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RESEARCH ARTICLE

Long-term tobacco plantation induces soil acidification and soil base cation loss

Yuting Zhang[1,2] · Xinhua He[3,4] · Hong Liang[5] · Jian Zhao[7] · Yueqiang Zhang[1,2] ·
Chen Xu[6] · Xiaojun Shi[1,2]

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Abstract Changes in soil exchangeable cations relative to soil acidification are less studied particularly under long-term cash crop plantation. This study investigated soil acidification in an Ali-Periudic Argosols after 10-year (2002–2012) long-term continuous tobacco plantation. Soils were respectively sampled at 1933 and 2143 sites in 2002 and 2012 (also 647 tobacco plants), from seven tobacco plantation counties in the Chongqing Municipal City, southwest China. After 10-year continuous tobacco plantation, a substantial acidification was evidenced by an average decrease of 0.20 soil pH unit with a substantial increase of soil sites toward the acidic status, especially those pH ranging from 4.5 to 5.5, whereas 1.93 kmol H^+ production ha^{-1} $year^{-1}$ was mostly derived from nitrogen (N) fertilizer input and plant N uptake output. After 1 decade, an average decrease of 27.6 % total exchangeable base cations or of 0.20 pH unit occurred in all seven tobacco plantation counties. Meanwhile, for one unit pH decrease, 40.3 and 28.3 mmol base cations kg^{-1} soil were consumed in 2002 and 2012, respectively. Furthermore, the aboveground tobacco biomass harvest removed 339.23 kg base cations ha^{-1} $year^{-1}$ from soil, which was 7.57 times higher than the anions removal, leading to a 12.52 kmol H^+ production ha^{-1} $year^{-1}$ as the main reason inducing soil acidification. Overall, our results showed that long-term tobacco plantation not only stimulated soil acidification but also decreased soil acid-buffering capacity, resulting in negative effects on sustainable soil uses. On the other hand, our results addressed the importance of a continuous monitoring of soil pH changes in tobacco plantation sites, which would enhance our understanding of soil fertility of health in this region.

Keywords Acid-buffering capacity · Ali-Periudic Argosols · Exchangeable base cations · *Nicotiana tabacum* · H^+ budget · Soil pH

Introduction

Soil acidification is a slowly natural process during soil formation and development (Van Breemen et al. 1984), and ~40 to 70 % of the world's arable land are affected by soil acidification (Rengel 2003), which leads to a significant reduction of food supply and sustainable soil use. Meanwhile, soil acidification has been recently accelerated due to acid deposition (Rengel 2003; Hoegberg et al. 2006), excessive ammonium-based fertilization (Guo et al. 2010; Lucas et al. 2011; Han et al. 2015; Tian and Niu 2015), and the mode of land use and plant growth (Jackson et al. 2005; Yang et al. 2012; Rice and Herman 2012).

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✉ Chen Xu
xuchen@cq.tobaccco.cn

✉ Xiaojun Shi
shixj@swu.edu.cn

[1] College of Resources and Environment, Southwest University, Chongqing 400716, China

[2] National Monitoring Station of Soil Fertility and Fertilizer Efficiency on Purple Soils, Chongqing 400716, China

[3] Centre of Excellence for Soil Biology, College of Resources and Environment, Southwest University, Chongqing 400716, China

[4] Department of Environmental Sciences, University of Sydney, Eveleigh NSW 2015, Australia

[5] College of Agriculture, Guizhou University, Guiyang 550025, China

[6] Tobacco Scientific Research Institute, Chongqing Tobacco Company, Chongqing 400716, China

[7] Technology Research Centre, Zunyi Tobacco Company, Zunyi 563000, Guizhou, China

Soil management practices can induce negative effects on soil properties, especially soil pH (Bortoluzzi et al. 2012). For instance, an accelerate soil acidification had resulted from the plantation of leguminous *Stylosanthes* species in Australian pastures (Noble et al. 2002) and in northeast Thailand's cropping systems (Lesturgez et al. 2006). A possible mechanism could be that a large quantity of soil Ca^{2+}, Mg^{2+}, and K^+ cations had been absorbed by such leguminous plants, leading to a proton (H^+) release from roots (Rengel 2003; Lesturgez et al. 2006). In the Chinese-intensive agricultural systems, the decrease of soil pH ranged from 0.30 to 0.80 U in the cash crop (vegetables, fruit, trees, tea, etc.) systems and from 0.13 to 0.76 U in the cereal or fiber crop (wheat, maize, rice, cotton, etc.) systems (Guo et al. 2010). On a tea plantation locating in eastern of China, soil acidification even occurred within 70-cm depth with a maximum 2.80 ΔpH change in the upper 17-cm depth (Alekseeva et al. 2011).

