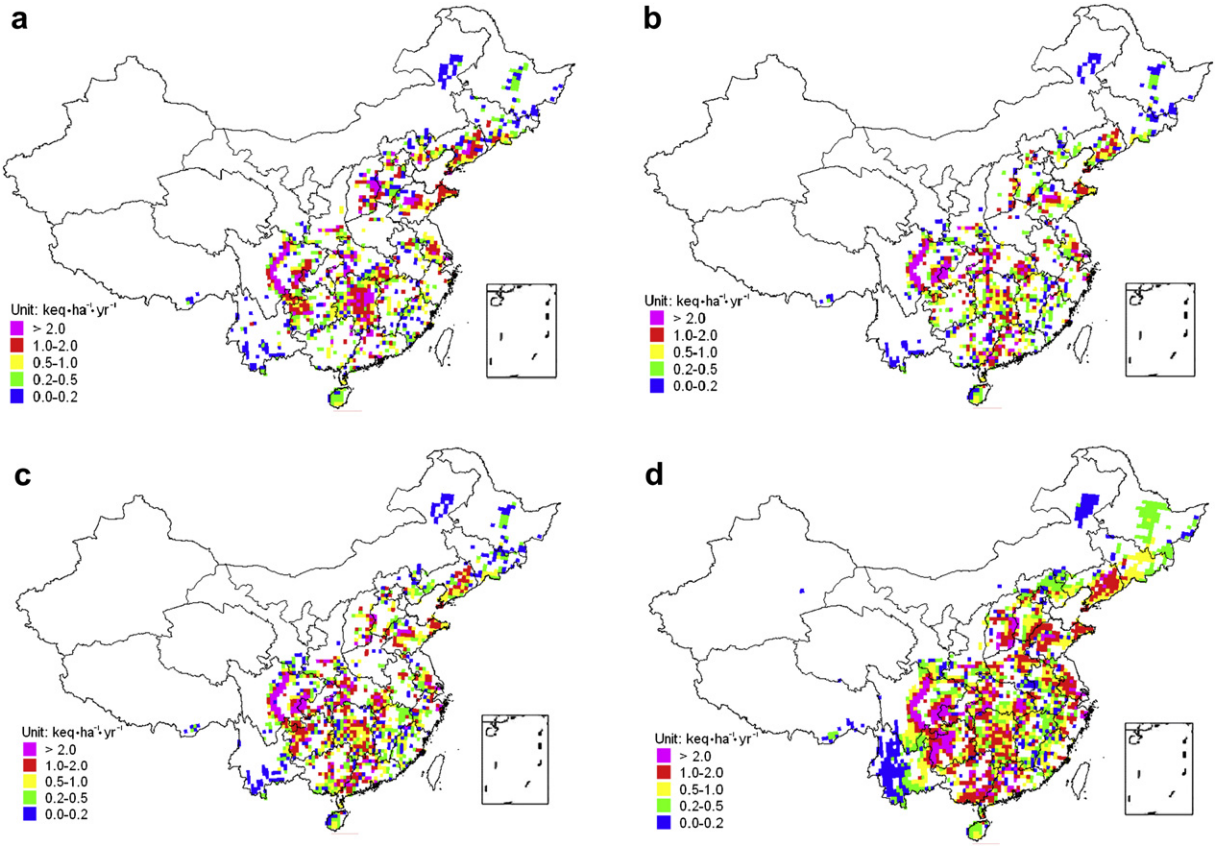

Fig. 3. Exceedances of critical load for acidification: (a) 2005; (b) 2020 base scenario; (c) 2020 control scenario; (d) 2020 with natural BC_D only. The spatial resolution is 36 × 36 km.

deposition is found in northwestern China due to the contribution of natural dust. South-central and eastern China are estimated to have the most significant deposition reductions because of emission controls of anthropogenic sources. Deposition of BCs will decrease by 66% and 56% from 2005 to the 2020 control scenario for the two areas respectively. Deposition from natural dust accounts only for 22%, 19%, and 34% of the totals for the 2005 and the 2020 base and control scenarios, respectively, illustrating the dominant contributions of anthropogenic sources even as control measures are tightened in the future.

4. Exceedance of critical load using a multipollutant assessment

The acidifying critical load (CL) below which harmful ecological effects do not occur designates a threshold of depositions of S, N, and BCs (S_D, N_D and BC_D, respectively). Based on a steady-status mass balance (SSMB), CL can be expressed with eqs (2)–(4), which imply that the threshold of S_D declines with a decrease of BC_D as well as with the increase of N_D, if the latter is larger than $CL_{\min}(N)$ (see Supporting information for details).

$$CL(S)_{\text{limit}} = BC_W - BC_U - ANC_{L,\text{crit}} \tag{2}$$

$$CL_{\min}(N) = N_I + N_U \tag{3}$$

$$S_D \le CL(S) = \begin{cases} BC_D + CL(S)_{\text{limit}} & (N_D \le CL_{\min}(N)) \\ BC_D + CL(S)_{\text{limit}} - (1 - f_{DE}) \times (N_D - CL_{\min}(N)) & (N_D > CL_{\min}(N)) \end{cases} \tag{4}$$

where BC_W is the weathering rate of base cations from soil minerals; BC_U and N_U are the vegetation uptake of base cations and nitrogen respectively; N_I is the net immobilization rate of nitrogen in the soil; f_{DE} is the nitrate lost by denitrification; $ANC_{L,\text{crit}}$ is the critical leaching of alkalinity. Those data are taken from previous studies for China (Duan et al., 2000, 2002, 2004).

