Higher soil acidification risk in southeastern Tibetan Plateau

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Abstract

Stable soil pH is a key property in maintaining an ecosystem’s structure, function, and sustainability. Increasing atmospheric deposition and grassland use on the Tibetan Plateau (TP) may increase the soil acidification risk, but we lack such information to date. Here, we evaluated the soil acidification risk in the TP, by comparing it with that in the Mongolia Plateau (MP) and applying the acid–base balance principles on atmospheric inputs, soils, and plants from 1980 to 2019. Cumulative acid input was lower in the TP than in the MP. Sulfur contributed more to acidity than nitrogen and atmospheric deposition contributed more to acidity than grassland use. Acid input was mainly influenced by local industry, animal husbandry and transportation in the MP, while in the TP it was also affected by the long-distance transportation of pollutants from South Asia and southern China. Overall, the TP was less acid-sensitive than the MP because of higher inorganic carbon content. However, soils in the southeastern TP, covering 21% of the total area, were acid-sensitive due to low levels of soil exchangeable base cation (EBCs) and lack of calcium carbonate. Coincidentally, the southeastern region has the highest concentration of acid input in the TP due to more rapid development and stronger influence of adjacent high acid deposition regions than others. Therefore, the acidification risk to the southeastern region is much higher than to other regions of the TP and the MP; in this region, the EBCs are likely to be depleted approximately 95 years earlier than in the MP. The findings of this study provide insights into the response of the TP to global change. For the ecosystem sustainability of southeastern TP, control of atmospheric acid deposition, especially sulfur deposition, in both local and adjacent regions and nations is required.
Keywords: deposition, uptake, nitrogen, carbonate, exchangeable base cation, depletion

1. Introduction

Soil pH significantly influences soil biogeochemical cycles, biodiversity, productivity, and many other factors in terrestrial ecosystems (Chytrý et al., 2007; Kirk et al., 2009). In the past decades, the release of SO₂ and NOₓ from fossil fuel combustion and high-energy combustion has led to a two- to seven-fold increase in atmospheric acid deposition relative to the time before the industrial revolution (Barak et al., 1997; Lu et al., 2014). Moreover, the emission of NH₃, which has strong acidification potential, has also increased rapidly with the development of agriculture and transportation (Du et al., 2015). Meanwhile, increased use of grasslands or forests for, such as, grazing, mowing, and harvesting, has removed base cations from the soil and further exacerbated soil acidification (Bolan et al., 1991; Fujii et al., 2012). The exposure of terrestrial ecosystems to long-term high levels of anthropogenic acid input may lead to soil acidification and significantly change ecosystem structure and function (Chen et al., 2013a). Understanding the risk of soil acidification is important to maintain the stable and sustainable development of regional ecosystems.

Changes in soil pH are generally assumed to reflect the level of soil acidification (Ji et al., 2014); however, this approach can only reflect past record of acidification rather than explore the acidification risk in the future. Using the principles of acid–base balance and integrating atmospheric properties, soil acidification processes, and the interactions between the atmosphere, soil, and vegetation provide a better assessment of the soil acidification risk (Fig. 1). Under natural conditions, dissolution of CO₂ released from roots and microbial activity and the ionization of organic acids are accompanied by the release of H⁺ (Fujii et al.,
2008; van Breemen et al., 1983); however, the natural acidification process is extremely slow, and more acid input is mainly contributed by atmospheric deposition and the loss of base cations caused by grassland utilization. Atmospheric deposition directly inputs H+ into the soil, and intensifies soil acidification through H+ production by the nitrogen cycle when nitrogen deposition enters the soil. In addition, plants take up base cations from the soil, and this process is accompanied by the release of H+ from plants. Once the plant is partially or completely removed due to grassland use, a portion of H+ released in the soil remains (Vries and Breeuwsma, 1987; Zhu et al., 2016). The input of excessive amounts of H+ to the soil and subsequent acidification is regulated by different buffering compounds in soils: by carbonates at pH > 7, by soil exchangeable base cations (EBCs) at 4.5 < pH < 7, and by aluminum compounds at pH < 4.5 (Bowman et al., 2008; Lieb et al., 2011). The methods for estimating soil acidification based on the principles of acid–base balance require a series of matched data of parameters related to the atmosphere, soils, and plants. Therefore, the lack of large scale studies of soil acidification based on this method is because of the high cost and increased time-consumption.

