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Key Points:
- Long-term changes of topsoil pH in forest ecosystems were explored
- Different pH dynamics were observed in broadleaved and coniferous forests
- Deposition of sulfur and nitrogen regulated the magnitude of soil pH change

Supporting Information:
- Text S1 and Figures S1–S8

Correspondence to:
Y. Yang,
yhyang@ibcas.ac.cn

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Long-term changes in soil pH across major forest ecosystems in China

Yuanhe Yang[1], Pin Li[2], Honglin He[3], Xia Zhao[1], Arindam Datta[4], Wenhong Ma[5], Ying Zhang[6], Xuejun Liu[7], Wenxuan Han[7], Maxwell C. Wilson[8], and Jingyun Fang[1,2]

[1]State Key Laboratory of Vegetation and Environmental Change, Institute of Botany, Chinese Academy of Sciences, Beijing, China, [2]Department of Ecology, and Key Laboratory for Earth Surface Processes of the Ministry of Education, Peking University, Beijing, China, [3]Key Laboratory of Ecosystem Network Observation and Modeling, Institute of Geographical Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing, China, [4]Earth Sciences and Climate Change Division, The Energy and Resources Institute, New Delhi, India, [5]College of Life Science, Inner Mongolia University, Hohhot, China, [6]School of Nature Conservation, Beijing Forestry University, Beijing, China, [7]College of Resources and Environmental Sciences, China Agricultural University, Beijing, China, [8]School of Life Sciences, Arizona State University, Tempe, Arizona, USA

Abstract Atmospheric acidic deposition has been a major environmental problem since the industrial revolution. However, our understanding of the effect of acidic deposition on soil pH is inconclusive. Here we examined temporal variations in topsoil pH and their relationships with atmospheric sulfur and nitrogen deposition across China's forests from the 1980s to the 2000s. To accomplish this goal, we conducted artificial neural network simulations using historical soil inventory data from the 1980s and a data set synthesized from literature published after 2000. Our results indicated that significant decreases in soil pH occurred in broadleaved forests, while minor changes were observed in coniferous and mixed coniferous and broadleaved forests. The magnitude of soil pH change was negatively correlated with atmospheric sulfur and nitrogen deposition. This relationship highlights the need for stringent measures that reduce sulfur and nitrogen emissions so as to maintain ecosystem structure and function.

1. Introduction

Fossil-fuel combustion and intensive agricultural fertilization have led to acidic deposition around the world. Although legislation aimed at reducing sulfur dioxide (SO_2) emissions [*Klimont et al.*, 2013] has reduced the rate of sulfur deposition in North America and Europe, anthropogenic SO_2 emissions in East Asia have continued to increase until 2006 [*X. K. Lu et al.*, 2010, *Klimont et al.*, 2013]. Meanwhile, emissions of reactive nitrogen from various anthropogenic sources have substantially increased since the industrial revolution, leading to an average atmospheric deposition rate of 105 Tg yr^{-1} at the global scale [*Dentener et al.*, 2006; *Galloway et al.*, 2008]. Acidic deposition can significantly alter soil chemical properties by lowering the soil pH [*M. Lu et al.*, 2011], often inducing the loss of exchangeable base cations (Ca^{2+}, Mg^{2+}, K^+, and Na^+) [*Lucas et al.*, 2011] and mobilizing exchangeable aluminum (Al^{3+}). Reduced availability of Ca^{2+} and Mg^{2+} in the soil decreases stress tolerance in various tree species, while increased availability of exchangeable Al^{3+} is detrimental to tree growth and ecosystem health [*Lieb et al.*, 2011]. These ecological impacts indicate that a profound understanding of soil pH change is crucial for evaluating the effect of acidic deposition on ecosystem structure and function [*Kirk et al.*, 2010] and can be important in developing control measures to improve soil quality [*Yang et al.*, 2012].

