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Phosphorus addition mitigates N_2O and CH_4 emissions in N-saturated subtropical forest, SW China

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Abstract. Chronically elevated nitrogen (N) deposition has led to severe nutrient imbalance in forest soils. Particularly in tropical and subtropical forest ecosystems, increasing N loading has aggravated phosphorus (P) limitation of biomass production, and has resulted in elevated emissions of nitrous oxide (N_2O) and reduced uptake of methane (CH_4), both of which are important greenhouse gases. Yet, the interactions of N and P and their effects on greenhouse gas emissions remain elusive. Here, we report N_2O and CH_4 emissions together with soil N and P data for a period of 18 months following a single P addition (79 kg P ha^{-1}, as NaH_2PO_4 powder) to an N-saturated, Masson pine-dominated forest soil at TieShanPing (TSP), Chongqing, south-western (SW) China. We observed a significant decline in both nitrate (NO_3^-) concentrations in soil water (5 and 20 cm depths) and in soil N_2O emissions, following P application. We hypothesise that enhanced N uptake by plants in response to P addition, resulted in less available NO_3^- for denitrification. By contrast to most other forest ecosystems, TSP is a net source of CH_4. P addition significantly decreased CH_4 emissions and turned the soil from a net source into a net sink. Based on our observation and previous studies in South America and China, we believe that P addition relieves N inhibition of CH_4 oxidation. Within the 1.5 years after P addition, no significant increase of forest growth was observed and P stimulation of forest N uptake by understorey vegetation remains to be confirmed. Our study indicates that P fertilisation of N-saturated, subtropical forest soils may mitigate N_2O and CH_4 emissions, in addition to alleviating nutrient imbalances and reducing losses of N through NO_3^- leaching.

1 Introduction

Anthropogenic activities have transformed the terrestrial biosphere into a net source of methane (CH_4), nitrous oxide (N_2O) and carbon dioxide (CO_2), leading to increased radiative forcing (Montzka et al., 2011; Tian et al., 2016). During the last decade, atmospheric concentrations of CO_2, CH_4 and N_2O have increased at rates of 1.9 ppm yr^{-1}, 4.8 and 0.8 ppb yr^{-1}, respectively (Hartmann et al., 2013). In China, the exponential increase of reactive nitrogen (N) input into the biosphere since the 1970s has likely led to more carbon (C) being sequestered in the biosphere (Cui et al., 2013; Shi et al., 2015). However, enhanced emissions of N_2O and CH_4 due to chronic N pollution potentially offset the cooling effect by C sequestration (Liu and Greaver, 2009; Tian et al., 2011).

Microbial nitrification and denitrification in soils account for about 60 % of N_2O emissions globally (Ciais et al., 2013; Hu et al., 2015). Although, microbial activity is often restricted in low pH soils of unproductive forests, surprisingly large N_2O emissions have been reported from acid, upland forest soils in southern China (Zhu et al., 2013b). Reported average N_2O fluxes in humid, subtropical forests range from 2.0 to 5.4 kg N_2O–N ha^{-1} yr^{-1} (Fang et al., 2009; Tang et al., 2006; Zhu et al., 2013b), which by far exceeds global averages for temperate or tropical forest ecosystems (Werner et al., 2007; Zhuang et al., 2012). This has been attributed to frequent shifts between aerobic and anaerobic conditions in soils during monsoonal summers, promoting alternating nitrification and denitrification (Zhu et al., 2013b) and causing

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large soil NO_3^- concentrations due to efficient cycling of deposited N in acid subtropical soils (Yu et al., 2016).

Chronically elevated rates of N deposition (30–65 kg N ha^{-1} yr^{-1}; Xu et al., 2015) have resulted in strong nutrient imbalances in southern Chinese forests, aggravating phosphorus (P) limitation (Du et al., 2016). Phosphorous deficiency in N-saturated forests restricts forest growth and thus limits its capability to retain N (Huang et al., 2015; Li et al., 2016), resulting in ample amounts of inorganic N (NH_4^+ and NO_3^-) being present in the soil solution. Accordingly, Hall and Matson (1999) observed much larger N_2O emission in P-limited than in N-limited tropical forests 1 year after repeated addition of N. Likewise, previous N manipulation studies in forests of southern China reported pronounced stimulation of N_2O emissions by N addition (Chen et al., 2016; Wang et al., 2014; Zheng et al., 2016), supporting the idea that P limitation causes forests to be more susceptible to N saturation and N_2O–N loss. In an N-limited tropical montane forest in southern Ecuador, P addition alone (10 kg P ha^{-1} yr^{-1}) had no effect on N_2O emissions during the first 2 years. However, N_2O emission was smaller when P was added together with N (50 kg N ha^{-1} yr^{-1}) rather than in treatments with N addition alone (Martinson et al., 2013). After continued fertilisation for 3 years, also P addition alone reduced N_2O emissions at these sites (Müller et al., 2015). In tropical China, with high-N deposition ($\sim$ 36 kg N ha^{-1} yr^{-1}; Mo et al., 2008), P addition (150 kg P ha^{-1} yr^{-1}) to an old-growth forest revealed a similar pattern, with no initial effect on N_2O emissions (0–2 years) but a significant longer-term effect (3 to 5 years) on N_2O emissions (Chen et al., 2016; Zheng et al., 2016). In a secondary tropical forests in southern China, Wang et al. (2014) found no effect on N_2O emissions of P alone (100 kg P ha^{-1} yr^{-1}), and in treatments combining P with N (100 kg N ha^{-1} yr^{-1}), N_2O emissions increased during the wet season. Meanwhile, they observed a significant increase in soil microbial biomass after P addition, which is in line with previous findings in tropical forest soils of southern China (Liu et al., 2012). Thus, they attributed the stimulating effect of P addition on N_2O emissions to the larger nitrification and denitrification potential of the increased soil microbial biomass. This was also proposed by Mori et al. (2014), based on results from a short-term incubation study with P addition, excluding plant roots.