The Tibetan Plateau (TP), as the world’s ‘Third Pole,’ is highly susceptible to global climate change. In the past decades, scientists have researched its response to global change factors such as increasing temperatures (Guo et al., 2017), CO2 enrichment (Zhao et al., 2010), and alterations in rainfall (Zheng et al., 2014). Unfortunately, the response of grassland soils in the TP to acid input, particularly to the increasing atmospheric acid deposition and grassland utilization, has not received much attention. This is because previous studies argued that the soils in the TP were high in inorganic carbon (Mi et al., 2008; Yang et al., 2012),
indicating a low sensitivity to acidity. However, the inorganic carbon content in the TP soils is spatially heterogeneous; soils with high inorganic carbon content mainly occur in the TP’s northwestern regions, with much lower levels in the southeastern regions. Coincidentally, the southeastern region is the major development area of the TP owing to its lower elevation and higher temperatures than other TP areas (Figs. S1). Furthermore, influenced by East Asian and South Asian monsoon (Fig. 2), it has relatively high levels of atmospheric acid deposition transported from central China and South Asia (Wang et al., 2010); even though little atmospheric acid deposition is created on the TP itself as it does not have highly developed agriculture and industries. Therefore, assessing the risk of soil acidification in the Earth’s Third Pole region is a prerequisite for maintaining its sustainable development.

For effective assessment of the soil acidification risk in the TP, we selected the Mongolian Plateau (MP) for comparative study (Fig. 2). The Mongolian Plateau is the most representative temperate grassland in the northern hemisphere. Compared with the TP, the MP has more developed industries and agriculture, and a higher ratio of carbonate area to total area. Different levels of human activity and soil acidity buffer capacity may lead to differences in their acidification risks. Meanwhile, due to different regional emission reduction strategies, the contribution of nitrogen and sulfur deposition to their acidity differs, and therefore quantification of contributions from different sources can provide guidance for future policy-making. Thus, the main objectives of the study were to: (1) quantify the levels of acid input and the contribution ratio of different factors between the two comparative plateaus; (2) ascertain the sensitivity to acid input of grassland soils between the two plateaus; (3) compare the durations of depletion of different acid buffering pools in the TP with those in
the MP. In particular, the assessment of soil acidification and its influencing factors may aid in formulating better strategies to manage existing policies and the regulatory framework to improve the region’s socioeconomic development while reducing the risk of acidification.

2. Materials and Methods

2.1 Study area

The TP and the MP are the representatives of alpine and temperate grasslands in the northern hemisphere, respectively. They both cover all of the main types of grasslands, such as meadow, steppe, tussock, herb-based wetland, and desert grassland (Fig. 2). The TP is the most extensive alpine polar grassland in the world (Yang et al., 2012). In the TP, the mean annual temperature (MAT) is –0.06 °C and the mean annual precipitation (MAP) is 661 mm. Soil types are mainly Cumulic Anthrosols, Calcaric Cambisols, and Haplic Luvisols. In contrast, the MP is an important part of the Eurasian steppe: the MAT is 2.67 °C, the MAP is 367 mm, and the main soil types are Calcaric Fluvisols, Calcic Gleysols, and Haplic Kastanozems (HWSD version 1.1). Economic development is much lower in the TP than the MP (Fig. 2). Therefore, comparative analyses of these regions should help in making better decision regarding their sustainability under the scenarios of high acid input from atmospheric deposition and grassland use.

2.2 Calculation of net $H^+$ input

2.2.1 Net $H^+$ input by atmospheric deposition

Atmospheric deposition not only inputs $H^+$ into the soil through acid deposition, but also
generates H\(^+\) through the nitrogen cycle process occurring after nitrogen deposition enters the soil, and this further acidifies the soil. Therefore, the net H\(^+\) input by atmospheric deposition is the sum of the two. Based on the principle of charge balance, the H\(^+\) input by acid deposition is calculated by Eq. (1):

\[
AC_D = SO_{4, D}^2^- + NO_{3, D}^- + Cl_{D}^- - NH_{4, D}^+ - BC_{D}^{2+}
\]  

where \(SO_{4, D}^2^-\), \(NO_{3, D}^-\), \(Cl_{D}^-\), \(NH_{4, D}^+\) are the total depositions of these elements; base cations \((BCD^{2+})\) is the sum of total deposition of Na\(^+\), Mg\(^{2+}\), K\(^+\), and Ca\(^{2+}\). All units are kmol ha\(^{-1}\) yr\(^{-1}\).