Soil pH dynamics have attracted significant attention in recent years, with extensive observations derived from the resurvey of previously sampled sites [e.g., *Lapenis et al.*, 2004; *Warby et al.*, 2009; *Hédl et al.*, 2011; *Yamada et al.*, 2012]. However, the results of these studies have been highly variable, ranging from significant decreases [*Lapenis et al.*, 2004; *Warby et al.*, 2009; *Bedison and Johnson*, 2010; *Hédl et al.*, 2011; *Yamada et al.*, 2012; *van der Heijden et al.*, 2013] to significant increases [*Kirk et al.*, 2010] in topsoil pH over the sampling interval. Further, some studies have reported no significant change in topsoil pH over the sampling period [*Hazlett et al.*, 2011; *McGovern et al.*, 2013]. These largely conflicting results suggest that more in-depth analyses of soil pH dynamics are necessary. Although it is evident that acidic deposition is very high in East Asia, soil pH dynamics have not been well studied in this region. It is therefore a priority to investigate long-term soil pH dynamics in East Asia [*Dentener et al*., 2006; *Klimont et al*., 2013] so as to develop mitigation strategies for maintaining soil productivity.

China is the most rapidly developing county in East Asia and the largest SO_2 emitter in the world while also suffering one of the highest levels of nitrogen deposition [*Pan et al*., 2013]. China is an ideal platform for exploring soil pH dynamics for several reasons. First, atmospheric sulfur [*Zhao et al*., 2009; *Z. Lu et al*., 2011] and nitrogen deposition [*Lü and Tian*, 2007; *Liu et al*., 2013] exhibit spatially distinct patterns across China, with higher deposition rates in the country's eastern regions. This large spatial gradient provides a natural platform to explore the relationship between soil pH dynamics and the strength of external acidic inputs. Second, soil conditions have been well documented in China, both historically and in present day, allowing for evaluation of soil pH change over sampling intervals. Historical records reach back to 1979 with the initiation of the National Soil Inventory, which described soil pH across the country from 1979 to 1989 [*National Soil Survey Office*, 1998]. More recently, soil pH has been reported in a number of individual studies that can be synthesized to generate a modern data set. Utilizing these data sets, two studies have described widespread increases in soil acidity across the major cropland [*Guo et al*., 2010] and grassland ecosystems [*Yang et al*., 2012] of China. However, a comprehensive analysis of temporal changes in soil acidity across forest ecosystems in China (~17% of total land area of the country) has not yet been undertaken.

In this study, we evaluated long-term variations in soil pH across major forest types in China from the 1980s to the 2000s. We utilized a data set synthesized from 240 data records arising from 130 articles published after 2000 and 247 soil profiles listed in the National Soil Inventory during the 1980s. The synthesized data set was then combined with related environmental parameters to construct an artificial neural network (ANN) to predict soil pH and its changes during the study period (1980s to 2000s). We further examined the relationship between soil pH change and external acidic deposition to test the hypothesis that external acidic inputs regulate soil pH dynamics.

2. Materials and Methods

2.1. Study Area

Forests cover 16.6% of the total land area in China. Major forested areas fall between latitudes 18.46°N and 52.97°N and longitudes 80.82°E and 133.60°E. The mean annual temperature (MAT) and mean annual precipitation (MAP) of forested areas during the 2000s were within the range of −4.3 to 25.1°C and 158.9 to 2142.2 mm, respectively. There are five major forest types in China: evergreen broadleaved forests, deciduous broadleaved forests, mixed coniferous and broadleaved forests, evergreen coniferous forests, and deciduous coniferous forests [*Chinese Academy of Sciences*, 2001].

2.2. Data Synthesis

Data from the National Soil Inventory of the 1980s [*National Soil Survey Office*, 1993, 1994a, 1994b, 1995a, 1995b, 1996], including the location for each soil profile and soil pH at different horizons, were used. Soil pH was measured potentiometrically using a pH meter in soil water suspension (air-dried samples and deionized water), with a soil:water ratio of 1:2.5 [*Bao*, 2000]; 247 soil profiles derived from the National Soil Inventory were used to represent the historical state of soil acidity (Figure S1 in the supporting information). These soil profiles covered all five of the major forest ecosystems across the country (n = 42, 58, 53, 81, and 13 for evergreen broadleaved forests, deciduous broadleaved forests, mixed coniferous and broadleaved forests, evergreen coniferous forests, and deciduous coniferous forests, respectively). These soil profiles have been widely used to explore the pattern of soil biogeochemistry at different soil horizon across China [*Xie et al*., 2007; *Yang et al*., 2007a, 2007b]. However, as topsoil is usually more sensitive to atmospheric acidic deposition than other horizons, we only used data from the upper horizon, with an average depth of 15 cm.