The change of land use also has an important effect on soil acidification. For instance, a global meta-analysis of 153 plantation types found that the conversion of non-forested lands to forests resulted in significant decreases of pH, Ca^{2+}, Mg^{2+}, and K^+ cations (Berthrong et al. 2009). In South America, the change from grassland to afforestation with fast growing exotic species (*Eucalyptus* sp.) resulted in the loss of base cations together with soil acidification (Céspedes-Payret et al. 2012). The continuous substantial mass loss of soil base cations induced by tree harvesting was the main reason inducing soil acidification (McDonnell et al. 2013).

Tobacco (*Nicotiana tabacum*) is an important cash crop in China, particularly in the Chongqing Municipal City, where it has an annual 45,000-ha tobacco plantation with 80,000 t of leaf production (CNBS 2012). Research on tobacco has focused on the harmful aspect of nicotine to human health, while currently limited information is available if the tobacco plantation could result in soil acidification. The objectives of the present study were therefore to address possible changes in soil exchangeable cations and pH after 10-year (2002–2012) long-term tobacco plantation from previous grain crop farmlands in southwest China and then to relate soil acidification to management strategies for sustainable soil utilization.

Materials and methods

Site description

The study site is located in seven metropolitan counties of Chongqing Municipal City (107° 24′–110° 11′ E, 28° 22′–31° 25′ N, 800–1200 m above the sea level), southwest China, where it has a subtropical climate. The mean annual highest and lowest temperature is 17.1 and 8.1 °C, respectively. The average annual rainfall is approximately 1100 mm, and frost-free period is about 230 days. The soil is classified as Ali-Periudic Argosols in the USDA Soil Taxonomic System. The faming system is a continuous tobacco plantation between May and October, while the land is fallow between December and March. In a ratio of nitrogen (N) (78 kg N ha^{-1})/P_2O_5/K_2O=8:12:25, a N/P/K compound fertilizer was applied 10–15 days before tobacco transplant, with a ratio of NH_4^+-N/NO_3^--N=7:3. The other two N (all in NO_3^--N)/P/K compound fertilizers were separately applied in a ratio of N (15 kg N ha^{-1})/P_2O_5/K_2O=20:15:10 and N (33 kg N ha^{-1})/P_2O_5/K_2O=13.5:0:44.5 at 7–10 and 30–40 days after tobacco transplant. Therefore, the N input (N fertilizer application) was 54.6 kg NH_4^+-N (3.9 kmol N) and 71.4 kg NO_3^--N (5.1 kmol N)ha^{-1} yr^{-1}, respectively. In addition, in 2010 and 2011, 1500-kg lime (CaO 80.8 % and MgO 4.2 %)ha^{-1} was annually applied at Fengdu (FD), Shizhu (SZ), and Wulong (WL).

Soil collection and analysis procedures

In 2012, the tobacco plantation areas were ~45,000 ha in the Chongqing Municipal City, which accounted for ~4.0 % of the whole tobacco plantation in China (CNBS 2012). With a 75-mm diameter auger, 20 soil cores (0–20-cm depth) from an area of ~20 ha were collected as one composite sample. From these seven tobacco plantation counties, a total of 1933 and 2143 soil samples from the same plantation field sites were then collected in March–April 2002 and March–April 2012, respectively (Fig. 1). After removing all visible debris, soils were air dried and then sieved (1 mm) for chemical analyses. Soil pH was determined using a glass electrode with a ratio of soil/water=1:2.5. Soil exchangeable K^+, Ca^{2+}, and Mg^{2+} cations were extracted with 1.0 M ammonium acetate, while exchangeable Ca^{2+} and Mg^{2+} cations were measured with a flame atomic absorption spectroscopy and exchangeable K^+ with a flame photometry (Lu 2000).

Tobacco plant sampling

From the seven tobacco plantation counties, a total of 647 whole tobacco plant samples were collected (Fengdu: FD, 31; Pengshui: PS, 152; Qianjinag: QJ, 75; Shizhu: SZ, 90; Wulong: WL, 60; Wushan: WS, 121; and Youynag: YY, 118) between July and September 2012 when tobacco plants were well matured. Plants were separated as leaves, stems, and roots, and oven dried at 70 °C. Their dry biomass was respectively recorded and ground into powder. Followed by a microwave extraction method, concentrations of K^+, Ca^{2+}, and Mg^{2+} cations and $H_2PO_4^-$, SO_4^{2-}, and Cl^- anions were measured with a flame atomic absorption spectroscopy and an ion chromatography (Lu 2000).