According to the research study by De Vries and Breeuwsma (1987), the total H\(^+\) production of the nitrogen cycle including nitrification, plant absorption, and denitrification is calculated by Eq. (2):

\[
AC_N = NH_{4,L}^+ - NH_{4,L}^- + NO_{3,L}^- - NO_{3,D}^-
\]

where \(NH_{4,L}^+\) and \(NO_{3,L}^-\) are the leaching amounts of NO\(_3^-\) and NH\(_4^+\), respectively. Since the mobility of NH\(_4^+\) in the soil is poor, and most of it is absorbed by plants or nitrated, we assume that no significant amounts of NH\(_4^+\) is leached from the soil (Duan et al., 2004). The mass balance equation of nitrogen, used to relate the deposition and leaching of nitrogen, is as follows:

\[
N_L = N_D - N_U - N_{de}
\]

where \(N_D\), \(N_U\), and \(N_{de}\) are the deposition of total nitrogen, plant absorption, and denitrification, respectively. \(N_L\) is the leaching amount of total nitrogen (since the leaching of NH\(_4^+\) is assumed to be 0, the leaching amount of total nitrogen is equal to that of NO\(_3^-\)).
Through our field surveys, we found that legumes were not the dominant species in the study area, so we did not consider the nitrogen fixation of legumes.

By summing up Eq. (1–3), we finally calculated the net $H^+$ input from atmospheric deposition using Eq. (4):

$$H_{dep} = SO_4^{2−} + (NH_x + NO_y - N_{de} - N_{u,dep}) - BC^{2+} + Cl^- \quad (4)$$

where the sum of $NH_x$ and $NO_y$ is the total nitrogen deposition; $N_{de}$ is the N that undergoes denitrification, calculated by the product of denitrification coefficient and net N input (Duan et al., 2001); and $N_{u,dep}$ is the uptake of N by plants from atmospheric deposition, the proportionality coefficient comes from the results of $^{15}$N tracer field studies (Templer et al., 2012). All parameters are expressed in kmol c ha$^{-1}$ yr$^{-1}$.

### 2.2.2 Net $H^+$ input via use of grasslands

Crop removal is another primary source of acidity. Plant roots excrete $H^+$ or $OH^-$ as they absorb cations or anions from the soil. Theoretically, if all the plant residues were returned in situ to the soil and no N losses or acid inputs occurred, the soil’s net acidification would be zero because of the deacidifying processes occurring during the decomposition of plant materials. However, under human interference, such as grazing and mowing, some elements are taken off (and can be considered permanently lost) from the grasslands, leading to a portion of the acidity remaining in the soil. Equations 5–7 were used to calculate the $H^+$ input via the use of natural grasslands by humans.

$$H_{use} = BC_{upt} \quad (5)$$

$$BC_{upt} = (BC_s \times Bio_s + BC_i \times Bio_i) \times 10^{-3} \quad (6)$$

where $BC_{upt}$ (kmol c ha$^{-1}$ yr$^{-1}$) is the net loss of base cations (BC) from grazing and mowing
grasslands. We did not calculate the N-uptake-induced proton caused by nitrogen absorption in this part because it has been calculated in atmospheric deposition.

\[
Bio_s = \text{Ratio}_s \times Bio \times C_{gra} \times 10
\]  
\[
Bio_l = \text{Ratio}_l \times Bio \times C_{gra} \times 10
\]

where \(BCS\) and \(BCL\) (mmol g\(^{-1}\)) are the element contents of BC in the stems and leaves, respectively; \(Bios\) and \(Bio_l\) (kg ha\(^{-1}\)) are biomasses of the stems and leaves consumed by grazing, respectively; and \(\text{Ratio}_s\) or \(\text{Ratio}_l\) and \(Bio\) (g m\(^{-2}\)) are the ratio of stem or leaf biomass to total biomass, and total biomass, respectively. \(C_{gra}\) is the grazing strength coefficient, which is determined by the number of standard sheep per hectare of grazing in different provinces. The N absorption of plants is calculated similarly, replacing the element content of BC with N.

### 2.3 Assessment of the soil buffering system

Soils can buffer atmospheric acid deposition using carbonates, soil EBCs, and aluminum compounds in different pH ranges. When the acid buffering reaches aluminum compounds, soil pH is already relatively low, and the heavy metals start to have irreversible toxic effects, which is why we only calculated the content of carbonates and EBCs using Eq. (9) and Eq. (10):

\[
Pool_{Ca} = C_{CaCO_3} \times BD \times T \times \left(1 - \frac{Ci}{100}\right) \times 10^5 / 50
\]

\[
Pool_{EBC} = C_{BC} \times BD \times T \times \left(1 - \frac{Ci}{100}\right) / 100
\]

where \(C_{CaCO_3}\) and \(C_{BC}\) are the weight-percent concentration of calcium carbonate (\%) and concentration of EBC (cmol\(\_\text{c} \) kg soil\(^{-1}\)) in the soil, respectively; \(BD, T,\) and \(Ci\) represent bulk density (g cm\(^{-3}\)), soil thickness (m; 0.1 m was used to represent topsoil), and percentage.
of rock fraction (diameter >2 mm), respectively; and $\text{Pool}_{\text{Ca}}$ and $\text{Pool}_{\text{EBC}}$ represent the content of calcium carbonate and EBCs in the topsoil (kmol$_c$ ha$^{-1}$), respectively.