The 2000s data set was synthesized from 130 articles, ultimately resulting in a soil pH data set representative of forested areas across the country. Articles published after 2000 were collected from China Knowledge Resource Integrated (CNKI) database (http://www.cnki.net/) and Institute for Scientific Information (ISI) Web of Science database (http://apps.webofknowledge.com) using "soil pH" or "soil acidity" as key words. To assure data consistency, the following four criteria were used: (i) Only articles that reported soil pH in forest ecosystems were considered, (ii) for manipulation experiments only data from the "control" plots were used, (iii) soil pH must have been determined following the same methodology as the National Soil Inventory (i.e., measured potentiometrically, soil water suspension, air-dried soils, deionized water, and soil:water ratio of 1:2.5; notably, studies that use other soil:water ratios were excluded from this synthesis), and (iv) only the surface layer data in mineral soils were used. Soil depth was concentrated within the top 30 cm in most studies, with the average close to the historical record (14.4 versus 15 cm). To clarify whether depth variability induces significant shifts in soil pH, we compared soil pH of surface and subsurface layers using data derived from the National Soil Inventory. Paired *t* test analysis revealed that forest soil pH did not exhibit a significant difference between the two soil layers (average depth interval: 0–15 versus 15–45 cm, $P = 0.161$; Figure S2), demonstrating that relatively small variability in soil depth would not induce large bias in subsequent analyses.

Based on the above-mentioned criteria, 240 forest soil pH records were synthesized from the literature (Figure S1 and Text S1). Similar to other data syntheses [e.g., *Liu et al*., 2013], it was difficult to obtain a random distribution of the synthesized sites across the study area. However, the synthesized data covered all five major forest types found in the study area ($n = 54$, 83, 36, 59, and 8 for evergreen broadleaved forests, deciduous broadleaved forests, mixed coniferous and broadleaved forests, evergreen coniferous forests, and deciduous coniferous forests, respectively). Interestingly, a marginally significant correlation was observed between the sample size and the area proportion covered by each of five major forest types ($r = 0.82$, $P < 0.1$), indicating that replicate numbers approximately reflect forest coverage across the study area. Again, similar to other data syntheses [e.g., *Bond-Lamberty and Thomson*, 2010], our synthesized measurements did not cover the entire decade equally (Figure S3a), with 48.8% of observations occurring in the late 2000s (2008–2010) and 7.5% of observations occurring in the early 2000s (2000–2002). However, the frequency distribution of the sampling year exhibited similar patterns between broadleaved (Figure S3b) and coniferous forests (Figure S3c), demonstrating that a potential regional bias in available soil data will not have a profound effect on subsequent model prediction.