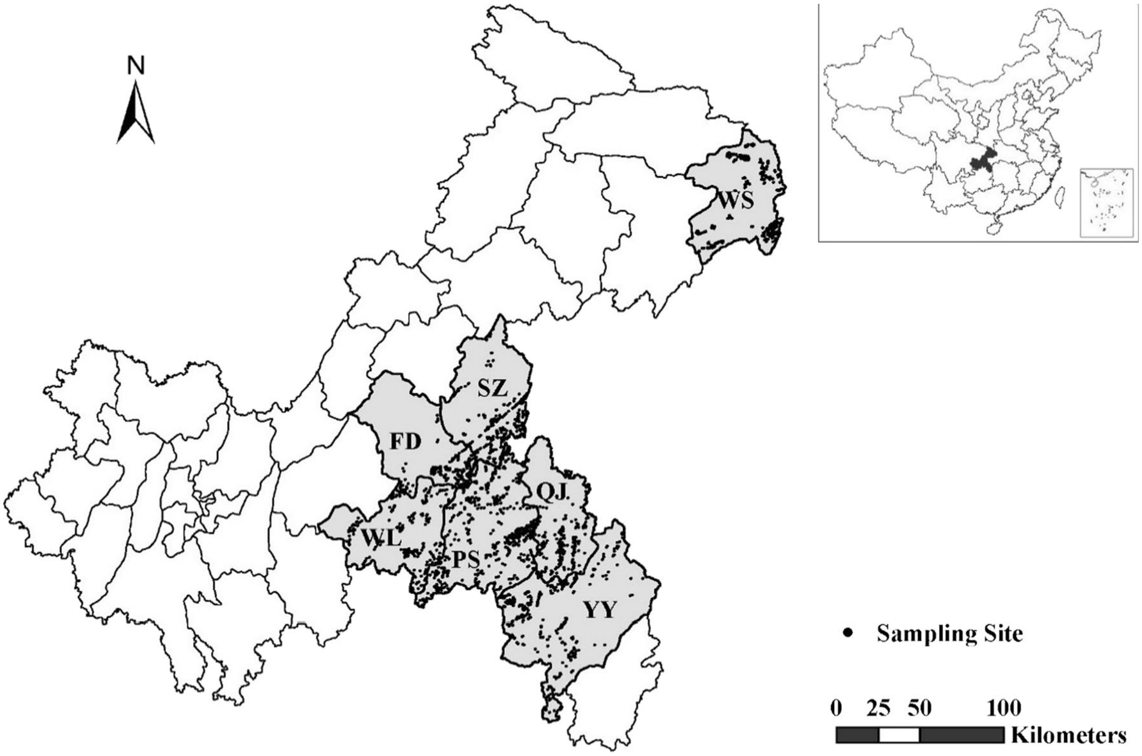


Fig. 1 Tobacco plantation sites at seven counties in Chongqing Municipal City, southwest China. Fengdu (FD, 111 and 137 samples in 2002 and 2012, respectively; hereafter the same), Pengshui (PS, 474 and 513), Qianjiang (QJ, 208 and 224), Shizhu (SZ, 275 and 286), Wulong (WL, 227 and 216), Wushan (WS, 152 and 385), and Youyang (YY, 486 and 382)

Estimation of H^+ budget

The contribution of fertilizer nitrogen (N) to soil acidification mainly includes nitrification of NH_4^+ to NO_3^- and leaching of NO_3^- from soil, while the uptake of NO_3^- makes soil more alkaline (Rengel 2003; Posch and Reinds 2009). Meanwhile, the amounts of base cations (K^+, Mg^{2+}, Ca^{2+}, etc.) and anions ($H_2PO_4^-$, Cl^-, SO_4^{2-}, etc.) are the dominated respective source of acidity and alkalinity, if the uptake and removal of these ions from soil by plants leave equivalent H^+ and OH^- in the soil (Rengel 2003; Posch and Reinds 2009). The H^+ budget is therefore estimated by the following equations (Verstraten et al. 1990; Posch and Reinds 2009; Guo et al. 2010):

$$H_N^+ = \left(NH_{4\,In}^+ - NH_{4\,Out}^+\right) + \left(NO_{3\,Out}^- - NO_{3\,In}^-\right) \quad (1)$$

$$H_{BC}^+ = \frac{K_{uptake}}{K_{mm}} + 2 \times \frac{Ca_{uptake}}{Ca_{mm}} + 2 \times \frac{Mg_{uptake}}{Mg_{mm}} \quad (2)$$

$$OH_{Anions}^- = \frac{P_{uptake}}{P_{mm}} + 2 \times \frac{S_{uptake}}{S_{mm}} + \frac{Cl_{uptake}}{Cl_{mm}} \quad (3)$$

Proton (H^+) production by fertilizer N is calculated by Eq. 1, where $NH_4^+{}_{In}$ is the NH_4^+ input from the chemical N fertilizer and the output of NH_4^+ ($NH_4^+{}_{Out}$) was assumed to be zero since NH_4^+ leaching is negligible in the Chinese agricultural systems (Guo et al. 2010). $NO_3^-{}_{In}$ is the input of NO_3^- from the chemical N fertilizer, while $NO_3^-{}_{Out}$ is the net balance between the fertilizer NO_3^- input and outputs (i.e., NH_3 volatilization, plant uptake, and denitrification losses) other than NO_3^- leaching, which is estimated from considerations as follows. Firstly, we assumed that all NH_4^+ was nitrified into NO_3^- in soil, since the nitrification was very intensive in topsoil at this region (Chen et al. 2008), and that NO_3^--N is the preferred N nutrition by tobacco (Zhang and MacKown 1993; Chen et al. 2008). Secondly, N losses from NH_3 volatilization, soil denitrification, and residual soil N from applied fertilizers were not included in the calculation because either was very low under tobacco plantation at this region, compared with the high N input rate (Chen et al. 2008). Therefore, the N output was the difference between N input from the fertilizer and N uptake by plants. Meanwhile, in Eqs. 2 and 3, K_{uptake}, Ca_{uptake}, Mg_{uptake}, P_{uptake}, S_{uptake}, and Cl_{uptake} signify the uptake amounts of K, Ca, Mg, P, S, and Cl in plant tissues, which are their ion concentration multiplied by their biomass, and K_{mm}, Ca_{mm}, Mg_{mm}, P_{mm}, S_{mm}, and Cl_{mm} signify the molar mass of each element.