### 2.4 Calculating duration of complete depletion of different acid buffering pools

The duration of depletion of different acid buffering pools was calculated to assess longevity (sustainability) of the acid neutralizing capacity to indicate the risk of soil acidification in grasslands. Recent research has shown that N and BC depositions in China have remained stable in recent years (Yu et al., 2019; Zhang et al., 2020), while S deposition has continued to decline (Yu et al., 2017); hence, we used the level of H$^+$ input in the 2010s to calculate the minimum duration of soil acid neutralization capacity under the relevant atmospheric deposition conditions (Eq. 11).

\[
\text{Deple}_i = (\text{Pool}_i - H_{\text{acc}}) / H_{2010s}
\]

where $\text{Deple}_i$ represents the predicted duration (years) of calcium carbonate or EBC depletion in soils, $H_{\text{acc}}$ is the accumulated H$^+$ input from the 1980s to the 2020s (kmol$_c$ ha$^{-1}$), and $H_{2010s}$ is the H$^+$ input in 2010s (kmol$_c$ ha$^{-1}$ yr$^{-1}$).

### 2.5 Data sources

We divided the period from 1980 to 2019 into four phases designated as the 1980s, 1990s, 2000s, and 2010s. We calculated the spatial pattern and input amount of acids in different phases. To calculate the H$^+$ input, data on the complete atmospheric deposition, concentrations of plant N and BC, plant leaf-to-stem ratio, grazing intensity, and denitrification coefficients were required. The sources of these datasets are summarized in
Table S1. The data on the soil acid buffering system based on soil pH, bulk density, rock fraction, calcium carbonate content, and EBC content in the 1980s were obtained from China’s Second National Soil Inventory, which were derived from the book of Soil Species of China (published by the Office of National Soil Survey, 1993-1996). The data of soil pH in the 2010s were compiled from relevant publications. Meanwhile, data on livestock raising, gross industrial production, and vehicle ownership were obtained from the China Animal Husbandry and Veterinary Yearbook, China Statistical Yearbook for Regional Economy, and Inner Mongolia Statistical yearbook. All calculations and analyses were performed using SPSS 13.0, R 3.4.3, and ArcGIS 10.4 software packages.

3. Results

3.1 Contributions of acid input from different sources

The average H⁺ input from atmospheric deposition during the 1980–2019 period was significantly higher in the MP than in the TP; similarly, the proportion of the area of H⁺ input to the total grassland area was also higher in the MP than the TP (56% vs. 25%). In both areas, S and N deposition contributed approximately 60% and 40% to acid inputs, respectively. Furthermore, base cations deposition and part of the N cycling process offset 55% and 18% of the potential acidity, respectively (Table 1).

The inputs of H⁺ from atmospheric deposition varied over time, and showed different patterns and magnitude (Fig S2 and S3). In the TP, both H⁺ input and the proportion of H⁺ input area to total grassland area increased until the 2000s, and then decreased. High acid input was observed in the eastern parts of the TP in the 1990s and the southern parts of the TP
in the 2000s. In contrast, the acid input in the MP peaked in the 1990s, and then increased again in the 2010s.

Contrarily, the H⁺ inputs from the utilization of grassland during 1980–2019, were estimated as 0.33 ± 0.25 and 0.56 ± 0.20 kmolec ha⁻¹ yr⁻¹ in the TP and the MP, respectively. The utilization of grasslands led to high acid inputs in the central and northeastern parts of the MP and higher inputs in the southeastern part of the TP than in other regions of the TP (Fig. S4). In addition, over time, the H⁺ input of grassland utilization showed different trends in the TP and the MP. In the TP, it continued to rise until the 2010s; however, it showed a downward trend in the MP from the 1980s to the 2010s (Fig. S5).

3.2 Cumulative acid inputs on the two plateaus

The regions of high cumulative acid input were mainly distributed in the central MP and the southeastern TP (Fig. 3A). From 1980 to 2019, the cumulative amount and the proportion of H⁺ input area to the total grassland area in the TP were lower than those in the MP (27.95 ± 20.43 vs. 32.80 ± 19.54 kmolec ha⁻¹; and 47% vs. 92%) Furthermore, the contribution of grassland utilization to acid input was much smaller than that of atmospheric deposition. However, the contribution of grassland utilization to the proportion of H⁺ input area to the total grassland area was much larger than that of atmospheric deposition (Fig. 3B and C).