As the sole biogenic sink for CH_4, upland soils play an important role in balancing terrestrial CH_4 emissions (Ciais et al., 2013; Dutaur and Verchot, 2007). Atmospheric CH_4 uptake in soil is mediated by the activity of methanotrophic bacteria, which oxidise CH_4 to CO_2 to gain energy for growth. Well-drained forest and grassland soils are dominated by yet uncultured, high-affinity methanotrophs residing in the upper soil layers (Le Mer and Roger, 2010). In addition to edaphic factors (pH and nutrients), parameters affecting the diffusion of CH_4 into the soil (soil structure, moisture, temperature) are believed to be major controllers for CH_4 uptake (Smith et al., 2003). A number of studies have shown that excess N affects CH_4 fluxes in forest soils (Liu and Greaver, 2009; Veldkamp et al., 2013; Zhang et al., 2008b). In general, N addition promotes CH_4 uptake in N-limited soils by enhancing growth and activity of methanotrophs, whereas excessive N input and N saturation inhibit CH_4 oxidation on an enzymatic level by substrate competition between CH_4 and NH_4^+ (Aronson and Helliker, 2010; Bodelier and Laanbroek, 2004). P-addition experiments in N-enriched soils have shown positive effects on CH_4 uptake (Mori et al., 2013a; Zhang et al., 2011), but the underlying mechanisms, i.e. whether P addition affects the methanotrophic community directly or alleviates the N-inhibition effect on CH_4 oxidation through enhanced N uptake (Mori et al., 2013b; Veraart et al., 2015), remain unresolved.

Subtropical forests in southern China show strong signs of N saturation, with exceedingly high-NO_3^- concentrations in soil water (Larssen et al., 2011; Zhu et al., 2013b). Little is known about how P addition affects N cycling and N_2O emission in these acidic, nutrient-poor soils. Likewise, the importance of increased inorganic N concentrations for soil–atmosphere exchange of CH_4, and how this is affected by P fertilisation, remain to be elucidated for soils of the subtropics. Here, we assessed N_2O and CH_4 fluxes in an N-saturated subtropical forest in south-western (SW) China under ambient N deposition and studied the effects of P addition on emission rates, nutrient availability and tree growth. We hypothesised that (i) P addition stimulates forest growth, (ii) stimulated forest growth results in increased N uptake by trees and understorey vegetation, and thus decreases the soil inorganic N concentration and (iii) P addition reduces soil N_2O emission and promotes CH_4 uptake.

2 Materials and methods

2.1 Site description

The study site TieShanPing (TSP) is a 16.2 ha subtropical forest (29°380′ N, 106°410′ E; 450 m a.s.l.), about 25 km north-east of Chongqing, SW China. TSP is a naturally regenerated, secondary mixed coniferous–broadleaf forest, which developed after clear cutting in 1962 (Larssen et al., 2011). The forest stand is dominated by Masson pine (*Pinus massoniana*) and has a density of about 800 stems ha^{-1} (Huang et al., 2015). TSP has a monsoonal climate, with mean annual precipitation of 1028 mm, and a mean annual temperature of 18.2 °C (Chen and Mulder, 2007a). Most of the precipitation ($>70\,\%$) occurs during summer periods (April to September). Soils are predominantly well-drained, loamy yellow mountain soil, classified as Haplic Acrisol (WRB 2014), with a thin O horizon (<2 cm). In the O–A horizon, soil pH is around 3.7, and the mean C / N and N / P ratios are 17 and 16, respectively. In the AB horizon, which has a slightly higher pH, mean C / N is well above 20.

Table 1. Ambient soil properties of the experimental plots at Tieshanping (TSP). Values are means and standard deviations in parenthesis $(n = 6)$[a]. Soils were sampled in August 2013.

	Soil layer	pH	Total C $g\,kg^{-1}$	Total N $g\,kg^{-1}$	Total P $mg\,kg^{-1}$	C / N	N / P
Block 1	O / A (0–3 cm)	3.7 (0.1)	80.7 (32.3)	4.8 (1.7)	308 (57)	17.0 (2.5)	15.5 (5.7)
	AB (3–8 cm)	3.8 (0.0)	23.9 (9.3)	1.3 (0.6)	–[b]	20.0 (3.0)	–
	B (8–20 cm)	3.9 (0.2)	8.6 (1.2)	< 0.05	–	–	–
Block 2	O / A (0–3 cm)	3.6 (0.1)	77.6 (13.4)	4.7 (0.8)	297 (44)	16.7 (1.3)	15.7 (2.8)
	AB (3–8 cm)	3.7 (0.1)	20.2 (5.3)	1.0 (0.3)	–	21.4 (3.3)	–
	B (8–20 cm)	3.9 (0.1)	7.1 (1.6)	< 0.05	–	–	–
Block 3	O / A (0–3 cm)	3.6 (0.1)	67.0 (15.5)	3.8 (0.8)	223 (45)	17.4 (0.6)	17.2 (3.7)
	AB (3–8 cm)	3.6 (0.1)	21.0 (7.9)	1.1 (0.5)	–	24.5 (4.6)	–
	B (8–20 cm)	3.8 (0.1)	7.2 (1.5)	< 0.05	–	–	–

	Soil Layer	P_{Al} $mg\,kg^{-1}$	Al_{ox} $mg\,kg^{-1}$	Fe_{ox} $mg\,kg^{-1}$	P_{ox} $mg\,kg^{-1}$	$P_{ox}/(Al_{ox}+Fe_{ox})$
Block 1	O/A (0–3 cm)	5.8 (1.4)	1700 (513)	1933 (350)	85.8 (22.6)	0.025 (0.008)
	AB (3–8 cm)	2.1 (0.6)	1217 (243)	1692 (493)	47.1 (22.0)	0.016 (0.007)
	B (8–20 cm)	< 1.0	1083 (90)	1158 (249)	29.3 (28.6)	0.012 (0.011)
Block 2	O / A (0–3 cm)	5.9 (1.0)	1500 (238)	1792 (215)	79.2 (21.5)	0.024 (0.007)
	AB (3–8 cm)	1.6 (0.4)	925 (149)	1517 (320)	37.2 (10.7)	0.016 (0.006)
	B (8–20 cm)	< 1.0	892 (209)	1033 (413)	16.1 (10.5)	0.009 (0.007)
Block 3	O / A (0–3 cm)	4.1 (0.9)	1367 (180)	1667 (168)	50.7 (10.9)	0.017 (0.003)
	AB (3–8 cm)	4.4 (4.0)	1075 (128)	1350 (150)	24.8 (8.3)	0.010 (0.002)
	B (8–20 cm)	< 1.0	992 (130)	875 (138)	8.0 (2.0)	0.004 (0.001)

P_{Al} is ammonium lactate-extractable P, Al_{ox} is oxalate-extractable Al, Fe_{ox} is oxalate-extractable Fe, P_{ox} is oxalate-extractable P.
[a] Water-extractable P was below a detection limit of $5\,mg\,kg^{-1}$, thus not presented in table, [b] Data not available.

The soil bulk density of the O–A horizon (~ 5 cm) is about $0.75\,g\,cm^{-3}$. Generally, soil water-filled pore space (10 cm) on the hillslopes ranges from 50 to 70 % (mean ~ 60 %; Zhu et al., 2013b). More details on soil properties are given in Table 1.

