

Simultaneous Assessment of Deposition Effects of Base Cations, Sulfur, and Nitrogen Using an Extended Critical Load Function for Acidification

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Base cations (BC) play an important role to prevent soil acidification. In certain acid sensitive areas, such as China, BC deposition is high and a considerable fraction is of anthropogenic origin. BC deposition might decrease in the future with the implementation of air pollution control measures. The effect of changes in BC deposition, however, has seldom been considered in critical load calculations based on the steady-state mass balance (SSMB) method. In order to better quantify the importance of the BC deposition for acid deposition mitigation policy, an extension of the SSMB method for critical load calculation for soil acidification is presented. The BC deposition is taken into account as a variable along with sulfur (S) and nitrogen (N) deposition, creating an S–N–BC critical load function. As a case study, critical loads of S and N for the Tie Shan Ping catchment in Chongqing in southwest China under variable BC deposition were calculated. Results indicate that abatement of BC deposition has significant impact on the critical loads of S and N. A 75% reduction in BC of assumed anthropogenic origin decreases the critical loads of acids by 58%. The current deposition does not exceed the critical loads, but if BC deposition from anthropogenic sources was controlled, then the exceedance would be considerable. Uncertainty analysis show that the size of the BC deposition of natural origin is the single parameter contributing the most to the steady-state S and N critical load. The extended critical load function can be used by policy makers to set more reasonable acidity control strategies in the future. The method also highlights for policymakers the “competition” between emission control of particulate matter driven by human health targets and potential increase of net acid load from such measures.

1. Introduction

Acid deposition has, for several decades, been recognized as a major environmental problem in Europe and North

America. In other parts of the world, such as China, acid deposition was recognized somewhat later, and it is considered among the most pressing environmental issues (1). Decreasing trends in acid deposition and related impacts have been clearly observed in Europe and North America (2, 3). On the contrary, in south and southwest China, the acid deposition problem is still increasing with rapidly growing energy demand and coal consumption. Substantial reductions in sulfur (S) emissions seem to be unlikely in the near future.

Sulfate is the dominant anion in precipitation in China (4). However, rapidly increasing emission of nitrous oxides caused by rapid increase in energy use and the number of motor vehicles, together with large emission of ammonia from the agriculture sector, give rise to considerable nitrogen (N) deposition in many areas (5). Hence N deposition is of increasing concern as an additional source for acidification.

Another important feature of regional air pollution in China is the high concentration of base cations (BC, i.e., Ca^{2+} , Mg^{2+} , Na^+ , and K^+) in the atmosphere (6–7). It is well-known that BC deposition may play an important role to prevent acidification. In northern China, the concentration of SO_2 and SO_4^{2-} can be extremely high (8), but due to a high concentration of BC the pH of precipitation is approximately neutral and the soil is not obviously acidified (9). In southern China, the influence of the desert areas in the north and northeast is less obvious, but the BC deposition is commonly much higher than that in Europe and North America (10, 11). Both anthropogenic and natural sources contribute to BC deposition. The industrial emission of calcium (Ca) in China has been estimated at 4.0 Mt in 2000, while the natural sources from wind-blown dust from northern China total 2.3 Mt (11). Comparatively, the overall industrial Ca emission in Europe is much less, approximately $0.8 \text{ Mt}\cdot\text{yr}^{-1}$ (12). A considerable fraction of BC in China is of anthropogenic origin; coal combustion, cement factories, other industry, intensive construction activities, road transport, and land use changes can all be significant sources. However, the size of the contribution to the deposition from the various sources is largely unknown.

As an effect-based approach, critical load of acidity has been defined as the highest deposition of acidifying compounds that will not cause chemical changes leading to long-term harmful effects on ecosystem structure and function (13, 14). Thus, critical load provides a threshold for allowable load of acidity, above which ecosystem damage may occur, including toxic metal (e.g., aluminum) mobilization, forest deterioration, or surface water acidification. In Europe, critical load estimates have been used as the basis for development of international agreements on emission reductions (15, 16). A steady-state mass balance (SSMB) model has been established for calculation of the critical load of S and N deposition for terrestrial ecosystems (17, 18). In the SSMB model, a critical deposition load of a combination of S and N is based on the ability of the ecosystem to produce input bases from BC weathering and deposition. The estimated background (natural) BC deposition is used in the method (18). However, very little data exist that can be used to calculate the natural fraction of BC deposition. Therefore, current BC deposition has commonly been used instead (19).