The accumulation input of acid in both the TP and the MP was significantly correlated with gross industry production (R² = 0.37, 0.49, p = 0.02, 0.01) and livestock cultivation (R² = 0.40, 0.66, p = 0.02, 0.001). Additionally, the acid accumulation input in the MP was also related to vehicle ownership (R² = 0.55, p = 0.01). Jointly, these factors accounted for 76% and
56% of spatial variations in the acid inputs of the MP and the TP, respectively (Fig. 4).

3.3 Different acid buffer pools in the two plateaus

The soil of the TP have a higher carbonate content but a lower ratio of carbonate area to the total area than those of the MP (Figs. S6 and S7). Carbonate-containing soils are mainly distributed in the northwestern regions of the TP and the central and western regions of the MP (Fig. S6). Conversely, EBCs in non-carbonate-containing soils were significantly lower in the TP than in the MP, but the ratio of non-carbonate-containing soil area to total soil area was higher in the TP than in the MP (36% vs. 3%, respectively) (Fig. 5A).

We estimated that, in the past 40 years, the total acid inputs have consumed only 0.4% of the soil calcium carbonate in the TP and 5% in the MP (Fig. S7A). However, over that period, EBCs decreased by 21% in the TP and 12% in the MP; approximately 1313 km² of soil was depleted of EBCs in the TP, which resulted in the release of Al³⁺ and Mn²⁺ (Fig. 5A).

3.4 Time until depletion of different acid buffer systems

In both the TP and the MP, calcium carbonate in carbonate-containing soils can last for several thousand years (Fig. S7B). Carbonate-containing soils accounted for 63% and 94% of the soils exposed to the area of acid input in the TP and the MP, respectively. However, in the TP, high acidity inputs were mainly concentrated in non-carbonate-containing soils, which led to higher acidification risk than in the MP. The EBCs may be depleted from these soils in approximately 312 years (Fig. 5B). In addition, the period until the depletion of these buffer systems in different grassland types shown in Table 2 showed that, of all the grasslands, the
4. Discussion

4.1 Contribution of different deposition composition to acidity

We first quantified the relative contributions of the primary sources of acid entering the soils of the TP and the MP. Atmospheric deposition was the most important source and the contribution of S deposition to acidity was higher than N deposition in both plateaus. This finding has also been demonstrated in forest ecosystems (Zhu et al., 2016). This occurs not only because the S deposition is still higher than the N deposition in these areas but also because the denitrification and plant absorption of N can partly offset potential acidity caused by N deposition. In addition, base cations deposition neutralized approximately 55% of the potential acidity. The findings of the present study are consistent with the results of studies on the acid neutralization capacity of atmospheric wet base-cation deposition by Zhang et al. (2020). High base cations deposition is the main reason for the lack of widespread acidification in China compared to Europe and North America. This finding demonstrated that various policies implemented by the Chinese government in the past several decades to control SO₂ emissions have been effective, and that these policies should be more vigorously enforced in the future. Furthermore, the development of industry related to high S emissions should be closely monitored in the TP considering its high risk of soil acidification.