Statistical analysis and mapping

The independent sample *t* test was performed to determine significant changes in soil pH and exchangeable base cations between 2002 and 2012, respectively. The relationships between soil pH and exchangeable base cations were analyzed by the Pearson correlation coefficient test. A multiple comparisons analysis was carried out by one-way ANOVA to determine significant differences between element uptake and proton production by the least significant difference test at $P<0.05$. All the statistical analyses were performed by the SPSS version 16.0 (SPSS Inc., Chicago, IL, USA). Meanwhile, the ArcGIS software version 10.2 (ESRI Inc., California, USA) was used to map the tobacco plantation sites in these seven counties in Chongqing Municipal City, southwest China.

Results

Changes in soil exchangeable base cations and pH

After 1 decade (2002–2012) continuous tobacco plantation, except for Fengdu (FD), Shizhu (SZ), and Wulong (WL), soil pH in four of the seven counties was significantly decreased, with the highest decline in Qianjiang (QJ) (ΔpH=0.66) (Fig. 2a and Table 1). Exchangeable base cations (K^{+}+Ca^{2+}+Mg^{2+}), however, showed significant changes within and across all seven regions (Fig. 2b and Table 1). After 1 decade, the whole data from all seven tobacco plantation counties showed an average decrease of 27.6 % total exchangeable base cations or of 0.20 pH unit if such changes equally occurred.

Soil pH site distribution percentages, except at pH of ≤4.5, >5.5–≤6.0, and >7.5, in the whole Chongqing Municipal City were obviously changed from 2002 to 2012 (Fig. 3). For example, soil pH ranging from >4.5 to ≤5.5 was distributed in 193 sites (10.0 % of the total 1933 sites) in 2002 while in 416 sites (19.4 % of the total 2143 sites) in 2012, indicating a substantial increase of soil sites toward an acidic status after 10-year tobacco plantation. In contrast, soil pH ranging from >6.0 to ≤6.5 was distributed in 387 sites (20.0 % of the total 1933 sites) in 2002 while in 297 sites (13.9 % of the total 2143 sites) in 2012, indicating a substantial decrease of soil sites toward a neutral status after 10-year tobacco plantation. In