By comparing the simulated depositions of S, N and BCs with CL, the distributions of CL exceedances in 2005 and 2020 are estimated and shown in Fig. 3. Besides the base and control scenarios, the CL exceedance in 2020 with deposition of natural BCs only is also illustrated.

In 2005, the areas exceeding CL covered approximately 15.6% of China's mainland territory, mainly in south-central, southwestern, and part of northeastern China (Fig. 3a). The total exceedance for the country is estimated at 2.2 Mt S, equaling 15% of national SO_2 emissions. In the 2020 base scenario, in which PM emissions are not well controlled and thus high deposition of BCs is projected, the share of area exceeding CL will decline to 14.1% and the total exceedance to 1.8 Mt S, despite an increase of CL exceedance in the southernmost part of south-central China (Fig. 3b). The reduction of the area exceeding CL and total exceedance level indicate recovery from acidification due to the SO_2 and NO_x controls.

However, in the more realistic 2020 control scenario, in which PM abatement policies reduce deposition of BCs across the country, the share of area exceeding CL will rise to 17.9% and the total exceedance to 2.4 Mt (Fig. 3c). Both values are even larger than those of 2005, implying higher risks of acidification, particularly in south-central and part of eastern China. If only the deposition of natural but not anthropogenic BCs is included for 2020, the share of area exceeding CL and total CL exceedance would reach 28% and 3.9 Mt S. These are 56% and 62% larger, respectively, than those in the control case (Fig. 3d). No significant exacerbation of acidification is found in northwestern China where natural dust sources are mainly located. That result confirms importance of anthropogenic BCs in limiting soil acidification, particularly in south-central and eastern China where the risks are highest.

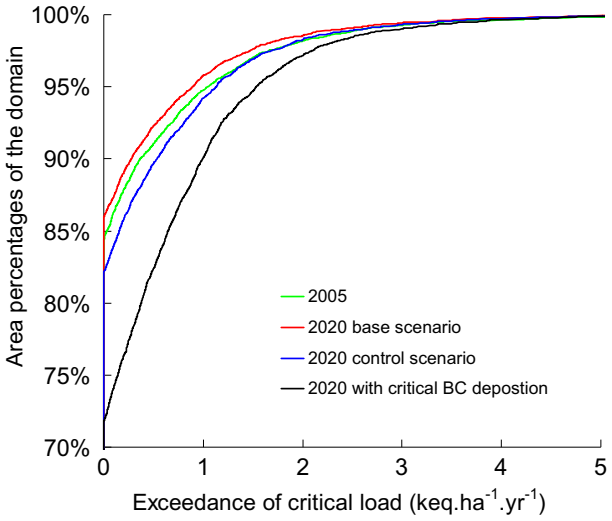

Fig. 4. Cumulative distributions of critical load exceedances in areas. Higher curves indicate relatively lower acidification risks at the national level. In 2005, for example, roughly 85% of the country's territory did not exceed the critical load; the remaining 15% suffers varied levels of soil acidification, represented by CL exceedances that increase from left to right until 100% of the territory is accounted for.

Cumulative distributions of CL exceedances in areas are illustrated in Fig. 4. It clearly shows that soil acidification at national level from 2005 to the 2020 base scenario will be reduced (from the green curve to the red one), due mainly to decreasing S deposition and increasing BCs. That progress will likely be delayed, however, or even reversed as PM emissions are abated by the policies assumed in the control scenario (from green to blue). Under projected S and N emissions in this study, the extreme case eliminating all anthropogenic PM is indicated by the black curve, with highest risks.

5. Discussion and conclusion

The simulated results are consistent with observations to the extent they are available in China. Observations of precipitation and vegetation throughfall have been conducted in different areas to understand the actual acidification conditions of the country. Both SO_4^{2-} and Ca^{2+} depositions as well as air concentrations in southwestern, south-central, and eastern China are found to be significantly higher than those in most other parts of the world (Larssen et al., 2006; Aas et al., 2007; Wang and Xu, 2009), reflecting large emissions of anthropogenic SO_2 and BCs. In accord with the simulation, long-term monitoring found partial correlation of an increasing trend of precipitation acidity with a decrease in concentrations of airborne PM at many sites across China, and that correlation cannot be explained by changes in natural sources (Tang et al., 2010). This result is strong evidence that the simulated process by which reductions in anthropogenic dust aggravate acidification is already taking place.