Annual inorganic N deposition at TSP measured in throughfall varies between 40 and $65\,kg\,N\,ha^{-1}$ (dominated by NH_4^+; Yu et al., 2016), while the annual bulk N deposition is from 20 to $30\,kg\,N\,ha^{-1}$ (Chen and Mulder, 2007b). According to regional data, annual P deposition via throughfall is $< 0.40\,kg\,ha^{-1}$ (Du et al., 2016). Strong soil acidification has been reported to cause severe decline in forest growth at TSP since 2001 (Li et al., 2014; Wang et al., 2007), and a decrease in abundance and diversity of ground vegetation (Huang et al., 2015). Pronounced N saturation with strong NO_3^- leaching from the top soil has aggravated P deficiency at TSP (Huang et al., 2015). The total P content in the O–A horizon is $\sim 300\,mg\,kg^{-1}$, while ammonium lactate-extractable P is smaller than $5\,mg\,kg^{-1}$ (Table 1).

2.2 Experimental design

Three blocks, each having two 20 m × 20 m plots, were established on well-drained soils of a gently sloping hillside. Adjacent plots were separated by at least 10 m buffer zone. In each block, plots were randomly assigned to a reference and a P treatment. On 4 May 2014, a single dose of P fertiliser was applied as solid $NaH_2PO_4{\cdot}2H_2O$, at a rate of $79.5\,kg\,P\,ha^{-1}$. The amount of P added was estimated from P adsorption isotherms (Supplement, Table S1 and Fig. S1), to ensure significantly increase in soil-available P. To apply P fertiliser evenly, we divided each plot into a 5 m × 5 m grid and broadcasted the powdered fertiliser by hand in each grid cell. The P dose applied at TSP was intermediate as compared to the $10\,kg\,P\,ha^{-1}\,yr^{-1}$ applied by Müller et al. (2015) to a mountain forest in Ecuador and the $150\,kg\,P\,ha^{-1}\,yr^{-1}$ applied by Zheng et al. (2016) to a subtropical forest in southern China.

The addition of $NaH_2PO_4^{'}2H_2O$ at the P-treated plots also resulted in an input of $59.0\,kg\,ha^{-1}$ of sodium (Na); 1 month after the fertiliser application, Na^+ concentrations in soil water of the P treatments were about $5\,mg\,L^{-1}$ at 5 cm depth and $3\,mg\,L^{-1}$ at 20 cm depth (Table S2). Although somewhat larger than in the reference plots ($0.52–1.31\,mg\,L^{-1}$), the Na^+ concentration in soil water of the P treatments are unlikely to have exerted a significant negative impact on plant and microbial activities.

Table 2. Soil pH, C, N and P contents in the O–A horizon (0–3 cm) in the references (Ref) and P treatments. Values are means and standard deviations in parenthesis ($n = 9$). P addition was conducted on 4 May 2014, after the first two sampling dates.

		pH	Total C g kg^{-1}	Total N g kg^{-1}	C / N	P_{Al} mg kg^{-1}	Total P mg kg^{-1}
2 August 2013	Ref	3.7 (0.1)^{ab*}	8.3 (2.3)^{ab}	0.5 (0.1)^{ab}	16.9 (1.1)^{b}	5.4 (1.4)^{a}	292 (46)^{ab}
	P	3.6 (0.1)^{b}	6.7 (2.0)^{b}	0.4 (0.1)^{b}	17.1 (2.1)^{ab}	5.1 (1.3)^{a}	260 (70)^{b}
2 May 2014	Ref	3.7 (0.1)^{a}	12.2 (4.2)^{a}	0.9 (0.3)^{a}	13.7 (1.5)^{b}	19.0 (8.0)^{a}	336 (65)^{a}
	P	3.8 (0.2)^{a}	9.0 (3.5)^{ab}	0.7 (0.2)^{ab}	14.2 (2.8)^{ab}	13.7 (5.2)^{a}	270 (72)^{a}
10 May 2014	Ref	3.8 (0.1)^{ab}	9.9 (2.1)^{a}	0.7 (0.2)^{ab}	14.0 (0.7)^{b}	15.4 (7.0)^{b}	304 (49)^{b}
	P	3.9 (0.3)^{a}	8.0 (1.9)^{a}	0.6 (0.1)^{b}	14.3 (1.3)^{ab}	174 (114)^{a}	572 (242)^{a}
2 December 2014	Ref	3.8 (0.1)^{a}	10.5 (3.6)^{a}	0.7 (0.3)^{a}	14.5 (1.3)^{ab}	14.2 (7.4)^{b}	328 (102)^{b}
	P	3.9 (0.2)^{a}	9.5 (2.1)^{a}	0.7 (0.1)^{ab}	14.0 (0.8)^{b}	66 (24)^{a}	442 (106)^{ab}
2 August 2015	Ref	3.9 (0.2)^{ab}	8.3 (2.2)^{ab}	0.4 (0.1)^{ab}	20.5 (2.5)^{a}	13.4 (6.2)^{b}	291 (61)^{a}
	P	4.0 (0.2)^{a}	6.5 (1.9)^{b}	0.3 (0.1)^{b}	19.7 (2.2)^{ab}	57 (36)^{a}	383 (136)^{a}

* Different letters indicate significant differences between references and P treatments ($p < 0.05$).

2.3 Sample collection and analyses

Within each plot, three ceramic lysimeters (P80; Staatliche Porzellanmanufaktur, Berlin) were installed at 5 and 20 cm soils near the plot centre in August 2013. To obtain water samples, 350 mL glass bottles with rubber stoppers were pre-evacuated, using a paddle pump, and connected to the lysimeters for overnight sampling. Between November 2013 and October 2015, we sampled soil pore water bi-monthly in the dry and dormant season and monthly during the growing season. All water samples were kept frozen during storage and transport. Concentrations of NH_4^+, NO_3^-, potassium (K^+), calcium (Ca^{2+}) and magnesium (Mg^{2+}) in soil water were measured at the Research Center for Eco-Environmental Sciences (RCEES), Chinese Academy of Sciences, Beijing, using ion chromatography (DX-120 for cations and DX-500 for anions).