Along with the target variable (soil pH), we synthesized other geographic, climatic, and vegetation parameters not documented in the National Soil Inventory or collated literature. Site names recorded in the National Soil Inventory were converted to geographic locations (longitude and latitude) using a digital map developed by the National Administration of Surveying, Mapping and Geoinformation (http://www.tianditu.cn/map/index.html). Based on these longitude/latitude coordinates, altitude was obtained from a digital elevation model (http://data.geocomm.com/dem/). The decadal average of MAT and MAP from 1979 to 1989 and 2000 to 2010 were extracted from a national climate data set (at a $0.1°\times0.1°$ resolution) (National Meteorological Information Center of China Meteorological Administration, http://www.nmic.gov.cn). Following vegetation descriptions in original publications, and in consultation with the vegetation atlas of China (scale: 1:1,000,000) [*Chinese Academy of Sciences*, 2001], the vegetation for each site was classified as one of the following groups: evergreen broadleaved forests, deciduous broadleaved forests, mixed coniferous and broadleaved forests, evergreen coniferous forests, and deciduous coniferous forests. A monthly normalized difference vegetation index (NDVI) data set was developed from the bimonthly NDVI during 1982–1989 and 2000–2010 (GIMMS NOAA/AVHRR NDVI composites; resolution: $0.083°\times 0.083°$) [*Tucker et al*., 2005] using the maximum value composition method [*Holben*, 1986]. This monthly NDVI data set was then used to calculate annual NDVI, which were then aggregated to grid cells of $0.1° \times 0.1°$. Finally, wet nitrogen deposition during 1979–1989 and 2000–2010 was estimated from the corresponding MAP using an empirical function ($y = 3.6281\exp^{0.001x}$, $r^2 = 0.22$, $P < 0.05$), which was developed using a data set synthesized by *Liu et al*. [2013]. Similarly, wet sulfur deposition during the two periods was predicted from MAP on the basis of a regression relationship ($y = 8.90 + 0.02x$, $r^2 = 0.23$, $P < 0.05$) reported by *Pan et al*. [2013].

2.3. ANN Simulation

ANN is an information processing paradigm that is inspired by biological nervous systems [*Hopfield*, 1982]. It is composed of a set of highly interconnected processing neurons. Each neuron receives a weighted set of inputs and responds with an output. The ANN is usually trained with a training data set, which adjusts the weight of the synaptic connections between neurons. Once trained, the ANN can be used to simulate complex relationships between inputs and outputs.

ANN has been widely used to predict some target variable using a set of environmental parameters [e.g., *Papale and Valentini*, 2003; *Beer et al*., 2010; *Yang et al*., 2014]. The ANN could simulate nonlinear relationships between the target variable and environmental parameters and might be more powerful for regional soil mapping [*Grimm et al*., 2008]. It has been demonstrated that ANN has a higher predictive capacity in regional soil mapping compared with multiple linear regression and regression kriging [*Li et al*., 2013]. As such, we adopted a feed-forward back propagation neural network (BPNN) to estimate soil pH across forested areas using related environmental variables. Using this BPNN, we predicted soil pH at sites without data during the 1980s (and 2000s) using sites with observations during the same period.

The architecture of a BPNN includes an input layer, a hidden layer, and an output layer (Figure S4). Given that topsoil pH exhibited systematic changes along geographic (Figure S5) and environmental gradients (Figure S6), seven variables (latitude, longitude, altitude, MAT, MAP, NDVI, and vegetation type) were included as input variables during BPNN analysis. Notably, the corresponding climate and NDVI data sets were involved in model simulation during the two periods. Both input and output variables were normalized between -1 and $+1$ before network training to avoid overfitting. The entire data set from each period was divided into three groups during network training: (a) training data set (70% of total data sets used to determine weights in the neural network), (b) validation data set (15% of total data sets used to calculate errors to avoid overtraining), and (c) testing data set (15% of total data sets used to evaluate the performance of the neural network) [*Papale and Valentini*, 2003; *He et al.*, 2006]. Both Pearson correlation coefficient and root-mean-squared error (RMSE) were calculated to evaluate errors produced during training, validation, and testing processes. The lowest error during both sampling periods was obtained with 11 neurons in the hidden layer. After network training, the constructed BPNN was used to predict soil pH using environmental variables (i.e., longitude, latitude, altitude, MAT, MAP, NDVI, and vegetation type). Finally, the predicted output ranging between -1 and 1 was rescaled to the original unit of soil pH (i.e., from 3 to 9), using the *mapminmax* function in MATLAB (the MathWorks, Natick, MA, USA). To obtain a robust estimation, we performed 5000 model simulations with bootstrapping methods before adopting the average prediction. ANN analyses were conducted using MATLAB (the MathWorks, Natick, MA, USA).