The European experience could be used by Chinese policymakers to mitigate emissions leading to acidification in a cost-effective way. The critical load concept has also been applied in China, and was taken into account when estab-

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lishing the acid rain control zone (8). Some studies have been carried out to calculate the critical loads of S, N, and acidity for soils at both the local and regional scales in China by different methods such as semiquantitative, SSMB, and dynamic modeling (20–22). A critical load map of China has been derived using the SSMB model (22). This map, however, does not take into account possible changes in BC deposition. To reduce the potential negative health effects from particulate matter in the urban atmosphere, strong measures will be taken to reduce the emissions of particulate matter and, thereby, also base cations in the future. Part of the reduction will occur simultaneously with SO₂ emission reductions and part will be independent of the acid gas emissions. Emissions from combustion sources can be reduced using flue gas particle scrubbers, emissions from roads can be reduced by paving the roads and emissions from construction activities reduced by changing construction practices. Also emissions from natural sources may decrease, for instance following afforestation of dry areas. Chinese total emissions of smoke and dust (which contain a proportion of base cations) in 2004 had decreased by 6 and 17%, respectively compared to that in 2000 (9). Moreover, BC emissions from area sources, such as small cement industrial workshops, have not yet been efficiently controlled, but will be reduced along with regulation implementation after control of large point sources. Over the same time period, the SO₂ emissions increased by 13%, due to increased energy production. Therefore, acidification in China might become more serious in the future, and an overestimate of the critical loads might arise when not taking the changes in BC deposition into account. This situation may also lead to a conflict between air pollution mitigation measures driven by health effects and regional scale ecosystems effects.

To avoid the limitation of the critical load calculation method when the BC deposition is high, an addition to the SSMB model is suggested, taking BC deposition into account together with S and N. This will allow variation of BC deposition as a third control component, and BC can be considered in evaluations together with the acids. As a demonstration, the critical load for soil acidification from S and N under variable BC deposition levels was calculated based on the data monitored in Tie Shan Ping (TSP) area near Chongqing in southwest China, using the extended S–N–BC critical load function. An uncertainty analysis was also carried out to estimate the relative importance of BC deposition to soil acidification.

2. Materials and Methods

Critical loads of S and N for terrestrial ecosystems can be calculated through the SSMB method (18), as shown in eqs 1–4 (see Supporting Information, Section 1 for details):

$$CL(S) + (1 - f_{DE})CL(N) = BC_D + BC_W - BC_U + (1 - f_{DE})(N_I + N_U) - ANC_{L,crit} \quad (1)$$

$$CL_{max}(S) = BC_D + BC_W - BC_U - ANC_{L,crit} \quad (2)$$

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

$$CL_{max}(N) = N_I + N_U + \frac{CL_{max}(S)}{1 - f_{DE}} \quad (4)$$

Calculated by this method, the critical load function can be plotted as a line with generally negative slope on an x–y plane defined by N and S deposition. Thus the SSMB model defines the maximum S and N deposition acceptable for a certain ecosystem with a given ecosystem protection criterion (see Figure 1, Supporting Information).