In August 2013, soils from the O–A (0–3 cm), AB (3–8 cm) and B (8–20 cm) horizons were sampled near the lysimeters for soil analysis. Total P and plant-available P contents were monitored in samples collected from the O–A horizons every 6 months, starting 2 days before P addition. Soil samples were kept cold (< 4 °C) during transport and storage. Before analysis, soil samples were air dried and sieved (2 mm). Soil pH was measured in soil suspensions (10 g dry soil and 50 mL deionised water) using a pH metre (PHB-4, Leici, China). Total soil C and N contents were determined on dried and milled samples, using a LECO elemental analyser (TruSpec$^{@}$CHN, USA). To measure total P, 1 g dry soil was digested with 5 mL of 6 M H_2SO_4 (Singh et al., 2005) and measured as ortho-phosphate by the molybdenum blue method (Murphy and Riley, 1962). Ammonium lactate-extractable (0.01 M) P and H_2O-extractable P (P_{Al} and P_{H_2O}, respectively) were measured as ortho-phosphate after extraction (1.5 g dry soil in 50 mL solution) (Singh et al., 2005). Ammonium oxalate-extractable (0.2 M) Fe, Al and P were measured by inductively coupled plasma optical emission spectroscopy (ICP-OES, Agilent, USA) after extraction (1.5 g dry soil in 50 mL solution).

From August 2013 onwards, we measured N_2O and CH_4 emissions in triplicate close to the lysimeters, using static chambers (Zhu et al., 2013b). The measurements were conducted bi-monthly in the dry and dormant season and monthly during the growing season, simultaneously with the sampling of soil pore water. To investigate the immediate effect of P addition on N_2O emissions, we also sampled the gas emissions once before (2 May) and 3 times (7, 10 and 12 May) after the P application. Gas samples were taken 1, 5, 15 and 30 min after chamber deployment; 20 mL gas samples were injected into pre-evacuated glass vials (12 mL) crimp sealed with butyl septa (Chromacol, UK), maintaining overpressure to avoid contamination during shipment. Mixing ratios of N_2O and CH_4 were analysed using a gas chromatograph (model 7890A, Agilent, USA) at RCEES, equipped with an electron capture detector (ECD) for detection of N_2O (at 375 °C with 25 mL min^{-1} Ar / CH_4 as make-up gas), a flame ionization detector (FID) for CH_4 (250 °C; 20 mL min^{-1} N_2 as make-up gas) and a thermal conductivity detector (TCD) for CO_2. Exchange rates between soil and atmosphere (emission/uptake) were calculated from measured concentration change in the chambers over time, applying linear or polynomial fits to the concentration data. Cumulative N_2O emissions over time were estimated by linear interpolation between measurement dates (Zhu et al., 2013b).

From October 2013 onwards, litterfall was collected during the first week of every month in five replicates per plot. Litterfall collectors were made of 1 m^2 nylon nets (1 mm mesh size), held in place by four wooden poles 0.8 m above the ground. Fresh litter was dried at 65 °C. In early November 2013 and 2014 (at the end of the growing season), we collected current-year pine needles from several branches of three trees in each plot. The collected needles were dried at

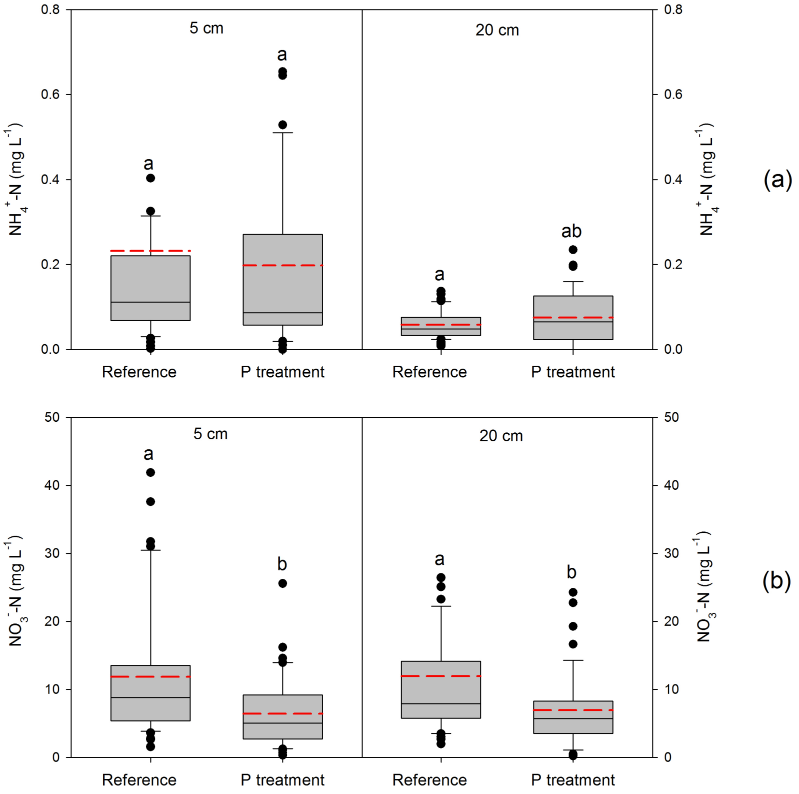

Figure 1. Box-and-whisker plots of NH_4^+ **(a)** and NO_3^- **(b)** concentration in soil water at 5 and 20 cm depths in the references and P treatments, throughout 1.5 years after the P addition; red dashed lines indicate mean values; different letters indicate significant differences ($p < 0.05$).

65 °C and the dry weight of 500 needles was determined. A subsample was dried at 80 °C and finely milled prior to chemical analysis at the Chinese Academy of Forestry. Total C and N were measured using an elemental analyser (FLASH 2000; Thermo Scientific; USA). The contents of K, Ca, Mg and P in the needles were determined by ICP-AES (IRIS Intrepid II; Thermo Scientific; USA) after digesting 0.25 g dry weight samples with 5 mL of ultra-pure nitric acid. In November 2013, and 2014, and in February 2015, we measured the height and the diameter at breast height (DBH) of 6 to 10 Masson pines (marked in November 2013; DBH > 5 cm) at each plot. DBH was then used to estimate the standing biomass of Masson pines based on standard allometric equations (Li et al., 2011; Zeng et al., 2008).

Daily average air temperature and daily total precipitation were monitored from July 2013 to November 2015 by a weather station (WeatherHawk 232, USA) placed on the roof at the local forest bureau, at about 1 km distance from the sampling site (Yu et al., 2016).