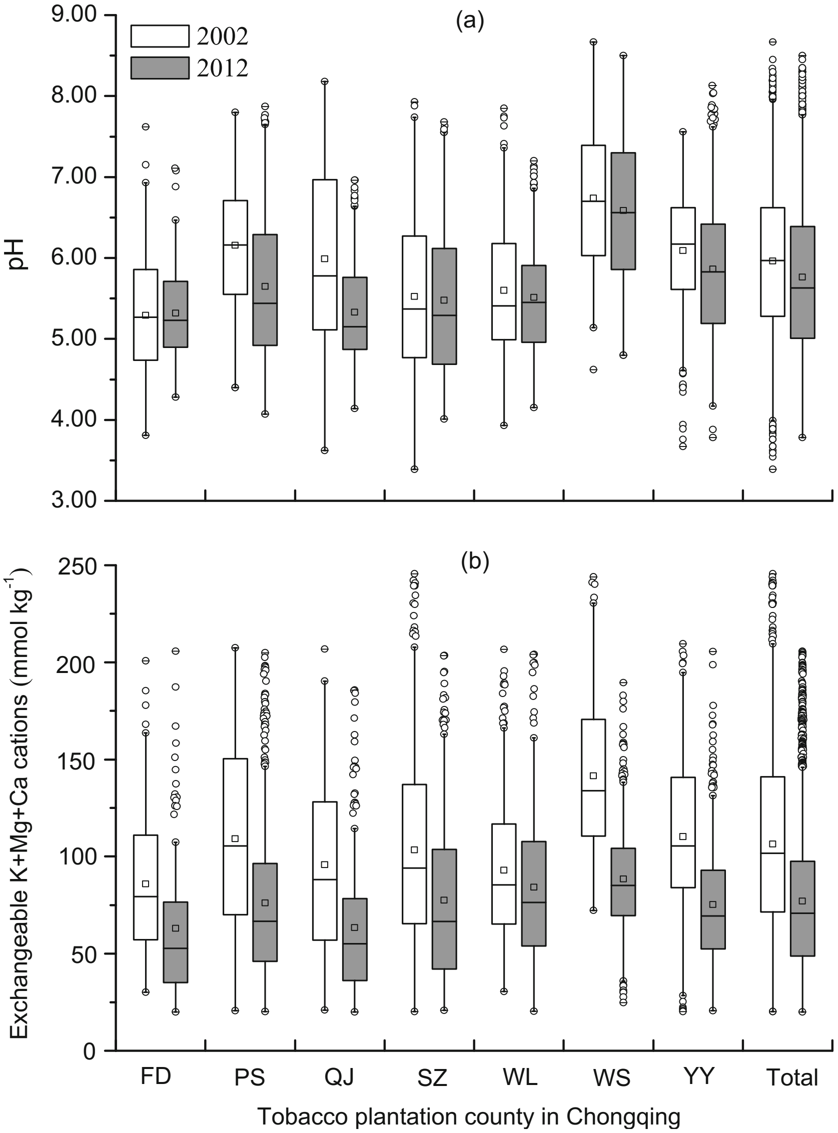


Fig. 2 Changes of soil pH (**a**) and exchangeable K, Ca, and Mg cations (**b**) after 10-year tobacco plantation (2002 versus 2012) at seven counties in Chongqing Municipal City, southwest China. FD Fengdu, PS Pengshui, QJ Qianjian, SZ Shizhu, WL Wulong, WS Wushan, YY Youyang. Total means the sum of all seven counties

Table 1 Average changes in soil pH and exchangeable base cations after 10 years of tobacco plantation (2002 versus 2012) at seven counties in Chongqing Municipal City, southwest China

	FD	PS	QJ	SZ	WL	WS	YY	Total
ΔpH	−0.03	0.50 ***	0.66 ***	0.04	0.09	0.15*	0.23***	0.20***
ΔBase cations (mmol kg^{-1})	22.8 ***	33.0 ***	32.3 ***	25.9 ***	8.8 *	24.0***	34.9***	29.4***

ΔpH and ΔBase cations indicate differences of pH and exchangeable base cations between 2002 and 2012, respectively

FD Fengdu, PS Pengshui, QJ Qianjian, SZ Shizhu, WL Wulong, WS Wushan, YY Youyang

*Significant difference between 2002 and 2012 at $P<0.05$

**Significant difference between 2002 and 2012 at $P<0.01$

***Significant difference between 2002 and 2012 at $P<0.001$

addition, with a normal distribution in 2002 to non-normal distribution in 2012, the GaussAmp fitting curve of soil pH changes did show an obvious increase toward a lower acidic pH direction while a decrease to a higher alkaline pH direction in 2012.

Relationships between soil pH and exchangeable base cations

At all seven sites, soil pH significantly positively correlated to exchangeable base cations (K^++Mg^{2+}+Ca^{2+}), with a general greater correlations in 2002 (R^2=0.51–0.76, $P<0.001$) than in 2012 (R^2=0.24–0.77, $P<0.001$) (Fig. 4). Meanwhile, except the Qianjiang and Wulong sites, the slope of the correlation equations was declined after 10-year tobacco plantation, indicating a change of correlativity between soil pH and exchangeable base cations. In addition, for the total data of these seven counties in the Chongqing Municipal City (Fig. 4h), one unit pH decrease consumed 40.3 and 28.3 mmol base cations per kilogram soil in 2002 and in 2012, respectively, indicating that less amounts of base cations were required to result in one unit pH decrease.

Proton budget estimated by tobacco harvest

Significantly higher uptake of elements by tobacco aboveground biomass patterned in the order of Ca>K>N>S>Mg>P≈Cl (Fig. 5a), while significantly higher H^+ production ranked as Ca uptake>K uptake>N_{In}-N_{Out}≈Mg uptake>P uptake≈Cl uptake>S uptake (Fig. 5b). The net H^+ production was positive under N_{In}-N_{Out}, Ca, K, and Mg uptake whereas negative with S, P, and Cl uptake (Fig. 5b). The H^+ production (H^+_N) under N_{In}-N_{Out} only accounted for ~13.4 % of the total H^+ production, while the uptake of Ca would cause plants to release about 9.83 kmol H^+ into per hectare field at 20-cm depth every year, which was about 68.0 % of total 14.45 kmol H^+ production by the three base cations (Fig. 5b).