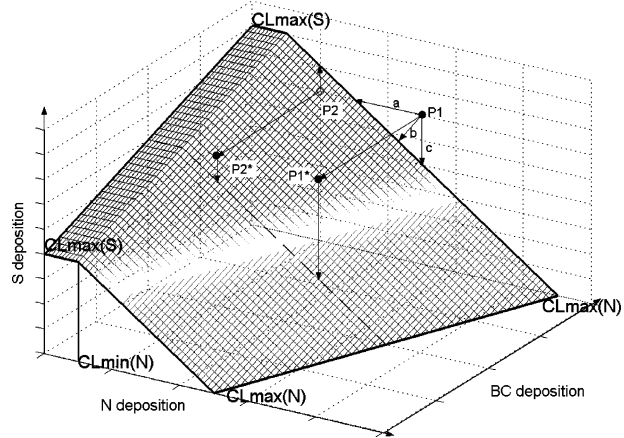


FIGURE 1. Extended S–N–BC critical load function. To avoid exceedance, depositions of S, N, and BC should be limited below the shaded surface. Critical loads of S and N decrease if BC deposition is decreased while $CL_{min}(N)$ remains constant. More effort to reduce S and/or N deposition should be taken 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*).

When defining critical load via eq 1, it is implicitly assumed that all the terms on the right-hand side, including BC deposition, are quantified as the long-term steady-state rates. However, it is difficult to define a long-term steady-state BC deposition that is only from natural sources, especially in areas where current BC deposition is high and a considerable fraction is of anthropogenic origin. In a more general view, if the BC deposition is considered variable instead of constant in the critical load calculation, eq 1 can be rewritten:

$$CL(S) + (1 - f_{DE})CL(N) - BC_D = BC_W - BC_U + (1 - f_{DE})(N_I + N_U) - ANC_{L,crit} \quad (5)$$

Therefore the critical load of S and N can be considered as approximately linear functions of the BC deposition. The extended sulfur (S)-nitrogen (N)-base cation (BC) critical load function is shown in Figure 1, taking changes in deposition of S, N, and BC into account. It is clearly indicated in the figure that both $CL_{max}(S)$ and $CL_{max}(N)$ would decrease with the reduction of BC deposition. However, $CL_{min}(N)$, which is associated only with N immobilization and vegetation uptake will remain constant regardless of BC deposition variation.

In the extended critical load function, each single point with coordinates of (N_D , BC_D , S_D) represents a deposition status. If current S and N depositions lie below the shaded surface, the deposition is acceptable, otherwise, the critical load is exceeded and ecosystem damage is expected at some time in the future. Therefore, measures to reduce S and/or N deposition are needed to prevent soil acidification. Taking point P1 in Figure 1 as an example, there are three kinds of measures to reduce acid deposition to meet the current requirement of critical load: route a (reducing N only), route b (reducing both N and S), and route c (reducing S only). The choice ultimately depends on a multidisciplinary consideration of environment, economy, and technology. Moreover, if BC deposition declines in the future, more efforts will be required to prevent soil acidification and ecosystem damage, because the critical load of S and N will be lower. For instance, when BC deposition level is reduced from P1 to P1*, more S and/or N deposition should be reduced. If current acid deposition does not exceed the critical load (e.g., point P2), attention should still be paid to the BC deposition: The critical load at current BC deposition level is not exceeded (P2), but

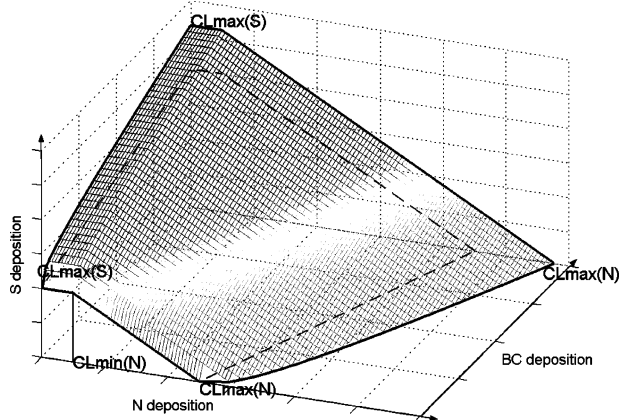


FIGURE 2. Nonlinear S–N–BC critical load function when $(\text{Bc}/\text{Al})_{\text{crit}}$ is applied as the chemical criterion. The broken lines indicate the linear surface as shown in Figure 1.

it may become exceeded if alkaline dust emissions are controlled and BC deposition declines ($P2^*$). This illustrates the potential of future acidification induced by reduced BC deposition, if S and N deposition are not reduced correspondingly.