2.4 Statistical analyses

Statistical analyses were performed using R version 3.3.1 (R Core Team, 2016). All data were tested for normality (Kolmogorov–Smirnov's test) and homoscedasticity (Levene's test) before further analysis. If not normally distributed, the data were normalised by logarithmic transformation. Considering heterogeneity among blocks, temporal variabilities of NO_3^- concentrations, N_2O and CH_4 fluxes were presented separately for each block. For time series data, we used linear mixed-effect (LME) models, to account for both repeated measurements and within-group variance of a stratification variable (block design). LME models were applied to test the effects of P addition on soil N_2O and CH_4 fluxes, NH_4^+, NO_3^-, K^+, Ca^{2+} and Mg^{2+} concentrations in soil water, as well as litterfall weight (Koehler et al., 2009; Müller et al., 2015). The analysis was based on data for plot means (the average of 3 subplot replicates) from three blocks. In LME models, treatments (reference or P addition) were considered fixed effects, while sampling time and plots were treated as random effects. We then assessed the significance of fixed effects through analysis of variance for LME models. One-way analysis of variance (ANOVA) was conducted to ex-

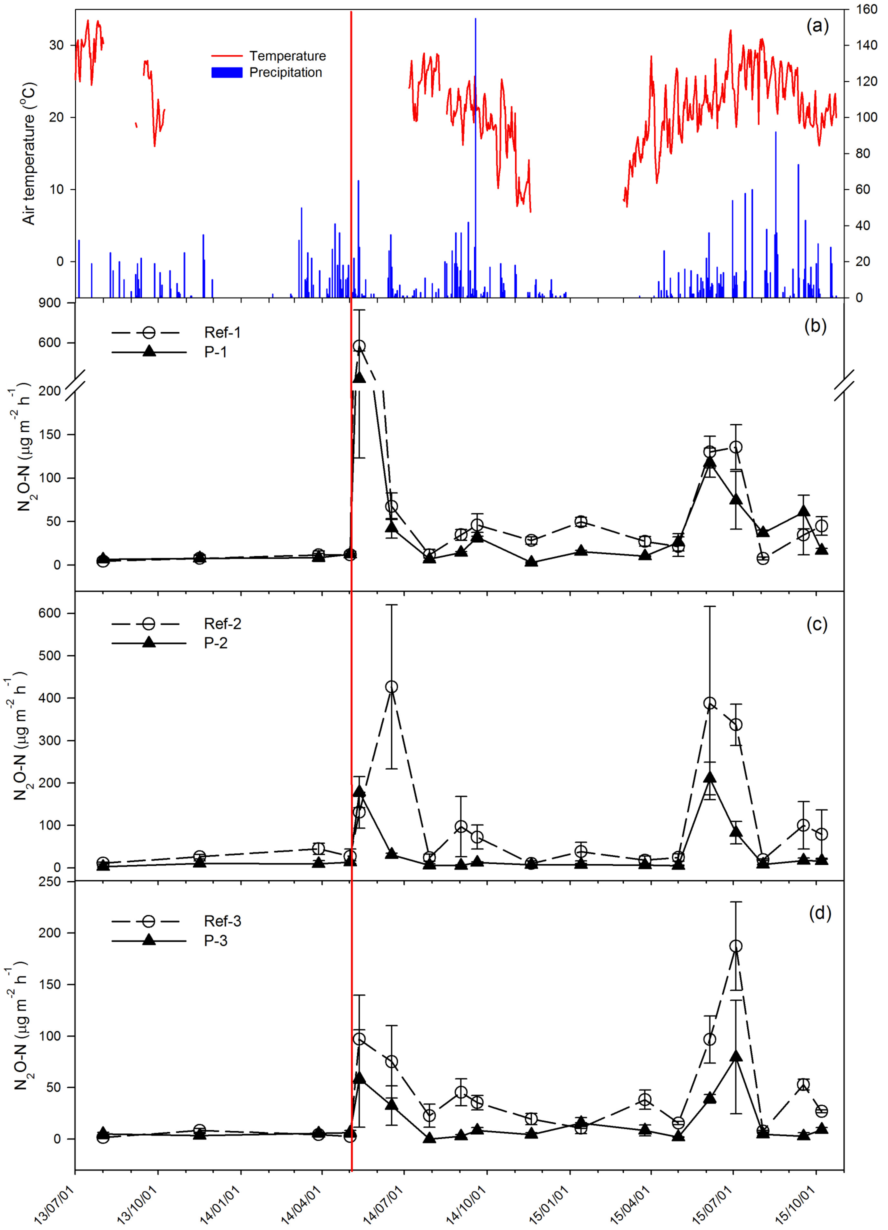

Figure 2. Daily mean air temperature and precipitation **(a)**, and monthly mean N_2O fluxes (±SE) in the references (Ref) and P treatments in each of the three blocks **(b–d)**; the red vertical line gives the date of P addition (4 May 2014).

amine the treatment effects on soil pH, nutrient contents in organic matter, tree biomass, 500-needle weight and needle nutrient content for each sampling. Significance levels were set to $p < 0.05$, if not specified otherwise.

3 Results

3.1 Nutrient concentrations in soil and soil water

Addition of P resulted in a significant increase in soil P content in the O–A horizon, both as P_{Al} and total P (Table 2). However, after 15 months, only P_{Al} indicated an enhanced P status, while total soil P did not differ significantly from background values at the reference sites. P addition had no significant effect on soil pH, or soil C and N content. The NO_3^- concentration in soil water collected at 5 cm depth varied seasonally, with significantly greater values (30–40 mg N L^{-1}) towards the start of the growing season in 2015 (April, Fig. S2), but not in 2014, likely due to dilution by abundant precipitation in February to March 2014. Addition of P resulted in significantly smaller NO_3^- concentrations in soil water at both 5 and 20 cm depths (Fig. 1b).

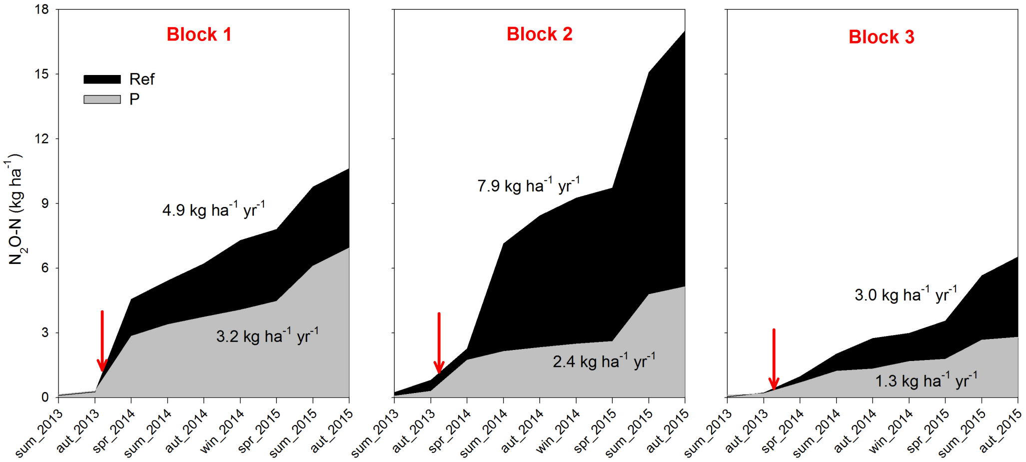

Figure 3. Cumulative N_2O emissions for three blocks in the references (Ref) and P treatments from summer 2013 to autumn 2015; the red arrows refer to the date of P addition (4 May 2014).

