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Research paper

# Effects of long-term nitrogen application on soil acidification and solution chemistry of a tea plantation in China



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### A R T I C L E I N F O

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#### ABSTRACT

In tea (Camellia sinensis) plantation areas, soil acidification mainly results from excessive nitrogen fertilization. However, the proposed theoretical explanations for soil acidification due to nitrogen fertilization are still lacking empirical validation because most studies have used short-term incubation periods or pot experiments. Here, both soil and soil solution samples were taken from a tea plantation field (Ultisol in USDA taxonomy system, or Alisol in WRB taxonomy system) treated using different nitrogen application rates: 0 (N0), 119 (N119), 285 (N285), and 569 (N569) kg N ha<sup>-1</sup> yr<sup>-1</sup> for 8 years (2006–2013). Soil pH and the concentrations of the relevant cations and anions were also determined. With no nitrogen fertilization (NO), the surface soil pH decreased from 4.16 to 3.32 after 8 years in the tea plantation. Compared with no nitrogen fertilization (N0), high nitrogen fertilization (N569) significantly decreased the soil pH from 3.32 to 3.15 and 3.67 to 3.35 in the soil at depths of 0-40 cm and 40-90 cm, respectively. However, the low (N119) and moderate (N285) nitrogen treatments showed non-significant effects upon soil pH. Our results confirm the previous findings that a high nitrogen application rate can accelerate soil acidification in a tea plantation, and that the subsoil is particularly susceptible to acidification after heavy nitrogen fertilization. Soil acidification also significantly decreased the nutrient base cations  $Ca^{2+}$ ,  $Mg^{2+}$ , and  $K^+$  in the soil. Our results suggest that heavy synthetic nitrogen fertilization should be partly replaced with compound or organic fertilizers to mitigate soil acidification and nutrient cation deficiency in tea plantation fields.

#### 1. Introduction

The tea plant (*Camellia sinensis*) is an important cash crop cultivated in many tropical and subtropical countries. Unlike many other crops, tea is a perennial leaf-harvested crop that grows best in acidic soil with an optimum pH between 4.0 and 5.5 (Ruan et al., 2007; Fung et al., 2008). Nevertheless, soil acidification can lead to the accumulation of aluminum (Al) and deficiencies in phosphorus (P), potassium (K), and magnesium (Mg) nutrients in tea field soil (Tachibana et al., 1995; Wang et al., 1997; Ruan et al., 2012). Concerning the quality of tea, soil acidification can increase the content of heavy metals (e.g., lead, Pb) and fluorine (F) in the harvested leaves, which may entail a risk to human health (Ruan et al., 2003; Jin et al., 2005; Zhang and Fang, 2007).

Generally, N fertilization or deposition is considered the major driver of soil acidification in croplands (Debreczeni and Kismanyoky, 2005; Schroder et al., 2011; Cai et al., 2015), grasslands (Martins et al., 2014; Chen et al., 2015), and forest ecosystems (Hogberg et al., 2006; Zhu et al., 2016). Specifically, it is thought that the accelerated soil acidification from N fertilization is directly caused by the production of protons via the nitrification process after ammonium nitrogen fertilization occurs (Barak et al., 1997; Zhou et al., 2014). The generated hydrogen ions (H<sup>+</sup>) are buffered by a suite of factors, including carbonate, silicate, exchangeable base cations, and the Al and/or iron (Fe) content, which depend on the soil pH (Bowman et al., 2008). Since base cations enter the soil solution as accompanying ions with the leaching  $NO_3^-$ , the buffering capacity of soil is consequently decreased under increasingly acidic conditions (Lucas et al., 2011).

How much N fertilization contributes to soil acidification remains unclear since the latter could happen naturally in tea plantation settings. Ding and Huang (1991) proposed that this might arise when the accumulated Al in the tea plant residue returns to the soil via litter fall

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and regular pruning. Recent studies have revealed that the root uptake of  $\rm NH_4^+$  and  $\rm Al^{3+}$ , and the subsequent release of protons, could also cause soil acidification in tea plantations (Ruan et al., 2004; Wan et al., 2012).