There are alternative chemical criteria for the SSMB calculation of critical alkalinity leaching ($\text{ANC}_{L,\text{crit}}$), and the choice of the chemical criterion influences the critical load calculation (18). The critical molar ratio of base cations to Al, $(\text{Bc}/\text{Al})_{\text{crit}}$, at which fine root damage might occur in the soil, is commonly used as the chemical criterion for forest (19), although the biological relevance of this ratio is scientifically disputed (23). $\text{ANC}_{L,\text{crit}}$ can be expressed by eq 6 (see Section 2, Supporting Information for the details):

$$- \text{ANC}_{L,\text{crit}} = 1.5 \frac{\text{BC}_D + \text{BC}_W - \text{BC}_U}{(\text{Bc}/\text{Al})_{\text{crit}}} + Q^{1-\frac{1}{\alpha}} \left[1.5 \frac{\text{BC}_D + \text{BC}_W - \text{BC}_U}{(\text{Bc}/\text{Al})_{\text{crit}} \cdot K^*} \right]^{1/\alpha} \quad (6)$$

In the extended function above (Figure 1), $\text{ANC}_{L,\text{crit}}$ was assumed independent of the BC deposition. However, if $(\text{Bc}/\text{Al})_{\text{crit}}$ is used as the chemical criterion, the $\text{ANC}_{L,\text{crit}}$ depends on the BC deposition (see eq 6) and the relation between BC deposition and S/N critical load is not simply linear any more. The critical load function can be redrawn also taking the impacts of the BC deposition on the chemical criteria into account, as a curvilinear surface instead of a linear one (Figure 2).

3. Catchment Application

To demonstrate the extended critical load function it was applied to a typical acidic catchment, Tie Shan Ping (TSP), located on a sandstone ridge 25 km northeast of Chongqing city, at 450 to 500 m asl in the southeast of the Sichuan basin, China (1, 4, 10).

The input variables and parameters for S and N critical load calculation, including deposition, weathering rate, and vegetation uptake of base cations (BC_D , BC_W , and BC_U), immobilization rate and vegetation uptake of N (N_I and N_U), water flux through root zone of soil (Q), $(\text{Bc}/\text{Al})_{\text{crit}}$, and pH-pAl equilibrium coefficients (α and $\log K^*$), were obtained either directly from field monitoring or indirectly by modeling and calculations based on monitored data. The uncertainties for input data were determined based on expert judgment, literature associated with the catchment, or ranges suggested by previous studies.

BC_D was estimated as $8.47 \text{ keq}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$, based on throughfall deposition data. BC_D was assumed to be normally

TABLE 1. Input Data for Calculating Critical Load of Tie Shan Ping

input parameter variable	unit	distribution	value	SE/range
BC_D	$\text{keq}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$	normal	8.47	(0.85)
fraction ^a		uniform	0.25	[0.25–0.75]
BC_W	$\text{keq}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$	normal	0.60	(0.12)
BC_U	$\text{keq}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$	triangle	0.25	[0.25–1.10]
N_U	$\text{keq}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$	triangle	0.21	[0.21–1.17]
N_I	$\text{keq}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$	triangle	0.17	[0.05–0.40]
f_{DE}		triangle	0.8	[0.7–0.9]
critical Bc/Al		uniform	2.0	[0.5–2.0]
α		normal	1.59	(0.08)
$\log K^*$		normal	3.11	(0.32)
Q	$\text{m}^3\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$	normal	5220	(1044)

^a The assumed ratio of natural BC deposition to the total BC deposition. Values in parentheses and square brackets indicate SE and range, respectively.

distributed with a standard error (SE) of 10% of the mean. The fraction of BC_D being of natural origin is highly uncertain and was assumed uniformly distributed from 25 to 75% of BC_D .

BC_W at TSP was calculated as $0.60 \text{ keq}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ using the PROFILE model (24). It was assumed to be normally distributed with SE of 20%, according to previous uncertainty analysis with the PROFILE model (25).