4.2 Spatiotemporal variability of acid input in different plateaus

The lower cumulative amount and ratio of acid input area to total grassland area in the
TP than in the MP (Fig. 3B and C) may be attributed to the differences in their economic
development and industrial structure. The gross domestic product and proportion of secondary
industry of the MP were 17.07 and 1.63 times as much as those of the TP, respectively,
leading to a higher level of S and N deposition in the MP than in the TP. Meanwhile,
grassland utilization is also a significant source of acid input for long-term over-utilized
grassland ecosystems and the grazing intensity in the MP was also higher than that in the TP
(Huang et al., 2016; Xie and Wu, 2014). The high cumulative acid inputs in the central parts
of the MP and the southeastern regions of the TP coincided with their higher population
density and economic level (Fig. S1D and E). The spatial variations in cumulative acid inputs
were significantly correlated with the gross industrial production and animal husbandry n in
both plateaus (Fig. 4). High amounts of SO$_2$ and NO$_x$ produced by industrial emissions were
important precursors of acid deposition (Ianniello et al., 2011). The numbers of livestock
affected not only the acid input from grazing, but also NH$_3$ emissions (Yu et al., 2019).
Furthermore, vehicle ownership was also positively correlated with acid input in the MP, and
this is because the MP’s industrial structure has undergone a transformation. The
transportation industry has become a factor that cannot be ignored, and vehicle exhaust is
becoming the main source of NH$_3$ emissions (Zheng et al., 2018). It is important to note that
local industry and animal husbandry explain only 56% of the TP variation, and much of the
acidity may be due to long-distance transport of pollutants from East Asia and southern China
influenced by the South and East Asian monsoons (Fig. 2). Studies by Fu et al. (2006) and
Wang et al. (2016) also confirmed this hypothesis because, their model predictions clearly
showed that the Tibetan anticyclone could trap anthropogenic emissions lifted from South
The temporal evolution of acid input in both plateaus (i.e., increasing, then declining) was mainly induced by changes in atmospheric deposition. Although grassland utilization changed during the 1980s–2010s, the contribution of grassland use to acid input was small, and the effects did not impact the overall trend. Socioeconomic changes and effective environmental policies were the primary factors influencing this change (Yu et al., 2019). The continued increase of acid inputs in the early years was mainly due to the rapid development of industry and livestock, accompanied by increasing energy consumption and grazing intensity since the reform and opening of China in the late 1970s (Guo et al., 2010; Zhang et al., 2018). Subsequently, in order to control SO$_2$ emissions, China began to amend its development model and implement a series of regulations and policies at the end of the 19$^{th}$ century, such as the adoption of coal desulfurization technology and the implementation of the Cleaner Production Promotion Law (Yu et al., 2019; Zhao et al., 2009), which sharply reduced the acid input. However, in the MP, the wet deposition of SO$_4^{2-}$ continued to increase in the 2010s, probably because of continuous urbanization. From the historical perspective of acid input, the development of society and economy is bound to be accompanied by a large amount of acid pollutant emissions due to the high demand for energy consumption. Thus, we maintain the balanced development of society and the environment only by vigorously developing and implementing new energy technologies with lower pollutant emissions and improved environmental protection measures.

4.3 Higher acidification risk in the southeastern region of the TP
Our results showed that the soils in the southeast of the TP were mostly non-carbonate-containing. This situation is mainly a consequence considerable great spatial heterogeneity of climate in the TP, as it is usually warm and wet in the southeast (with an annual average temperature of ~20 °C and precipitation up to 1000 mm) and cold and dry in the northwest (with an annual average temperature below 0 °C and precipitation < 50 mm) (Wang et al., 2012) (Fig.S1). During the long process of natural acidification, precipitation leaching occurs when the amount of precipitation is greater than the level of evaporation, in which case the alkaline substances exchanged by H⁺ are easily leached out of the soil, thereby reducing soil alkalinity (Slessarev et al., 2016). The soils in this area mainly rely on EBCs for buffering acidity, and the buffering capacity of the TP soils is much lower than that of the MP soils, which primarily contain carbonates. The acid buffer capacity of 1% carbonate in the topsoil is approximately 40 times higher than the EBCs of 1 mmol kg⁻¹. Thus, the surface soils in southeastern TP have higher acid sensitivity than those in the MP. With the most intense human activities, the southeast of the TP is the most economically developed region, and it is also affected by long-distance pollutants from central China and South Asia, thereby incurring a double acidification risk. This high risk means that significant soil pH decline and heavy metal release are expected within the next few centuries. In contrast, the soils exposed to the area of acid input in the MP are mostly carbonate-containing soils, which can maintain the pH at high levels for millennia.

There are still some uncertainties in this study. Because soil acidification is a complex process, researchers often simplify the calculation by considering only the acid input from human activities but ignore natural acidification, such as the dissociation of organic acid and
carbonic acid. Meanwhile, we ignore the supplement of exchangeable basic cations by soil mineral weathering, because it is relatively slow in the southeast of TP and north of MP (Duan et al., 2000). This practice may lead to some underestimation of soil acid input and soil acid buffer capacity. Therefore, to verify the reliability of our results, we also collected soil pH data published from the 1980s and 2010s for the TP and the MP, matched the data by the random forest method, and finally calculated the change in soil pH. Data reliability analyses demonstrated that in the TP, the changing trends in soil pH supported our main findings that the soils of the TP have a greater risk of acidification than the soils of the MP because of higher acid sensitivity (Fig. 6).