In general, the concentration of NH_4^+ in soil water was small ($< 0.5\,mg\,N\,L^{-1}$) and not affected by P addition (Fig. 1a). At both depths, mean soil water concentrations of Mg^{2+} and Ca^{2+} were significantly smaller in the P treated than the reference plots, and the overall cationic charge declined significantly in response to P addition (Fig. S3).

3.2 N_2O and CH_4 fluxes: effects of P addition

In the reference plots, N_2O fluxes varied seasonally (Fig. 2), showing a significant relationship with daily precipitation (Fig. S4a), but not with daily mean temperature (Fig. S4b). Mean N_2O fluxes were generally below $50\,\mu g\,N_2O–N\,m^{-2}\,h^{-1}$ in the dry, cool season, but reached values of up to $600\,\mu g\,N_2O–N\,m^{-2}\,h^{-1}$ in the growing season (Fig. 2). Cumulative N_2O emissions were estimated with seasonally averaged fluxes, and they differed greatly among the three blocks (Fig. 3), of which block 2 had the greatest annual N_2O emission ($7.9\,kg\,N\,ha^{-1}$). CH_4 fluxes in the reference plots also varied greatly among blocks (Fig. 5). Net emission of CH_4 was observed in summer 2013 ($\sim 80\,\mu g\,CH_4–C\,m^{-2}\,h^{-1}$) in blocks 1 and 2, whereas block 3 showed net uptake. From spring 2014 until October 2015, CH_4 fluxes were less variable in all blocks, with values fluctuating around zero. A longer period of net emission was observed in block 3 during the dry season 2014. The fluxes did not correlate with precipitation or air temperature (Fig. S5c, d).

Mean N_2O fluxes during the 1.5 years after P addition were significantly smaller in the P treatment than in the reference (Fig. 4). The P addition resulted in a 50 % ($3\,kg\,N_2O–N\,ha^{-1}\,yr^{-1}$ on average) reduction of cumulative N_2O emission (Fig. 3). No immediate effect (within days) of P addition on N_2O emission was observed (Fig. S5).

In the 1.5 years following P addition, mean CH_4 fluxes indicated net CH_4 emission ($\sim 3.8\,\mu g\,CH_4–C\,m^{-2}\,h^{-1}$) in the reference, whereas net CH_4 uptake ($\sim 6.5\,\mu g\,CH_4–C\,m^{-2}\,h^{-1}$) was observed in the P treatment (Fig. 6). The suppressing effect of P addition on CH_4 emission was significant, in accordance with what was found for NO_3^- concentration and N_2O emission.

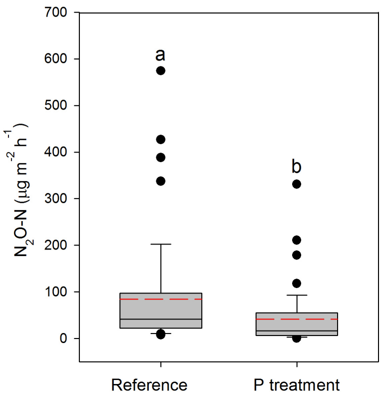

Figure 4. Box-and-whisker plots for N_2O fluxes in the reference and P treatment throughout 1.5 years after the P addition; red dashed lines indicate mean values; linear mixed-effect models were used to test the P treatment effect; different letters indicate significant difference ($p < 0.05$).

3.3 The effect of P addition on tree growth

Throughout the 2-year experimental period, we observed no significant change in tree biomass in response to P addition (Table S3). Likewise, there was no effect of P treatment on the 500-needle weight. Between the two samplings in 2013 and 2014, we found differences in chemical composition of the pine needles, but the difference between the reference and P treatment was not significant. Also, the C / N and N / P ratios of the needles (40 and 16, respectively) were not affected

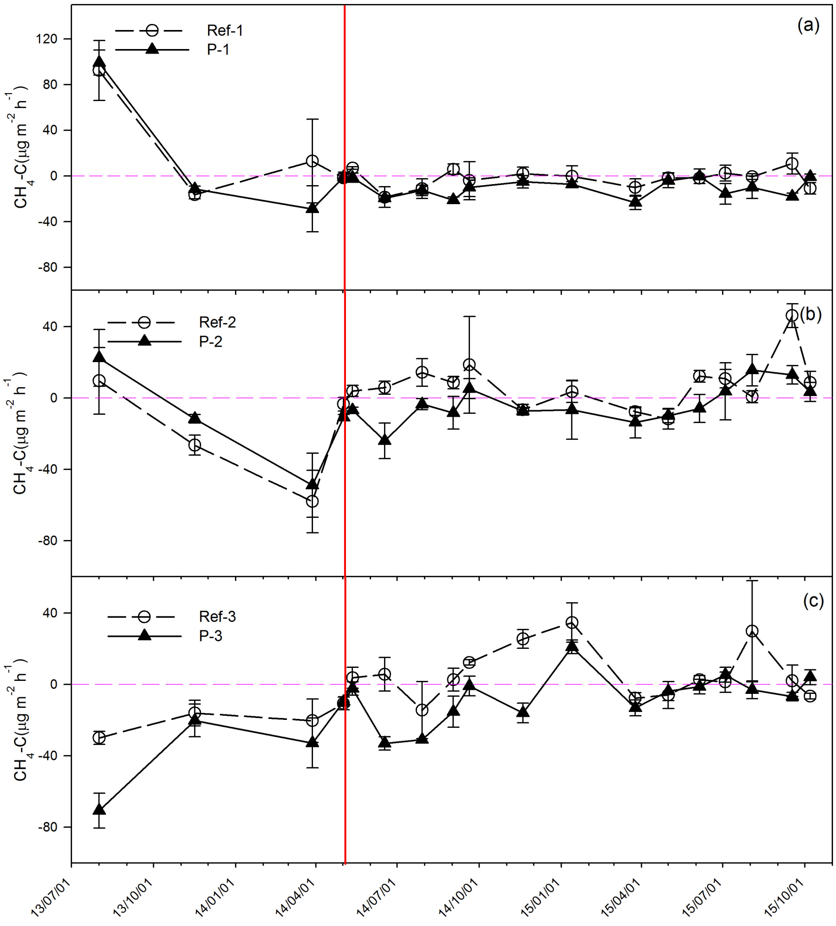

Figure 5. Monthly mean CH_4 fluxes (±SE) in the references (Ref) and P treatments for three blocks **(a–c)**; the horizontal broken line indicates zero flux the red vertical line refers to the date of P addition (4 May 2014).