However, several processes may also weaken the rate of soil acidification in tea plantation soil. The contribution of nitrification to soil acidification could be diminished by reductions in the availability of substrate, as tea plants preferentially assimilate  $NH_4^+$  (Ruan et al., 2007). Additionally, the soil acidification rate may be reduced by less nitrate leaching, since the nitrification rate is typically inhibited by a low soil pH (Kemmitt et al., 2005). Moreover, soil acidification can be mitigated by the presence of soil buffering minerals, such as iron oxides (Li et al., 2012) and kaolinite in the soil (Lesturgez et al., 2006). So, rather than the total production of protons per se, soil acidification strongly depends on the soil buffering capacity and the depletion of the soil base cation pool (Bowman et al., 2008). But in the absence of longterm observations, what we know of soil acidification and its theoretical explanation has come mostly from short-term incubation studies or pot experiments. The long-term effects of N applications on soil acidification in tea plantation systems are not certain; nor have these effects upon soil chemistry and soil solution been clearly elucidated yet.

In this study, we conducted an 8-year-long field experiment that considered a range of N additions in Zhejiang, an important green tea production area in China, for which we tested the soil pH and the ion concentrations in the soil solution. Our objectives were 1) to reveal the contribution of N fertilization to soil acidification in a tea plantation; 2) to illustrate the extent of cation and anion changes in the soil solution under a high N application in a tea plantation; and 3) to distinguish and evaluate the main factors controlling soil pH in the tea plantation.

#### 2. Materials and methods

#### 2.1. Site description

The field experiment was located at Hangzhou, in Zhejiang province in China ( $120^{\circ}05'E$ ,  $30^{\circ}10'N$ ), within the country's main region for green tea cultivation. The experimental site has a monsoon climate, with 1533 mm yr<sup>-1</sup> in precipitation and a mean annual temperature of 17.0 °C. The soil at the site is classified as Ultisol (Soil Survey Staff, 2014) or Alisol (IUSS Working Group WRB, 2015), developed from parent material of a Quaternary eolian red deposit, with a texture that is loamy clay. The surface (0–20 cm) soil properties that existed at the site prior to the experiment are shown in Table S1.

Before the experiment, tea plants of the variety 'Longjing 43' grew for 6 years in the studied field. They had been planted in single lines 1.5-m apart, with an in-line distance of 0.33 m; this represents a planting density of approximately 60 000 plants ha<sup>-1</sup>. To sustain the growth and the yield of these tea plants, 450 kg ha<sup>-1</sup> of urea and 4500 kg ha<sup>-1</sup> of organic fertilizer (rapeseed cake) were applied yearly before the experiment.

#### 2.2. Experimental design

The field experiment began in 2005 and consisted of four treatments of N application rates:  $0 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  (control, N0), 119 kg N ha<sup>-1</sup> yr<sup>-1</sup> ('low N', N119), 285 kg N ha<sup>-1</sup> yr<sup>-1</sup> ('moderate N', N285), and 569 kg N ha<sup>-1</sup> yr<sup>-1</sup> ('high N', N569). The N fertilizer was urea, and it was split into four seasonal applications; i.e., 30% in early February, before spring tea; 20% in late May, after spring tea but before summer tea; 20% in early July, before autumn tea; and 30% late October, after autumn tea. Phosphorus, as super phosphate, and potassium, as potash of sulfate, were both applied in October as the base fertilizer of all the treatments (Table S2). All the fertilizers were applied in the band furrows (at a depth of 15 cm) between two rows of tea plants, and then covered with soil after their application. were randomly distributed. Each plot was  $24 \text{ m}^2$  in size and consisted of two 8-m-long lines of tea plants. The entire experimental area was on flat terrain, and the plots were separated from each other by one blank line of tea plants or a 1-m-wide segment.