Because of extremely high elevations on the TP, its ecosystems are inherently fragile and unstable under environmental changes (Chen et al., 2013b). Under dramatic environmental changes, the thin surface soils and low biological buffering capacity decrease the resistance of these alpine ecosystems to soil acidification, resulting in the death of several plant species and even the collapse of entire ecosystems. Van et al. (2011) showed that soil acidity could explain most of the variations in herb species richness in the United Kingdom. Chen et al. (2013a) proved that soil acidification reduced ecosystem productivity by altering below-ground communities and soil properties. Therefore, in the TP, which is an ecologically fragile area with a high acidification risk, it is essential to handle the relationship between economic development and ecological protection carefully and properly. The identification and quantification of acid input sources is a crucial step in developing a reasonable strategy for ensuring the TP’s sustainability. According to the results of the present study, the source of acid in the TP was mainly S deposition, which primarily originates from power generation,
industry, and transportation development (Vet et al., 2014). Furthermore, acidogenic pollutants transmitted long-distance from adjacent high acid deposition areas are another significant source of acidity. In South Asia, countries such as India and Nepal have rapidly developing industries and booming populations, which will lead to a continued long-term impact on the TP in the future. In addition to enforcing the development of local clean energy and reducing the heavy industry presence, joint control of acid deposition by multiple adjacent countries should also be considered to maintain ecological sustainability on the Tibetan Plateau.

5. Conclusions

To the best of our knowledge, this study is the first evaluation of acidification levels in the TP based on the principle of acid–base balance. We showed that S deposition contributed more than N deposition to acidity in the TP and the MP. More importantly, the non-carbonate-containing soils in the TP’s southeastern parts are more sensitive to acid than soils in the MP. Coincidently, as high acid inputs are concentrated in this region because of rapid regional development and deposition from south Asia and southern China, all evidence suggests that, compared to the MP, the southeastern region of the TP (21% of the TP) will face a greater risk of acidification in the future. In addition, as the most ecologically fragile region in China, increased soil acidification may irreversibly affect the structure and function of the entire alpine ecosystems that dominate Tibet. Therefore, increasing environment protection by decreasing heavy industries with high S emissions or promoting clean energy in the southeastern regions of the TP and adjacent areas (both southern China and South Asian...
countries) is essential for alleviating the risk of acid deposition in the soils and maintaining sustainability with reasonable economic development of the plateau in the future.
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Zhu QC, De Vries W, Liu XJ, Zeng MF, Hao TX, Du EZ, et al. The contribution of atmospheric deposition and forest harvesting to forest soil acidification in China since 1980. Atmos Environ 2016; 146: 215-222.
Table 1 Contribution of different components of atmospheric deposition and soil nitrogen cycle to soil acid input for the Tibetan Plateau (TP) and the Mongolian Plateau (MP)

| Year | S deposition | N deposition | N loss in N cycle | Base cations deposition |
|------|--------------|--------------|-------------------|------------------------|
| TP   |              |              |                   |                        |
| 1980s| 42 †         | 58           | -15‡              | -77                    |
| 1990s| 62           | 38           | -19               | -43                    |
| 2000s| 83           | 17           | -8                | -49                    |
| 2010s| 58           | 42           | -26               | -58                    |
| Average| 61          | 39           | -17               | -57                    |
| MP   |              |              |                   |                        |
| 1980s| 58           | 42           | -19               | -63                    |
| 1990s| 68           | 32           | -13               | -44                    |
| 2000s| 45           | 55           | -29               | -55                    |
| 2010s| 64           | 36           | -15               | -55                    |
| Average| 59          | 41           | -19               | -54                    |

† A contribution greater than zero represents a positive contribution to acid input
‡ A contribution less than zero represents a negative contribution to acid input
| Acid buffering substances | Grassland types       | Acid input area (km²) | Proportion of acid input area (% of total) | Duration to complete depletion of acid buffering substances (years) |
|---------------------------|-----------------------|-----------------------|-------------------------------------------|---------------------------------------------------------------|
| CaCO₃                     | Meadow                | 102,891               | 35                                        | 3008 ± 2426†                                                 |
|                           | Steppe                | 472,167               | 42                                        | 1870 ± 2461                                                  |
|                           | Tussock               | 3159                  | 5                                         | 1880 ± 1300                                                  |
|                           | Herb-based wetlands   | 38,560                | 42                                        | 2632 ± 2429                                                  |
|                           | Sparse grassland      | 149,488               | 20                                        | 5424 ± 3627                                                  |
| Exchangeable base cation  | Meadow                | 78,087                | 26                                        | 359 ± 307                                                    |
|                           | Steppe                | 103,005               | 9                                         | 302 ± 254                                                    |
|                           | Tussock               | 424                   | 1                                         | 581 ± 496                                                    |
|                           | Herb-based wetlands   | 20,603                | 22                                        | 373 ± 252                                                    |
|                           | Sparse grassland      | 26,360                | 4                                         | 412 ± 318                                                    |

† Data were represented as mean ± standard deviation
Figure captions

Fig. 1 Core processes of soil acidification. The black arrows represent the source and input of atmospheric deposition. The red and blue arrows represent hydrogen production and consumption, respectively. The orange arrow represents the process without hydrogen generation and consumption. The thinner lines represent natural acidification. The dotted box shows the buffer system of the soil at different pH stages.