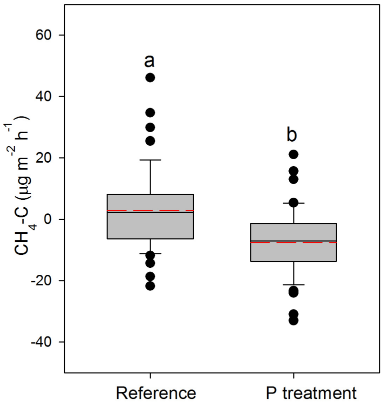

Figure 6. Box-and-whisker plots of CH_4 fluxes in the reference and P treatment throughout 1.5 years after the P addition; red dash lines indicate mean values; linear mixed-effect models were used to test the P treatment effect; the different letters indicate significant difference ($p < 0.05$).

by P addition. Monthly litterfall varied seasonally in both reference and P treatment (Fig. S6), but no significant difference was found between the two treatments.

4 Discussion

N_2O emission rates in the reference plots were relatively large (Fig. 2), with mean values close to 100 µg N_2O–N m^{-2} h^{-1} (Fig. 4). This is within the range of N_2O emission rates previously reported for well-drained hillslope soils at TSP (Zhu et al., 2013b), but greater than the rates reported for other forests in southern China. For instance, N_2O emission rates averaged to 37 µg N_2O–N m^{-2} h^{-1} in unmanaged sites at Dinghushan (Fang et al., 2009; Tang et al., 2006) and 50 µg N_2O–N m^{-2} h^{-1} in N-fertilised sites (Zhang et al., 2008a). TSP reference plots emitted on average 5.3 kg N_2O–N ha^{-1} yr^{-1} (Fig. 3), which is about 10 % of the annual N deposition (50 kg N ha^{-1} yr^{-1}) (Huang et al., 2015). These fluxes are well above average fluxes reported for tropical rainforests (Werner et al., 2007). Large N_2O emissions at TSP are likely due to the large N deposition rates (Huang et al., 2015). A similar trend of increasing N_2O emissions with increasing N deposition rates has been reported for a wide range of ecosystems (Liu et al., 2009). Also, warm–humid conditions during monsoonal summers may stimulate N_2O emissions (Ju et al., 2011), as monsoonal rainstorms trigger peak fluxes (Pan et al., 2003). The positive correlation between precipitation and N_2O emission peaks (Fig. S4a) may indicate the importance of denitrification as the dominant N_2O source. This is supported by recent ^{15}N tracing experiments at TSP (Yu et al., 2017; Zhu et al., 2013a).

Addition of P caused a significant decline in soil inorganic N in soil water (predominantly NO_3^-; Fig. 2), particularly during summers, when NO_3^- concentrations were relatively large (Fig. S2). At the same time, annual N_2O emissions decreased by more than 50 % (Figs. 3 and 4). These findings are consistent with a number of previous studies (Baral et al., 2014; Hall and Matson, 1999; Mori et al., 2014), which attributed the reduction of N_2O emissions in P-treated soils to decreased NO_3^- availability and thus less denitrification. The attenuation of soil NO_3^- by P addition at TSP may reflect stimulated N uptake by plants and/or soil microorganisms. In a similarly N-rich, tropical forest in southern China, Chen et al. (2016) reported a stimulation of net N mineralisation and nitrification after 6 years of bi-monthly P addition, despite reduced soil NO_3^- concentration. Therefore, it is likely that plant uptake plays a more important role in P-induced N retention than immobilisation by soil microbes. However, during our study period of 2 years, we did not find significant increase of N uptake based on tree biomass and foliar N content measurements (Table S3). An alternative explanation could be that P addition stimulated of N uptake by ground vegetation, which remains to be confirmed.

In contrast to our study, P-addition experiments in southern Ecuador (Martinson et al., 2013) and southern China (at Dinghushan Biosphere Reserve (Zheng et al., 2016) found no effect of a single P addition on N_2O emission during the first 2 years after application. However, significant reduction in N_2O emission was observed after 3 to 5 years of continuous P addition, both at the Ecuadorian and the Chinese sites (Chen et al., 2016; Müller et al., 2015). For the montane forest site in Ecuador, the observed delay in N_2O emission response to P addition may be explained by the relatively low ambient N deposition ($\sim 10\,\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$) and small N_2O fluxes ($\sim 0.36\,\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$ in the reference plots) (Martinson et al., 2013; Müller et al., 2015). In addition, the moderate amount of P added ($10\,\mathrm{kg\,P\,ha^{-1}\,yr^{-1}}$; Martinson et al., 2013) could have resulted in an insignificant P effect in the first 2 years. The Dinghushan site in southern China receives $36\,\mathrm{kg}$ inorganic $\mathrm{N\,ha^{-1}\,yr^{-1}}$ by throughfall (Chen et al., 2016; Fang et al., 2008), which is similar to the inorganic N deposition at TSP (Chen and Mulder, 2007b; Huang et al., 2015). However, soil KCl-extractable inorganic N ($\sim 40\,\mathrm{mg\,N\,kg^{-1}}$; Zheng et al., 2016) and NO_3^- leaching ($\sim 20\,\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$; Fang et al., 2008) at the Dinghushan site are several-fold smaller than at our site ($\sim 100\,\mathrm{mg\,N\,kg^{-1}}$ and $\sim 50\,\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$, respectively) (Huang et al., 2015; Zhu et al., 2013b). Also, the mean N_2O emission rates in the reference plots ($10\,\mathrm{\mu g\,m^{-2}\,h^{-1}}$) at Dinghushan were smaller than at TSP ($> 50\,\mathrm{\mu g\,m^{-2}\,h^{-1}}$; Fig. 4). These indicate that the Dinghushan forest has stronger N assimilation and is thus less N rich than TSP forest. Therefore, we suggest that the response of N_2O emission to P addition may depend on the N status of the soil. The fact that numerous studies found apparent suppression of N_2O emission in short-term experiments (< 2 years) in N + P treatments, but not in treatments with P alone, supports this idea (Müller et al., 2015; Zhang et al., 2014b; Zheng et al., 2016).

Another study in southern China reported increased N_2O emissions during 2 years after P addition, in a secondary mixed forest (Wang et al., 2014). While suppression of N_2O emission by P has been attributed to increased plant N uptake (Mori et al., 2014), increased N_2O emission is generally explained by enhanced microbial growth (Liu et al., 2012) and denitrification activity (Ehlers et al., 2010; He and Dijkstra, 2015). P stimulation of N_2O emission by microbial denitrification should be rather fast, as indicated by Mori et al. (2013c) in a short-term (1 week) incubation experiment with soils from an *Acacia mangium* plantation. Unlike Mori et al. (2013c), we did not find increased N_2O emissions within a week after P addition at our site (Fig. S5). This may suggest that the denitrifier community at TSP was not responsive to the P applied, probably because TSP hillslope soils have large denitrification potentials (Zhu et al., 2013c).