2.4. Statistical Analyses

Based on ANN simulation, a data set of pH values (either measured or simulated) at all sites was created for both sampling periods. A paired *t* test was then performed to detect statistical differences in soil pH between the 1980s and the 2000s. Using measured data alone, we conducted an unpaired *t* test to examine soil pH dynamics in four of the five major forest types (excluding deciduous coniferous forests due to the small sample size; $n = 13$ and 8 during the 1980s and 2000s, respectively). Ordinary least-squares regression was used to examine the effects of atmospheric acidic deposition (sulfur and nitrogen deposition) on soil pH dynamics. To further examine whether a real relationship exists between soil pH change and acidic deposition, we conducted partial correlation analysis using either longitude or latitude as a controlling factor. Statistically, the partial correlation between soil pH change (y) and acidic deposition (x) given a controlling variable of longitude or latitude (z) is the correlation between the residuals R_x and R_y derived from the two linear regressions of x with z and of y with z, respectively. It represents the correlation between soil pH change and acidic deposition after the influence of longitude or latitude has been removed from both soil pH change and acidic deposition. All statistical analyses were performed using the R software package [*R Development Core Team*, 2012].

3. Results and Discussion

3.1. ANN-Based Estimation

RMSE values comparing predicted and measured soil pH for training, validation, and testing data sets during the 1980s were 0.34, 0.66, and 0.52, respectively (Figures 1a–1c). This exhibits linear relationships between predicted and measured soil pH. Moreover, Pearson correlation coefficients (r) comparing predicted and measured values for the 1980s were 0.96, 0.86, and 0.92 for the training, validation, and testing data sets, respectively. Similarly, predicted soil pH for the 2000s were linearly associated with related measurements, and r values comparing predicted and measured values were 0.95, 0.90, and 0.91 for the training, validation, and testing data sets, respectively (Figures 1d–1f). Moreover, data points occurred near the 1:1 line, with an RMSE of 0.35, 0.56, and 0.52 for the training, validation, and testing data sets, respectively. These results demonstrated that the ANN approach provided reliable predictions of soil acidity for both study periods. To further illustrate this point, we conducted a Monte Carlo analysis to evaluate ANN-based predictions. Specifically, we extracted soil pH and related environmental variables with bootstrapping techniques and used the selected data set to train the ANN model. We then predicted soil acidity for those sites without actual measurements. These analyses were repeated 5000 times and testing results were summarized for