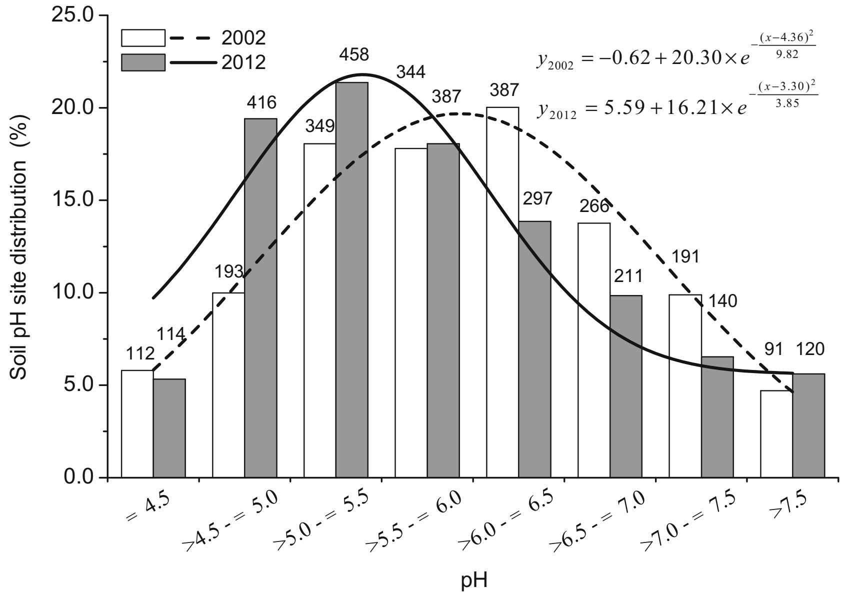


Fig. 3 Soil pH site distribution percentage in the whole Chongqing Municipal City (seven sampling counties) in 2002 (site n=1933) and 2012 (site n=2143). Numbers above the bars are site numbers