The reference plots at TSP showed net CH_4 emission for extended periods (Figs. 5 and 6). Also, long-term CH_4 fluxes sampled between 2012 and 2014 on hillslope soils near-by (Fig. S7; Zhu et al., unpublished data) showed net CH_4 emission. This is in contrast to the generally reported CH_4 sink function of forested upland soils (Ciais et al., 2013; Dutaur and Verchot, 2007). For example, net CH_4 fluxes reported for well-drained, forest soils in southern China range from -30 to $-60\,\mathrm{\mu g\,CH_4\text{–}C\,m^{-2}\,h^{-1}}$ (Fang et al., 2009; Tang et al., 2006; Zhang et al., 2014a). Since aerated upland soils typically provide favourable conditions for microbial CH_4 uptake (Le Mer and Roger, 2010), the net emission observed in our sites is unlikely to be due to enhanced CH_4 production, but rather due to suppressed CH_4 consumption. One general explanation for the net CH_4 emission at TSP could be inhibition of CH_4 oxidation by NH_4^+, which competes with CH_4 for the active site at the methane monooxygenase enzyme (Bodelier and Laanbroek, 2004; Zhang et al., 2014a). The concentration of NH_4^+ in the soil water was rather small ($< 0.5\,\mathrm{g\,L^{-1}}$; Fig. 1), which does not preclude, however, that NH_4^+ availability from the soil exchangeable pool may have been high. Zhu et al. (2013b) found extraordinarily high KCL-extractable NH_4^+ in TSP surface soils, likely reflecting the large atmogenic NH_4^+ input at the TSP site (Huang et al., 2015). On the other hand, Reay and Nedwell (2004) found that NO_3^- inhibits methanotrophic activity in acidic soils, where NH_3 is scarce. Possible mechanisms are the toxicity of denitrification intermediates (e.g. NO_2^-; Wang and Ineson, 2003) and the osmotic effect of high-NO_3^- concentration (Hütsch et al., 1996). This deduction can be supported by the high-NO_3^- concentration in the acidic soils at TSP (Figs. 1 and S2).

P addition had a significant impact on CH_4 fluxes, changing the soil from a net source to a net sink on an annual basis (Fig. 6). However, the uptake rates of CH_4 in the P treatments remained smaller than those reported for forest soils in tropical China (Tang et al., 2006; Zhang et al., 2008b). The stimulating effect of P addition on CH_4 uptake is consistent with previous studies (Mori et al., 2013a, b; Zhang et al., 2011), and has been attributed to alleviating N inhibition of methane oxidation. P addition may also result in a change of the taxonomic composition of the methane oxidising community (Mori et al., 2013a; Veraart et al., 2015). Alternatively, CH_4 oxidation may be stimulated by increased CH_4 diffusion into the soil, due to enhanced root growth and increased soil water loss due to transpiration in P-amended plots (Zhang et al., 2011). Given the strong N enrichment of TSP forest (Huang et al., 2015), it is likely that the reason for the observed reduction in CH_4 emissions in response to P fertilisation is due to alleviating direct NH_4^+ inhibition of methane monooxygenase (Veldkamp et al., 2013), rather than due to P stimulation of methanotrophic activity (Veraart et al., 2015).

Shortly after fertiliser application, we observed a modest, albeit significant, increase of Na^+ concentration in soil water (Table S2). Other studies have documented the potential toxicity of excess Na^+ in soil water to plant and microbial activities (Rengasamy et al., 2003; Wong et al., 2008). However, Na^+ toxicity to a degree affecting N turnover processes in our plots is unlikely, as Na^+ concentrations in soil water, within 1 month after application (Table S2), did not exceed $5\,\mathrm{mg\,L^{-1}}$, which is far smaller than the values commonly assumed to cause toxicity (40 to $100\,\mathrm{mg\,L^{-1}}$) (Bernstein, 1975). Frequent precipitation at TSP (Yu et al., 2016), both prior and following the addition of $NaH_2PO_4 \cdot 2H_2O$ (Fig. 2), may have diluted and leached Na^+, thus preventing toxic effects.

P application significantly increased plant-available P in the P-limited TSP soil (Table 2). Meanwhile, concentrations of leachable base cations (K^+, Mg^{2+}, Ca^{2+}) in soil water decreased (Fig. S3), as expected from the reduction of NO_3^- concentrations in the P treatments, which represent a major decline in mobile anions in the P-treated soils (Mochoge and Beese, 1986). We observed no sign of stimulated forest growth or increased N uptake by trees within the relatively short period of our study (Table S3 and Fig. S6), making it difficult to link the observed reduction in inorganic N in the soil solution (Fig. 1) to plant growth. When interpreting the observed P effect on NO_3^- concentrations in soil water, several aspects need to be considered. First, 2 years of observation may be too short to detect any significant increase in tree growth, due to NO_3^- uptake, given the commonly large variabilities in tree biomass estimates (Alvarez-Clare et al., 2013; Huang et al., 2015). Second, a significant proportion of the added P, and of excess N, may have been assimilated by the understorey vegetation, which was not assessed in this study. Previously, understorey biomass has been reported to quickly respond to P addition (Fraterrigo et al., 2011). Third, as long-term N saturation and acidification at TSP have reduced forest health (Lu et al., 2010; Wang et al., 2007), we may not expect immediate response of forest growth to P addition. Large needle N / P ratios (17–22, Table S3) indicated that P limitation for tree growth was not relieved 1.5 years after P addition (Li et al., 2016). Therefore, enhanced N uptake by understorey growth may have been the main mechanisms responsible for observed NO_3^- decline in the P-treated soil (Hall and Matson 1999).

Our study suggests that N-saturated TSP soils act as a regional hotspot for N_2O (Zhu et al., 2013b) and CH_4 emissions. Within the short experimental period of 1.5 years, P fertilisation was shown to significantly decrease NO_3^- concentrations in soil water and to overall reduce N_2O and CH_4 emissions. These findings provide a promising starting point for improving forest management towards greenhouse gas abatement targets, taking into account the P and N status of subtropical soils in the region.

Data availability. Data of gas fluxes, soil water chemistry and forest growth can be found in the Supplement, document 2.

The Supplement related to this article is available online at https://doi.org/10.5194/bg-14-3097-2017-supplement.

Competing interests. The authors declare that they have no conflict of interest.

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