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The contribution of atmospheric deposition and forest harvesting to forest soil acidification in China since 1980

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HIGHLIGHTS

- Forest soil pH in China decreased by 0.36 units on average in the periods between 1981–1985 and 2006–2010.
- The strongest acidification occurred in southwest China and/or semi-Luvisols.
- Atmospheric deposition contributed 84% to the H^+ production causing forest soil acidification.
- H^+ production induced by forest growth contributed 16% to the forest soil acidification.
- On average, base cation deposition neutralized 31% of the potential acid input.

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ABSTRACT

Soils below croplands and grasslands have acidified significantly in China since the 1980s in terms of pH decline in response to acid inputs caused by intensified fertilizer application and/or acid deposition. However, it is unclear what the rate is of pH decline of forest soils in China in response to enhanced acid deposition and wood production over the same period. We therefore gathered soil pH data from the Second National Soil Inventory of China and publications from the China National Knowledge Infrastructure (CNKI) database in 1981–1985 and 2006–2010, respectively, to evaluate the long-term change of pH values in forest soils. We found that soil pH decreased on average by 0.36 units in the period 1981–1985 to 2006–2010., with most serious pH decline occurring in southwest China (0.63 pH units). The soil type with the strongest pH decline was the semi-Luvisol (0.44 pH units). The decrease in pH was significantly correlated with the acid input induced by atmospheric deposition and forest harvesting. On average, the contribution of atmospheric deposition to the total acid input was estimated at 84% whereas element uptake (due to forest wood growth and harvest) contributed 16% only. Atmospheric deposition is thus the major driver for the significant forest soil acidification across China.

1. Introduction

Since the Second Industrial Revolution, the world has got into a period of booming industrial production, with elevated emissions of acidifying compounds, notably sulphur dioxide (SO_2), nitrogen oxides (NO_X) and ammonia (NH_x), to the atmosphere (Carmichael et al., 2002; Mylona, 1996; O'Neil et al., 2012). Subsequently, a series of environmental problems occurred (Dickerson et al., 1997; Haines et al., 1980; O'Neil et al., 2012), one of which was soil acidification (Van Breemen et al., 1982). Much research has been carried out on acidification of non-agricultural (forest) soils since the 1980s, especially in Europe (Van Breemen et al., 1984) and the US (Johnson et al., 1982) in response to deposition of SO_2, NO_X and NH_3, recently being summarized in De Vries et al. (2015). As a consequence, international co-operation to combat the emissions of those pollutants has been carried out. Such reductions have

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occurred in Europe since 1975 for SO_2 (Mylona, 1996) and since 1990 for NO_x (Vestreng et al., 2009) and to a lesser extent for NH_3 (Waldner et al., 2014; www.EMEP.int), which was accompanied by the slow soil recovery. For instance, Kirk et al. (2010) found that soil pH increased between 1978 and 2003 in Wales and England in response to decreased acid deposition.

Unlike Europe, SO_2 and NO_x emissions have largely increased in China since 1980 (Smith et al., 2011), causing a high increase in SO_2 and NO_x deposition (Duan, 2000; Liu et al., 2013), leading to enhanced soil acidification (Liu et al., 2010). In recent decades, China became one of the countries with the most elevated acid deposition rates due to increasing emissions of sulphur (S) and nitrogen (N) compounds (Duan, 2000; Liu et al., 2013; Pan et al., 2013; Du et al., 2014, 2015). Though S deposition is still among the highest in the world, it has decreased in China since 2005 because of the implementation of several policies to reduce SO_2 emission (Fang et al., 2013). However, N emissions continue to rise due to intensive cultivation and livestock production, as well as traffic and industrial development (Pan et al., 2013). This has led to a decline in ratios of sulphate (SO_4^{2-}) to nitrate (NO_3^-) concentrations in wet deposition (Fang et al., 2011; Wang et al., 2012). In China, reduced-N (NH_3 and NH_4^+) plays a more important role than oxidized-N (NO_x and NO_3^-) deposition (Larssen et al., 2011; Du et al., 2014). Both the high rates and composition shifts of acid deposition may have significantly affected soil acidification in China (Liu et al., 2010; Yang et al., 2015).

Although S (SO_2 and SO_4^{2-}) and N (NH_x and NO_x) deposition have received considerable attention (Bowman and Cleveland, 2008), the important role of base cation (BC, including calcium (Ca^{2+}), magnesium (Mg^{2+}), potassium (K^+) and sodium (Na^+) ions) deposition should not be neglected (Larssen and Carmichael, 2000), since the potential acid input equals the sum of S and N deposition corrected for base cation deposition. Furthermore, forest felling for timber use, leading to removal of BC from the soil, further accelerates soil acidification (Rowell and Wild, 1985). Significant site-specific soil acidification in Chinese forests has been reported previously by many researchers (e.g. Liu et al., 2010; Huang et al., 2014).