Fig. 2 Direction of long-distance transmission induced by the East Asian and South Asian monsoons, and basic characteristics of the Tibetan Plateau (TP) and the Mongolian Plateau (MP): mean annual temperature (MAT), mean annual precipitation (MAP), and gross domestic product (GDP). Data with the same letters indicate no significant difference at $P = 0.05$. The orange arrows represent the summer wind direction and the blue arrow represents the winter wind direction. The small red arrows represent atmospheric acid deposition from south Asian countries and southern China to the TP over long distances due to monsoons.

Fig. 3 Spatial patterns (A), cumulative acid input and contribution of different sources of acidity (B), and the area receiving acid input as a proportion of the total grassland area and contribution of different sources to the proportion of the area receiving acid input (C) during 1980–2019. Error bars are standard deviations. Data with the same letters indicate no significant difference at $P = 0.05$. TP, Tibetan Plateau; MP, Mongolian Plateau. From 1980 to 2019, cumulative acid input was mainly observed in the southeast of TP and the central of MP. Both flux and the area receiving acid input as a proportion of the total area of cumulative acid
input were lower in the TP than in the MP. Atmospheric deposition contributed more to
acidity but less to the area receiving acid input than grassland use types.

Fig. 4 Variation partitioning among the factors influencing the cumulative acid input in the
Tibetan Plateau (A) and the Mongolian Plateau (B) obtained by redundancy analysis.
Livestock, livestock raising; GIP, gross industry production; and vehicle, vehicle ownership.
The spatial variation of cumulative acid input in the Tibetan Plateau is mainly influenced by
livestock raising and industrial development, and livestock breeding has a greater influence.
In the Mongolian Plateau, livestock raising, industrial development, and vehicle ownership
jointly explained 76% spatial variations of cumulative acid input

Fig. 5 Exchangeable base cation (EBC) content in the 1980s (A) and duration of depletion of
EBCs (B) as a result of atmospheric deposition and grassland utilization in the 2010s in the
Tibetan Plateau (TP) and the Mongolian Plateau (MP). The values on the arrows represent the
rate at which EBCs were consumed by the acid input during 1980–2019. Error bars are
standard deviations and data with the same letters indicate no significant difference at $P = 0.05$. The content of EBC in the 1980s was lower in the TP than in the MP. However, the
consumption of EBC was higher in TP than in MP. Moreover, EBC in the TP will be
exhausted sooner than in the MP.

Fig. 6 Soil pH in the 1980s and 2010s (A), and reductions in soil pH in different soils types (B)
and different plateaus (C) between the 1980s and 2010s. Error bars are standard deviations,
and data with the same letters indicate no significant difference at $P = 0.05$. 

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Fig. 1

SO₂ + O₃ → 2SO₂, SO₂ + H₂O → H₂SO₄
SO₂ + H₂O → H₂SO₃, 2H₂SO₃ + O₂ → 2H₂SO₄
2NO + O₂ → 2NO₂, 3NO₂ + H₂O → 2HNO₃ + NO

Emission → Wet Deposition → Dry Deposition

BC deposition → Nitrogen deposition

Acid deposition

NH₃

BC

SO₂

NO₂

N₂O/N₂

Leaching Loss

Ca²⁺, Mg²⁺, K⁺, Na⁺, NO₃⁻, SO₄²⁻, HCO₃⁻
Fig. 2
Fig. 3
Total variation explained: 56%

Total variation explained: 76%

Fig. 4
Soil content of EBC (kmol ha⁻¹)

Duration of depletion of EBC (years)

A

Decrease 21%

B

Decrease 12%

Fig. 5
Soil pH

![Bar chart](Fig. 6)

**Fig. 6**
Acknowledgements

This work was supported by the Nature Science Foundation of China (31872690, 31570471, 31988102), Chinese Academy of Sciences Strategic Priority Research Program (XDA19020302), the National Key R&D Project of China (2017YFA0604803).

Jianxing Zhu acknowledges support from National Postdoctoral Program for Innovative Talents (BX20180300) and Postdoctoral Science Foundation of China (2019M650829). We are grateful to the ecological stations and all monitors from the Chinese Ecosystem Research Network (CERN) for sample collecting. Data share should contact N. P. He (henp@igsnrr.ac.cn) or Q. F. Wang (qfwang@igsnrr.ac.cn)

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