The N_U and BC_U of masson pine (main vegetation type at TSP) was 0.21 and $0.25 \text{ keq}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ with a range of 0.21–1.17 and 0.25–1.10 according to Duan et al. (26). Flux balances at TSP indicate that there is a considerable sink of BC in the soils (approximately $1\text{--}5 \text{ keq}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$), that should be taken into account in the calculation of the critical load. The N_I was estimated from the total amount of soil N divided by the period of soil formation (27). Present N in the rootzone at TSP was approximately $1700 \text{ keq}\cdot\text{ha}^{-1}$ (10), and N_I was set to $0.17 \text{ keq}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$. A range between 0.05 and $0.40 \text{ keq}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ was supplied by Hao et al. (28).

The f_{DE} for the yellow earth at TSP was set to 0.8, a bit higher than the value suggested for clay soils in European applications, but close to that for forest soils in north China (29). The range for f_{DE} was assumed to be 0.7–0.9.

A triangular distribution was tentatively adopted for BC_U , N_U , N_I and f_{DE} , in which the estimated value is assumed most likely, the probabilities for the interval limits are set to 0, and a linear probability distribution is assumed between the most likely value and the interval limits.

The $(\text{Bc}/\text{Al})_{\text{crit}}$ for the vegetation at TSP (mainly masson pine) was suggested as 2.0 with a uniform distribution from 0.5 to 2.0. The α and $\log K^*$ were estimated as 1.59 and 3.11 respectively based on the linear regression between pH and pAl^{3+} in soil water at TSP (10). Q was monitored as $5220 \text{ m}^3\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$, and assumed to follow a normal distribution with SE 20% of the mean value.

A summary of input data with uncertainty assumptions for the critical load calculation in TSP is shown in Table 1.

Based on eqs 5 and 6, the extended S–N–BC critical load function was determined for TSP catchment, described as a surface in Figure 3. To draw the surface, the characteristic values, e.g., $\text{CL}_{\text{max}}(\text{S})$, $\text{CL}_{\text{max}}(\text{N})$ under current BC deposition, $\text{CL}_{\text{max}}(\text{S})^*$, $\text{CL}_{\text{max}}(\text{N})^*$ under assumed natural BC deposition (25% of current deposition), and $\text{CL}_{\text{min}}(\text{N})$, were first calculated through eqs 2–4, and then labeled in the figure. The calculated critical loads at TSP are generally higher than the results for sensitive areas in Europe, partly due to higher temperature and humidity in the subtropics (23). The critical loads are highly dependent on the BC deposition. If BC deposition is cut by 75%, critical loads of S and N decline by 58%.

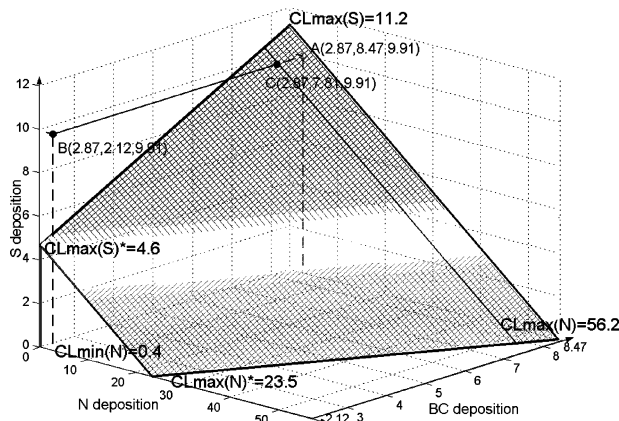


FIGURE 3. The S–N–BC critical load functions of TSP. $CL_{\max}(S)/(N)$ and $CL_{\max}(S)/(N)^*$ represent the maximum S/N critical load under current BC deposition and 25% of current BC deposition, respectively. $(Bc/Al)_{\text{crit}}$ was used as the chemical criterion to calculate the $ANC_{L,\text{crit}}$ based on the eq 6 and the critical load function is a curvilinear surface. However, the curvilinear section is too small compared with the whole range, and thus not obvious in the figure.