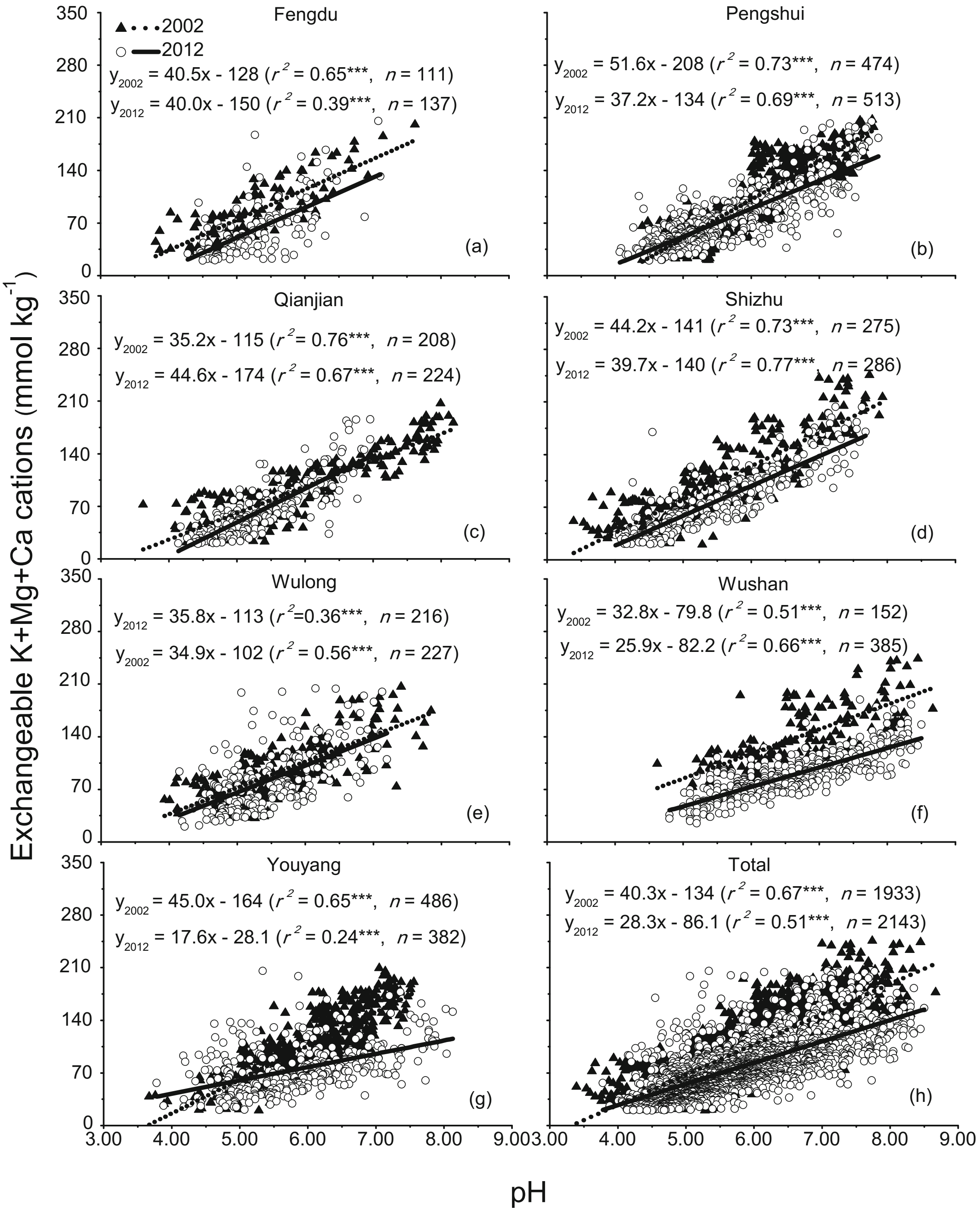


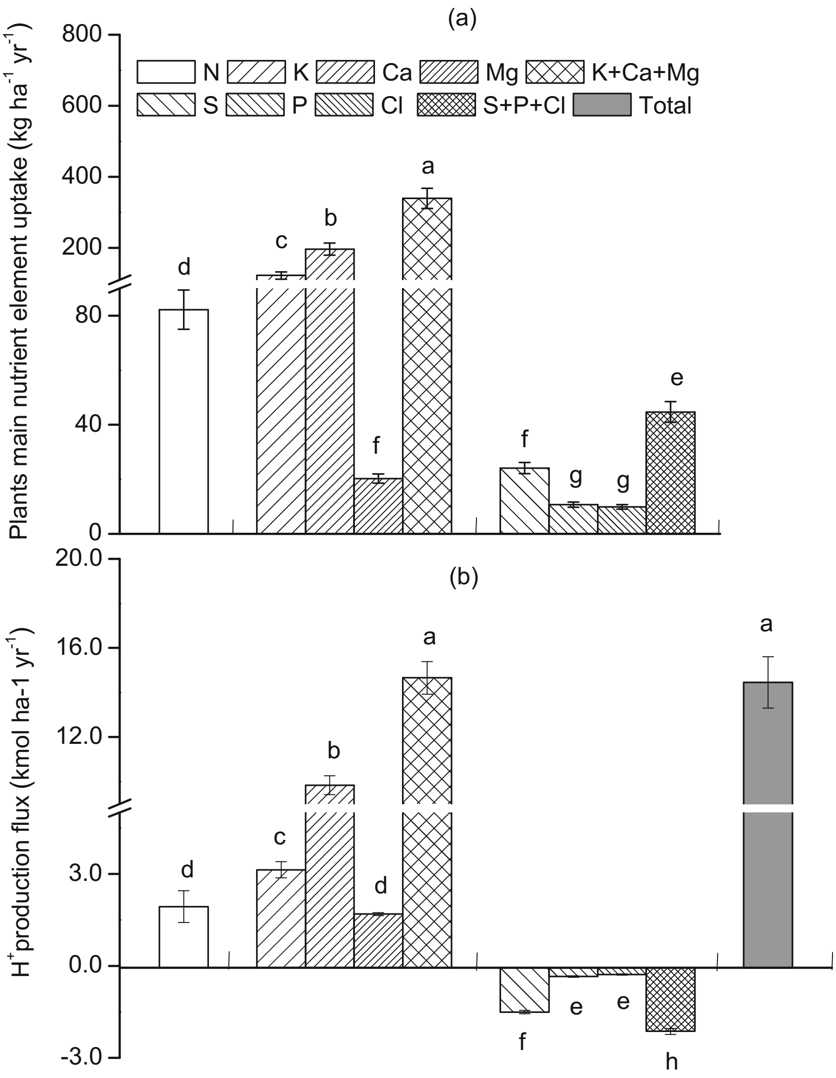
Fig. 4 Relationships between soil pH and exchangeable base cations in 2002 and 2012 in the seven counties, Chongqing Municipal City, southwest China. Total means the sum of all seven counties

Discussions

Soil acidification after tobacco plantation

In most Chinese-intensive agriculture systems, soil acidification is the main reason to cause land degradation and is becoming more challengeable to land sustainability (Guo et al. 2010). With a total of 1.2 million ha of flue-cured tobacco plantation, China is the major tobacco producer in the world. The continuously intensive tobacco plantation has resulted in serious soil acidification in China. For instance, soil pH ≤5.5 soil area was increased by 12.3 % with a 0.020 pH unit year^{-1} acidification rate after 10-year tobacco plantation in the Chongqing Municipal City (Fig. 2a). Such acidification rates in Chongqing were compatible with the average national 0.014–0.023 pH unit year^{-1} in China and could relate to soil

Fig. 5 Plant main nutrient element uptake (**a**) and H^+ production under fertilizer nitrogen and other ions uptake during tobacco plantations (**b**) in the seven counties, Chongqing Municipal City, southwest China in 2012. H^+ production in the N column indicates its equilibrium (N_{In}-N_{Out}) of N input (N fertilizer application, etc.) and output (N uptake by plants and N leaching, etc.). Different letters above columns indicate significant differences between different ion uptake



quality and biogeochemical cycling of soil nutrients, crop yield, and then food security (Guo et al. 2010). Meanwhile, soil acidification could also accelerate the accumulation of toxic aluminum, manganese and heavy metal ions in soil (Bolan et al. 2003; Wang et al. 2006), the decrease of biodiversity and ecosystem productivity (Rousk et al. 2010; Chen et al. 2013), and fertilizer use efficiency (Schroder et al. 2011; Duan et al. 2011).