#### 2.3. Soil sampling and analysis

In February 2013, soil samples were collected from the topsoil layer (i.e., 0–40 cm depth), where most roots are found, and from the subsoil layer (i.e., 40–90 cm depth) with a 5-cm-diameter stainless steel corer. Each composite soil sample consisted of six or seven randomly selected points within each plot; these subsamples were completely mixed, and any roots, plant residues, and stones were removed by hand. The fresh soil was passed through a 2-mm-mesh sieve and divided into two parts: one was stored at 4  $^{\circ}$ C in a refrigerator, while the other was air-dried for the later determination of soil pH, exchangeable cations, and the water-soluble ions.

Soil pH was measured in pastes of 1:1 (w/v) in deionized water with an ORION 3 STAR pH meter (Thermo Ltd., USA). Available  $NH_4^+$  and  $NO_3^-$  were extracted by 2 M of KCl and quantified by a Flow Injection Analyzer (SAN + +, SKALAR Ltd., Netherlands).

The soil exchangeable cations were extracted with 0.1 M of BaCl<sub>2</sub> at a ratio of 1:50 (w/v) for 30 min, and passed through 0.45-µm-sized cellulose–acetate paper filters for further measurement (Lu et al., 2014). From the ensuing extracts, the contents of base cations (Ca<sup>2+</sup>, Mg<sup>2+</sup>, K<sup>+</sup>, Fe<sup>3+</sup>, Mn<sup>2+</sup>, and Na<sup>+</sup>) were determined by inductively coupled plasma atomic emission spectroscopy (ICP-AES; Thermo Jarrell Ash Ltd., USA), while those of exchangeable Al<sup>3+</sup> and H<sup>+</sup> were determined by NaOH-neutralization titration. The available P, available K, and available sulfur (S) in the soil were extracted by shaking 2.5 g of soil with 25 ml of a Mehlich-3 solution for 5 min (Mehlich, 1984); their contents were determined by ICP-AES.

Free Al/Fe oxides and amorphous Fe/Al oxides were extracted by using the Sodium citrate-bicarbonate-dithionite (DCB) method and the acid ammonium oxalate method, respectively (Mehra and Jackson, 1960).

The water-soluble ions (K<sup>+</sup>, Na<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup>,  $SO_4^{2-}$ , F<sup>-</sup>, and Cl<sup>-</sup>) in the soil samples were extracted with deionized water at a ratio of 1:5 (w/v) for 30 min, and after filtration the samples were analyzed for their cations and anions. NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> were quantified in a SAN<sup>++</sup> Flow Injection Analyzer (SKALAR Ltd., Netherlands). The cations (K<sup>+</sup>, Na<sup>+</sup>, Ca<sup>2+</sup>, and Mg<sup>2+</sup>) were measured by using ICP-AES and the anions (SO<sub>4</sub><sup>2-</sup>, F<sup>-</sup>, and Cl<sup>-</sup>) measured by using Ion Chromatography (Dionex DX-2000, USA).

#### 2.4. Soil solution sampling and analysis

In each plot, two ceramic porous lysimeters (Model 1900, Soilmoisture Equipment Corp., Santa Barbara, CA, USA) were installed at soil depths of -15 cm (i.e., within the main rooting zone) and -90 cm (i.e., beyond the rooting zone) in October 2012 to collect their respective soil solutions. These solutions were sucked into bottles using a vacuum pump after each rain event from March to October 2013. The volumes of collected soil solution were recorded by the weighting method.

All the soil solution samples were passed through 0.45-µm filters and stored in a refrigerator at 4 °C. Their ion concentrations (K<sup>+</sup>, Na<sup>+</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, Al<sup>3+</sup>, NH<sub>4</sub><sup>+</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, F<sup>-</sup>, and Cl<sup>-</sup>) were determined as described above. The mean ion concentrations for each soil solution depth per treatment level at a specific sampling date were calculated from the respective volume-weighted average of the replicates.

#### 2.5. Data analysis

Each treatment consisted of four replicate plots, with all 16 plots

Soil cation exchange capacity (CEC) was calculated as the sum of

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