The present analysis has powerful implications for policy-making: China's current program of emission controls, taken as a whole, is unlikely to achieve its longstanding goal of reduced acidification. Unanticipated side effects of the control of primary PM and thus BCs, particularly from the cement and lime industries, may wholly counteract the benefits to regional acidification of reduced emissions of acid precursors, including large-scale abatement of SO_2

achieved since 2006. Moreover, such implications are probably even stronger than those indicated by the experiences in developed countries. The U.S. national emissions of SO_2 and PM_{10} (as a surrogate for BCs) were reported to be reduced by 36% and 24%, respectively, from 1990 to 2005, the 15 years following enactment of the 1990 amendments to the Clean Air Act (USEPA, 2010b). Based on long-term observation of a forest catchment, however, a previous study indicated that the amendments would not be adequate to protect surface waters and forest soils of the northeastern U.S. against further anthropogenic acidification (Likens et al., 1996). For China, the control scenario of the current study estimates that the emissions of SO_2 and BCs would be reduced by 22% and 49%, respectively, from 2005 to 2020. The much higher percentage decline of BCs and lower decline of SO_2 thus suggest that acidification abatement may be even less likely in China under current anticipated control policies than the U.S. experienced in 1990–2005. Despite this, ongoing PM control efforts must be continued in China because of the critical benefits of reduced aerosol pollution and avoided associated damages to public health. This suggests that policy-makers may have little choice but to pursue even more stringent SO_2 and NO_x controls.

The implications are not limited to China. Since China dominates the anthropogenic emissions (Zhang et al., 2009) and contributes significantly to long-range transport of acidifying pollutants in East Asia (Lin et al., 2008), the effectiveness of its policy response will impact other Asian countries. As important, other swiftly industrializing nations with acid rain pollution and high PM concentrations will face similar trade-offs in pollution control. A study analyzing the chemical composition of precipitation and throughfall in southern Brazil found a stronger neutralization of acid deposition by alkaline cations in urban and industrial areas versus rural sites, highlighting the significance of industrial PM and the need to research the effects of gradual control of anthropogenic sources (Casartelli et al., 2008). Even in developed countries like the U.S., studies have advised greater understanding of the effects of deposition of BCs on ecosystem acidification (Kelly et al., 2002). A recent U.S. study suggested that greater emission controls might be needed to protect sensitive forest sites from acidification under reduced depositions of BCs in the future (Wu and Driscoll, 2010). These results concur with the current study that prior understanding that natural sources dominate alkaline dusts may underestimate the role of BCs in acidification. Quantitative analyses at larger scales, regionally or nationally, are also needed to improve policy strategies to control acidification.

Currently the concept of CL and the SSMB approach are widely applied at the national to continental scales for estimating ecosystem acidification, determining the required emission reduction of acidifying precursors, and modeling the recovery process of threatened ecosystems in Europe (Moncoulon et al., 2007; Hettelingh et al., 2007, 2008). While accepted as a basis for emission control and acid rain mitigation in China (Hao et al., 2000), the limitations of the methodology should also be recognized. First, the concept of CL can be applied to natural ecosystems such as forests and grasslands but not to croplands. Compared to fertilizer application, acid deposition is a minor contributor to the declining soil pH over the past 30 years in Chinese agricultural lands (Guo et al., 2010). The policy implications drawn from CL exceedances should thus concern areas of appropriate ecosystems only. Secondly, although statistical analyses found significant relationships between CL exceedance and the threat by loss of species in European forests (Augustin et al., 2005; Hettelingh et al., 2008), such analyses have not been well conducted for China. Severe and increasing defoliation and forest mortality have been observed in some catchments with high acid deposition in southern China, implying rising ecosystem acidification. The association between forest health and acidification, however, needs further investigation because insect attack is also believed to play an important role (Larssen et al., 2006). Moreover, the assumption of SSMB concerns marginal ecosystem S retention, which could be important on buffering acidification in subtropical and tropical climates. Based on limited catchment monitoring in southern China (Larssen et al., 2006; Vogt et al., 2006), the S retention was small by comparing the S contents between the deepest soil lysimeter and throughfall (Hicks et al., 2008). Therefore the uncertainties from excluding the effects of S retention are considered only modest in this study. When evaluating the acidification of surface water, the effects of S retention would be more significant (Hicks et al., 2008). Finally, application of the SSMB method cannot inform understanding of the temporal development of acidification and recovery processes. How fast the impacts of CL exceedance would occur is difficult to estimate. One study has initiated investigation of the temporal dimensions of acidification in Asia (Hicks et al., 2008), but there are currently very few empirical observations to support such analyses and results will be highly uncertain. Long-term observations on the effects of ecosystem acidification and recovery are thus required for better understanding of the true scale of acid rain pollution as well as for validation of simulations.

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Appendix. Supporting information

Emission inventories of BCs, description, performance, and uncertainty of the model for simulating deposition of BCs, and the methods of calculating critical load are included.

Supporting information associated with this article can be found, in the online version, at doi:10.1016/j.envpol.2011.05.018.

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