Soil acid-buffering capacity and pH decrease with decreasing exchangeable base cations

Cation exchange is an important soil acid-buffering mechanism and causes an exportation of base cations by consuming and exchanging H^+ (Yang et al. 2013; Lucas et al. 2011), which will adsorb by soil clay and exist as soil potential acid. In this study, the exchangeable base cations were significantly decreased in all these seven tobacco planting counties after 10-year long-term continuous tobacco plantation (Fig. 2b), in spite of 1500 kg lime ha^{-1}, was annually applied in Fengdu (FD), Shizhu (SZ), and Wulong (WL) in 2010 and 2011. However, the ΔBase cations were comparatively lower in these three limed counties than other four non-limed counties, indicating that the external supplement of Ca^{2+} and Mg^{2+} with lime had compensated with the loss of base cations and also the increase of OH^- after the reaction of lime with water in the soil, in turn for some resurgence of pH, at FD, SZ, and WL (Table 1). Meanwhile, the exchangeable base cations significantly ($P<0.001$) negatively related to soil pH (Fig. 4). Base cations were critically important to buffer soil acidification process at the early acidification stage (Rengel 2003; Tian and Niu 2015), while Al could play a major role in buffering acidification when soil base cations were in a depletion status (Rengel 2003; Schroder et al. 2011). Unfortunately, changes in soil exchangeable Al have not been analyzed in this study. When soil pH decreases under 4.0, soil Al-hydroxide could be massively dissolved and then become exchangeable for inducing soil Al toxicity (De Vries et al. 1989). In this study, the consumption of base cations obviously decreased from 40.3 to 28.3 mmol kg^{-1} soil, resulting in one unit pH decrease after 10-year long-term continuous tobacco plantation (Fig. 4h). Such pH decrease could thus exhaust almost all soil base cations and decrease soil acid-buffering capacity from a long-term point of view. These results further confirmed that both soil acid-buffering capacity and pH values could decrease with the decrease of exchangeable base cations (Nelson and Su 2010; Klaminder et al. 2011; Xu et al. 2012).

Plant uptake and removal of base cations as a major mechanism of soil acidification

In agriculture systems, the main reasons for causing soil acidification include dry and wet acid deposition, the excess use of nitrogen fertilizer, and plant biomass removal as alkaline composition (Guo et al. 2010; Peters et al. 2011; Lu et al. 2014; Tian and Niu 2015). Studies have showed that fertilization with ammonium nitrogen (NH_4^+–N) could promote H^+ production to intensify soil acidification (Malhi et al. 2000; Chien et al. 2008; Guo et al. 2010; Schroder et al. 2011; Han et al. 2015). By contrast, fertilization with nitrate nitrogen (NO_3^-–N) had little effect on soil acidification (Malhi et al. 2000; Chien et al. 2008; Miao et al. 2011). In general, tobacco prefers to take up NO_3^-–N, rather than NH_4^+–N (Zhang and MacKown 1993; Chen et al. 2008), and the uptake of NO_3^- from soil by tobacco would leave an equivalent OH^- in the soil (Rengel 2003; Posch and Reinds 2009). Because NO_3^-–N was supplied for the preferred uptake by tobacco, the H^+ production from N input (N fertilizer application, etc.) and output (N uptake by plants, N leaching, etc.) was significantly lower in the present study (1.93 kmol H^+ ha^{-1} year^{-1}) than in other Chinese cropping systems (20 to 33 kmol H^+ ha^{-1} year^{-1}; see Guo et al. 2010).

Soil acidification in tobacco plantation fields is mainly due to the uptake and removal of base cations by aboveground tobacco biomass (Fig. 5). The removal of soil mineral nutrients from biomass harvest without returning to soil did result in H^+ accumulation in farming systems and directly led to the decrease of base cations and pH of soil (Lesturgez et al. 2006; Santonoceto et al. 2002; Randall et al. 2006). In general, 339.23 kg base cations ha^{-1} year^{-1} could be removed with tobacco harvest, which was 7.57 times higher than the removal of anions, leading to a production of 12.52 kmol H^+ ha^{-1} year^{-1}, which was much higher than a production of 1.93 kmol H^+ ha^{-1} year^{-1} from the N uptake and accumulation (Fig. 5). Furthermore, the uptake of calcium was also a major H^+ production pathway in tobacco plantation (Fig. 5). Releasing of H^+ from plant roots also had significantly positive relationship with excessive absorption of base cations (Rengel 2003).

Conclusions

Soil acidification induced by a decade long-term continuous tobacco plantation has become a major constraint on soil quality and tobacco productivity in Chongqing Municipal City, southwest China. The continuously annual removal of tobacco biomass had exported soil base cations and hence resulted in substantial soil acidification and decreased the soil acid-buffering capacity. As a result, other farming management strategies including application of gypsum, limestone and organic manure, straw return, or mulching residual biomass back into the soil under tobacco plantation or crop rotations could alleviate soil acidification and maintain crop production. Moreover, a continuous monitoring of soil pH changes would further enhance our understanding of long-term tobacco plantation on soil fertility and health.

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