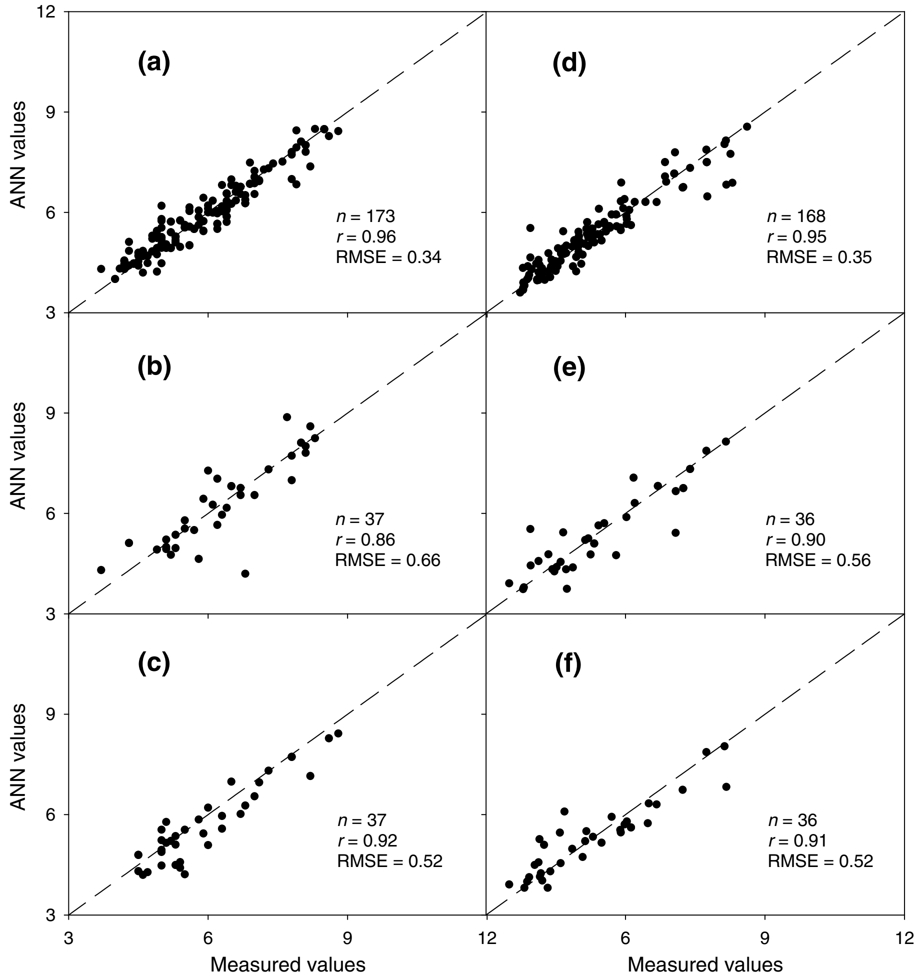

Figure 1. Predicted versus measured soil pH during the training, validation, and testing processes for the two time periods. (a, d) Training data set, (b, e) validation data set, and (c, f) testing data set.

each run. Our analyses revealed that RMSE was relatively low in most cases across five major forest types, with the average ranging from 0.56 in deciduous coniferous forests to 0.75 in evergreen broadleaved forests during the 1980s and from 0.68 in mixed coniferous and broadleaved forests to 0.79 in evergreen coniferous forests during the 2000s (Figure S7). The overall RMSE across all forest types was estimated at 0.69 and 0.75 during the 1980s and the 2000s, respectively, confirming the reliability of the ANN approach. In addition, predicted soil pH during the 2000s were positively correlated with the corresponding values during the 1980s (r = 0.62, $P < 0.05$; Figure 2a). This provides indirect evidence that the ANN approach generated reasonable predictions during both sampling periods.

3.2. Soil pH Dynamics in the Broadleaved and Coniferous Forests

Comparisons of soil pH between sampling periods revealed substantial differences among forest types (Figure 2b). Despite of higher pH and greater pH range at some sites during the 2000s, soil pH in evergreen and deciduous broadleaved forests during the 2000s was significantly lower than during the 1980s (n = 96 and 141, respectively; $P < 0.05$), while there were no significant differences in the other three forest types (n = 89, P = 0.34 for mixed coniferous and broadleaved forests; n = 140, P = 0.39 for evergreen coniferous forests; n = 21, P = 0.34 for deciduous coniferous forests). Interestingly, the magnitude of the decrease in soil pH recorded in evergreen forests (−0.6) was within the range of pH change observed across China's major croplands (−0.8 to −0.3) [*Guo et al.*, 2010] and grassland ecosystems (−0.8 to −0.4) [*Yang et al.*, 2012]. However, the magnitude of the decrease in soil pH across deciduous broadleaved forests (−0.2) was smaller than those reported in other ecosystems. Unpaired t tests of the change in pH (testing for significant differences from 0 trend) also