Soil acidification is mostly defined as a decrease in soil pH (e.g. Krug and Frink, 1983), but production of protons in the soil is only partially reflected in changes of the pH, being an intensity factor. For this reason, Van Breemen et al. (1983) defined soil acidification as a decrease in the acid-neutralizing capacity (ANC) of the soil, being a capacity factor. Whether changes in the ANC, which is due to net acid production in the soil, is reflected in observable changes in soil acidity (pH) depends on the buffering rate of the soil due to weathering and cation exchange (Ulrich, 1981; De Vries et al., 1989). Simultaneous information on both acid production, causing a change in ANC, and pH changes thus give information on both soil acidification and the sensitivity of the soil to acidification.

Large scale decline in soil pH has been demonstrated for both croplands (Guo et al., 2010) and natural grasslands (Yang et al., 2012) in China, during the 1980s and 2000s, in response to acid production cause by enhanced N fertilization and/or acid deposition, respectively. Nevertheless, very limited information is available on large scale pH changes in Chinese forest soils over the last two or three decades in response to acid production cause by forest management and acid deposition. Recently, a paper on regional forest acidification has been published, showing significant soil pH decrease only in broadleaved forests across China (Yang et al., 2015). However, their research was still based on a limited dataset (487 samples) and no evaluation was made of the linkage between pH change and proton (acid) production by acid deposition and forest growth nor with the sensitivity of soils to acidification.

In this paper we further evaluated pH changes in forest soils over China from 1981–1985 to 2006–2010. The evaluation was based on a very large dataset (5598 samples), allocated to six regions and five soil clusters, with different sensitivities to acid inputs, respectively. The observed pH decline was evaluated for those soil clusters and regions, by calculating the acid inputs caused by SO_2, NO_X and NH_x deposition and forest uptake in different regions. Base cation deposition as well as the fate of N were accounted for to assess the net proton production and related this to the pH decrease in Chinese forests. The assessment of acid inputs by deposition and forestry growth was conducted to gain insight in the relative contribution of these drivers to soil pH decrease, which provides valuable information for predicting the future forest acidification trend.

2. Materials and methods

Soil pH datasets were collected to assess pH changes in six major sub-regions and among five major soil types in China using an unpaired t-test (2.1). To gain insight in the main sources of soil acidification in various regions, an assessment was then made to calculate the proton production (H^+) from deposition (2.2) and forest growth (2.3) in each region. The pH change in each region was evaluated and compared with the proton production driving these changes.

2.1. Evaluation of soil pH changes by unpaired t-tests

Approach: In assessing pH from 1981–1985 to 2006–2010, we focused on pH changes in the top 30 cm of the forest soils in six major sub-regions according to Fang et al. (2001), i.e. East, North, Northeast, Northwest, South central and Southwest (Fig. 1). We used an unpaired t-test to evaluate the significance in changes in the soil pH status between the early 1980s (1981–1985) and the late 2000s (2006–2010) across China, since pH data in the two periods are not paired but have different locations and numbers of measurements. The analysis was made after testing that the pH values in the two periods were normally distributed.

Soil groups were clustered according to their soil genetic classification (Shi et al., 2004; Zhao and Shi, 2007), as well as their sensitivity to acidification (Van Breemen et al., 1983; De Vries et al., 1989). Soil types were distinguished into 5 clusters, i.e., Ferralsols, Luvisols, Semi-Luvisols, Calcareous soils and Other Soils.

(i) Ferralsols include highly weathered acidic red soils, yellow soils and latosols, predominantly distributed in low latitudes and humid tropical and subtropical zones (e.g. central and south part of China). These soils have mostly a low pH, a low cation exchange capacity (CEC) and a low base saturation (BS) implying that these soils are in the aluminium (Al) buffer range with a limited sensitivity to pH change.

(ii) Luvisols include slightly acidic dark brown soils, yellow brown soils, brown soils, Albic soils and yellow-cinnamon soils. These soils mainly occur in the eastern humid monsoon climate zone (e.g. east China), and have high organic carbon contents and slightly acidic to neutral pH values, implying that these soils are in the BC exchange buffer range, being sensitive to acidification.

(iii) Semi-Luvisols include black soils, grey soils and cinnamon soils, mainly occurring in semi-humid and semi-arid zones (e.g. north China). These soils do often have carbonate ($CaCO_3$) in the subsoil, but the topsoil is mostly in the BC exchange buffer range, being sensitive to acidification.

(iv) Calcareous soils include desert soils, calcium soils, dark loessial soils and alkali-saline soils, mostly occurring in semi-arid and arid zones, for instance in Northwest and North