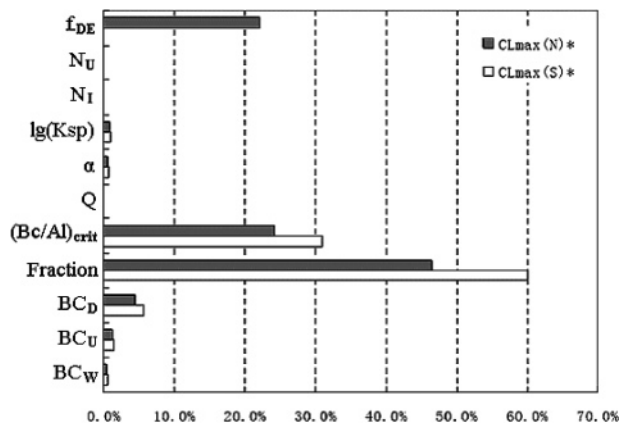


FIGURE 4. The variance contributions of different parameters to $CL_{\max}(S)^*$ and $CL_{\max}(N)^*$ in TSP.

Average N, BC, and S deposition in 2002 and 2003 at TSP were 2.87, 8.47, and 9.91 $\text{keq}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ (point A in Figure 3). With these deposition values, the critical load was not exceeded, although very close to the current S deposition. The high BC deposition therefore mitigates exceedance of the critical loads. The acidifying deposition ceiling would be reached when BC_D decreased to 7.81 $\text{keq}\cdot\text{ha}^{-1}\cdot\text{yr}^{-1}$ i.e. 92% of current value (point C), and would be far exceeded when BC_D decreased by 75% (point B), if S or N deposition is unchanged. Based on these results, acidification effects are a realistic risk in the future at TSP.

Uncertainty analysis was carried out using a Monte Carlo method to calculate the contribution to the overall uncertainty from various input parameters (30). The input data with their uncertainty assumptions were placed in the Monte Carlo framework, and 100 000 simulations were performed.

The uncertainty of the natural BC deposition fraction contributed most to both $CL_{\max}(S)^*$ and $CL_{\max}(N)^*$ variance in TSP (60 and 46% respectively) (Figure 4). This confirmed that the critical load and thus the target for emission control in China can be influenced greatly by the uncertainty in BC deposition estimation, especially in the areas where high current BC deposition occurred. $(Bc/Al)_{\text{crit}}$ also contributed to considerable uncertainty in critical loads for S and N (31 and 24%, respectively). The transformation of N in ecosystems occurs by many biochemical processes, and is much more complicated than S. The variation of f_{DE} contributed considerably to uncertainty in $CL_{\max}(N)^*$ (22%). N_I and N_U , though

quite uncertain, had very little effect on $CL_{\max}(N)^*$. However, they could greatly affect $CL_{\min}(N)$, which was much smaller than $CL_{\max}(N)^*$ in this study. The uncertainties of other parameters were of little importance on the results.

4. Discussion

The influence of BC deposition on acidification in the future has been assessed in some model applications. The dynamic acidification model MAGIC has been used to predict future acidification development in two catchments in southern China based on scenarios with different future deposition of S and Ca (6). The results indicated that with large decreases in BC deposition, increased soil acidification can be expected even with considerable S emission reductions. An and Huang (7) used a long-term soil acidification model (LTSAM) to show that a large reduction in BC deposition could decrease the critical load heavily for red earth in Zhejiang, China. Although critical load calculation based on current BC deposition might be acceptable in Europe due to relatively low deposition levels, it would lead to wrong conclusions in the areas with high current BC deposition, e.g., in China.