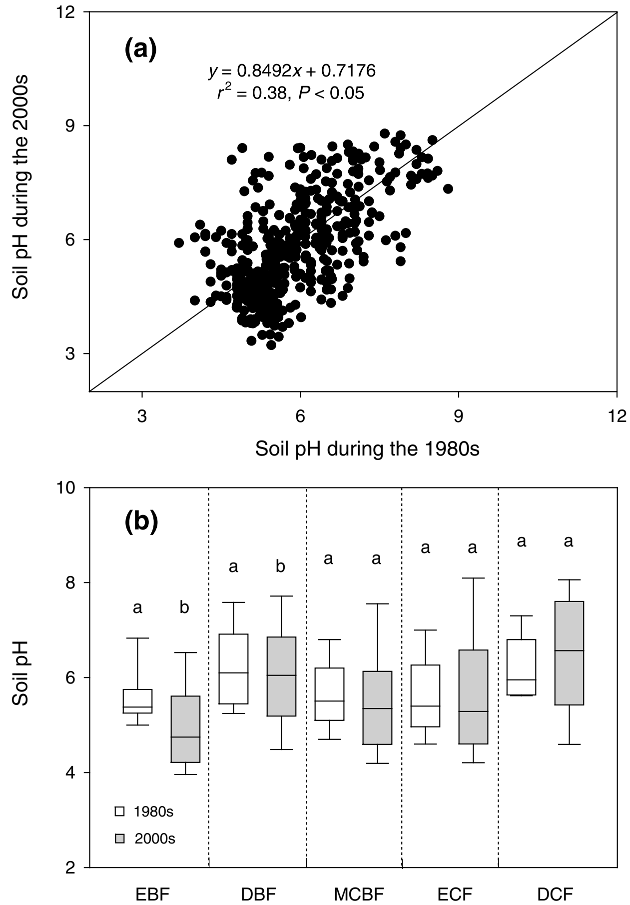

Figure 2. Comparison of (a) soil pH between the 1980s and the 2000s and (b) box-and-whisker plots showing soil pH changes across various forest types during the monitoring period. The line in Figure 2a is a 1:1 line. Different letters above each box in Figure 2b denote significant differences between the two periods. EBF: evergreen broadleaved forests, DBF: deciduous broadleaved forests, MCBF: mixed coniferous and broadleaved forests, ECF: evergreen coniferous forests, and DCF: deciduous coniferous forests.

confirmed the results of the paired *t* tests, showing a significant decrease for evergreen and deciduous broadleaved forests, but no significant change for evergreen coniferous forests (Figure S8). Nevertheless, paired *t* test analysis should provide more reliable results as it considers data generated by the ANN simulation.

Soil pH decreased significantly in broadleaved forests; however, the decrease was not significant for coniferous forests. This difference could be driven by differences in wet acidic deposition in these two forest types (24.5–36.1 versus 18.5–30.7 kg S ha^{-1} and 8.6–15.0 versus 6.1–12.0 kg N ha^{-1} during 1980s–2000s). To test this hypothesis, we examined the relationship between the magnitude of soil pH change and wet sulfur/nitrogen deposition during 1980s–2000s. Our analysis indicated that the magnitude of soil pH change was negatively correlated with the strength of atmospheric sulfur deposition ($r = -0.37$, $P < 0.05$; Figure 3a) and nitrogen deposition across the study area ($r = -0.37$, $P < 0.05$; Figure 3b), suggesting that differences in atmospheric acidic deposition can partially explain different patterns of soil pH dynamics observed across forest types. To further explore whether the relationship between soil pH change and acidic deposition is simply a spatial artifact, we examined the partial correlations between these variables using either longitude or latitude as a controlling factor. Our results showed that the partial correlations between soil pH change and sulfur deposition and between soil pH change and nitrogen deposition were still significant ($P < 0.05$). Overall, these analyses demonstrate that acidic deposition modifies the magnitude of soil pH change.

Dry acidic deposition including both sulfur and nitrogen deposition could also contribute to soil acidification. It has been reported that dry deposition is generally high in China. For instance, *Pan et al.* [2013] demonstrated that both particulate and gaseous dry sulfur deposition accounted for 68% of total sulfur deposition across 10 sites in northern China. Similarly, a national-scale synthesis by *Lü and Tian* [2007] reported that the proportion of dry deposition ranged from 9.6% to 53.6% (averaging 23.5%) of total nitrogen deposition across the country. Undoubtedly, these relatively high levels of dry acidic deposition could stimulate acidification processes in forest soils [*Larssen et al.*, 2006; *Zhao et al.*, 2009; [*X. K. Lu et al.*, 2010]. However, it is challenging to quantify the contribution of dry acidic deposition on regional soil pH dynamics due to the paucity of spatially explicit dry acidic deposition data across the country.

Finally, soil acidification in broadleaved forests could be due to reduced soil exchangeable base cations. It has been widely reported that forest growth in China has accelerated over past several decades [*Fang et al.*, 2014]. This enhanced growth could have led to sequestration of soil base cations in woody biomass. Heavy precipitation events have also become more frequent across the country [*Zhai et al.*, 2005]. The increased frequency of high precipitation could stimulate leaching of base cations from the surface soil. Together, enhanced uptake by plants and increases in the frequency of high precipitation could reduce soil base cations

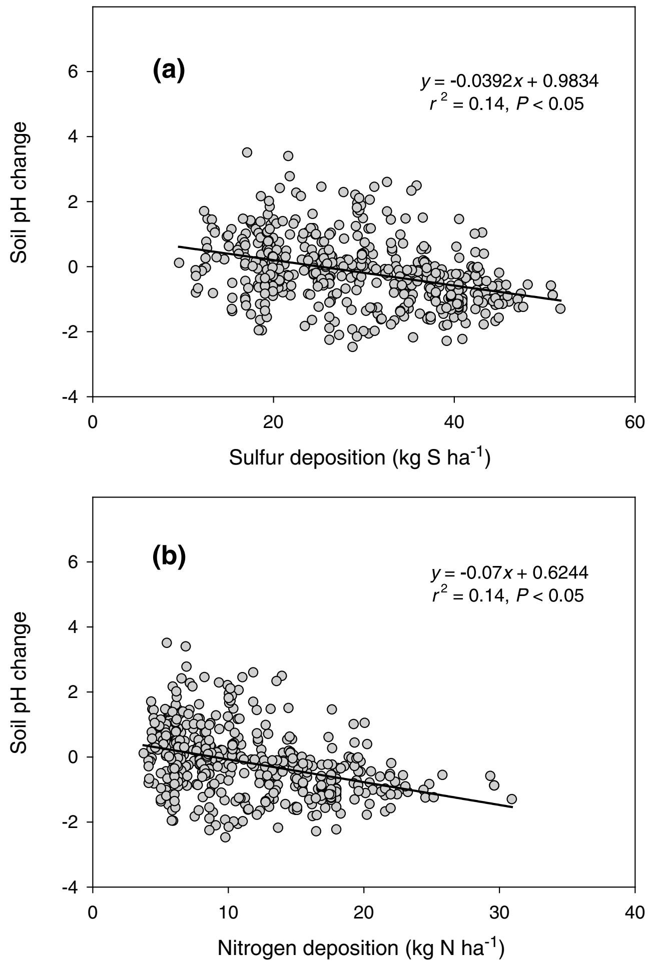

Figure 3. (a and b) Soil pH dynamics versus atmospheric acidic deposition across forested areas in China.

in broadleaved forests [*Guo et al*., 2010], weakening the buffering capacities of the soil and making the soil more sensitive to external acidic deposition. In addition, elevated base cations leaching under acidic deposition may also be a significant driver of soil acidification observed in broadleaved forests.

3.3. Scientific and Policy Implications

Our findings have important implications for understanding the potential effects of acidic deposition on forest ecosystems. The acidification of forest soils observed in this study together with those reported from cropland [*Guo et al*., 2010] and grassland ecosystems [*Yang et al*., 2012] in China indicates that atmospheric acidic deposition has already induced negative impacts on all of the major ecosystems of the country. This acidification may lead to serious ecological consequences such as species losses [*Z. Lu et al*., 2010] and productivity reduction [*Mo et al*., 2008]. Moreover, the mobilization of Al^{3+} ions in the soil due to buffering processes may exert toxic effects on tree growth and ecosystem services in forest ecosystems [*Lieb et al*., 2011]. Further studies should be conducted to clarify the dynamics of soil base cations and exchangeable Al^{3+} across forest ecosystems. This study also implies urgency in establishing measures aimed at improving environmental quality and retarding the adverse effects of soil acidification on ecosystem structure and function. It is essential to reduce industrial SO_2 emissions by continuously implementing flue gas desulfurization and phasing out inefficient units [*Larssen et al*., 2006; *X. K. Lu et al*., 2010]. Additionally, along with measures that reduce SO_2 emissions, it is important to control industrial NO_x emissions by using low NO_x burners or installing selective catalytic reduction systems in power plants, while also reducing agricultural NH_3 emissions through long-term changes in farming practices [*Zhao et al*., 2009].

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