The critical load concept is made for policy purposes, and would be of little use if not able to assist in decision making. The S–N–BC critical load function, as developed here, could provide for more reasonable and useful critical load estimation in possible future scenarios. Policy makers would be able to clearly understand the necessity and required amount of acid deposition abatement along with the BC deposition reduction, and therefore plan a comprehensive emission control strategy for both acid compounds and particle matter. Taking the Chongqing city for example, the PM_{10} level in the year 2004 was $142 \mu\text{g}\cdot\text{m}^{-3}$, and thus nearly 30% of PM emission is required to be cut, in order to meet public health standards (the national standard of PM_{10} is $100 \mu\text{g}\cdot\text{m}^{-3}$ for residential areas). If this reduction is taken into account in the extended critical load function at TSP, 20% of SO_2 should be simultaneously cut to prevent soil acidification. According to the previous government plan, however, only 10% reduction of SO_2 is required. Therefore, it is clearly shown for policy makers that soil acidification in TSP is an urgent risk, and more corresponding SO_2 emission abatement is needed. Even in Europe, the S and N emission ceilings based on critical loads should be reevaluated in the context of visible reduction of BC emission and deposition. Hedin et al. (31) indicated the historical steep declines in BC deposition in Europe, and recent monitoring data from Europe confirm the declining trends. In Finland, for example, BC concentration in bulk precipitation decreased by 15–35% because of the emission reduction actions both in Finland and elsewhere in Europe (32). This reduction is partially a consequence of reduced S deposition. Under this situation, the S–N–BC critical load function can be applied for policy makers to quantify the effects of variation of BC deposition, and make more efficient strategies to prevent acidification in the future.

It is important to distinguish between natural and anthropogenic BC deposition for the critical load estimate. However, it is quite difficult to evaluate the current status and future trend of anthropogenic BC deposition due to lack of data. Long-term monitoring of BC deposition and modeling work of emission and dispersion is thus strongly recommended in order to improve the acidifying critical load results.

In the European mapping of critical loads, $(Bc/Al)_{\text{crit}}$ was most commonly applied as the effects criterion and most commonly set to 1.0, although the dose response relationship is quite uncertain (33). Besides $(Bc/Al)_{\text{crit}}$, the chemical criteria for $ANC_{L,\text{crit}}$ calculation contain critical aluminum concentration $[Al]_{\text{crit}}$, critical pH $[H]_{\text{crit}}$, and critical aluminum mobilization rate Al_w , etc (18). However, $[Al]_{\text{crit}}$ is usually for drinking water (groundwater) protection, and $[H]_{\text{crit}}$ is used

for organic soils (e.g., peatland and bogs). With regard to Al_w , the necessary parameter p (the stoichiometric ratio of Al to BC weathering in primary minerals) is very uncertain in China. Therefore, $(Bc/Al)_{crit}$ is applied in the catchment calculation. The value for main vegetation types in Chinese subtropical areas was suggested to lie in the range 1.0–2.0, but little data are available to test this (22). At TSP, the molar ratio of BC to Al in soil water was found within or above this range. The absolute Al concentrations were much higher than commonly found in Europe (10), hence the influence of BC on the Al toxicity may be weakened and harmful effect to plants might occur (34). Therefore, the rationality of this approach applied in heavy polluted areas needs to be further assessed.

There are also limitations in the critical load calculation with the extended SSMB function. The method describes steady-state conditions without any dynamic consideration, and thus cannot illustrate the process of recovery from soil acidification, which becomes an important basis for control strategies in Europe where considerable reduction in acid deposition has been achieved (35). Under this situation, some other strategy analysis tools such as Dynamic Impact Analysis (35) and dynamic target load (36) have been developed to estimate recovery from soil acidification. Unlike Europe, however, China is still suffering heavy air pollution and high amounts of acid deposition, and is thus at the beginning stage of emission control and deposition abatement. Calculating critical load with SSMB is still useful and appropriate for acidification control policy.

Adsorption and desorption of sulfate are not taken into account in the SSMB. Sulfate desorption might occur along with decreasing S deposition in the future and then delay the reversal of soil acidification (37). Even though it has been reported that the capacity for sulfate desorption in south Chinese soils is quite large (38), the current amount of S accumulated in TSP is limited and cannot influence the acidification process very sharply if S deposition does not increase in the future (10). Therefore, the process of sulfate adsorption and desorption in the soil was neglected in this study.

Acknowledgments

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Supporting Information Available

SSMB method for critical load calculation; $ANC_{L,crit}$ calculation using $(Bc/Al)_{crit}$; and site description of TSP catchment. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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