Dear Editor,

Thank you for considering the second revision of our manuscript "**Phosphorus addition mitigates N_2O and CH_4 emissions in N-saturated subtropical forest, SW China**" (bg-2016-470) by Yu et al., for publication in Biogeosciences.

We have revised our manuscript (R2), based on the reviewers' comments. In the point-to-point response, the changes refer to the line numbers in the manuscript version with marked-up changes (attached following the response).

We thank you again for handling our manuscript.

Best regards,
Longfei Yu on behalf of all coauthors
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Comments from Reviewer #2

1) Scientific Significance:

The primary objective of this study is to determine how phosphorus availability influences the efflux of N2O and CH4 in a nitrogen-saturated pine-dominated subtropical forest. While we have a fair understanding how the availability of nitrogen influences these processes, we have a very poor understanding how phosphorus can directly, or indirectly influence greenhouse gas production. Prior research on this subject has been inconclusive and this study will help add to the body of literature to advance our understanding of soil nutrient availability and climate forcing.

2) Scientific Quality:

Much improve from the early versions – mixed model is the way to go. My primary concern is the low replication (just three 20 x 20 m plots for each treatment) and perhaps overall discussion needs to be tempered.

R: We thank the reviewer for the critical but constructive comment. Our study explores the effect of phosphorus (P) on N cycling in the N-saturated Tieshanping (TSP) forest (Huang et al., 2015). Indeed, our field observations indicate that P application significantly reduces N_2O and CH_4 emissions from upland soils at TSP. We agree that the current field set-up has its limitation and that our findings cannot be readily scaled up. For this, further research is needed, including work at other sites. The experiment was done in triplicate, as more replicates were not feasible given the available man-power and budget. In terms of replication, our experiment, investigating GHG emissions, does not differ much from others, which also used 20 x 20 m^2 plots in triplicates (Muller et al., 2015; Martinson et al., 2013) or even 10 x 10 m^2 plots in triplicates (Zheng et al., 2016; Wang et al., 2014) for each treatment.

3) Presentation Quality:

The discussion needs minor revisions. A fair amount of the discussion is just a summary of the results with modest interpretation or explanation. While it is fine to report how these results are similar/dissimilar with other work, it needs to go beyond that and provide new insights and tieback with the present study. What do the result consistent with this study have in common, but uncommon with work that is inconsistent? More effort is needed in the discussion to explain the 'why' of the results. Certain interesting passages need to be developed (e.g. lines 313-315)

R: We thank the reviewer for the general suggestion. In the revised version, we now expand our explanation of the results. Major changes are made in three sections of the discussion:

1) P effect on soil NO_3^- concentration (line 301-310): we attribute the attenuation of soil NO_3^- concentration to P stimulation of N uptake by plants and soil microorganisms. Based on the reported increase of N mineralization and nitrification rates in a similar site from South China, we suggest that microbial uptake of N is less likely than the plant uptake. Although this is not supported by our measurement of tree biomass and foliar N during two years, we provide an alternative explanation, which is N uptake by understory vegetation.

2) Comparison of P effect on N_2O emission with other studies (line 324-335): In comparison with TSP, the slow response of N_2O emission to P addition at the Ecuadorian site is suggested to be a result of both low ambient N deposition and low P dose. The Chinese site (Dinghushan) receives similar inorganic N input via throughfall as TSP. But we suggest the Dinghushan site to be less N-rich than TSP, based on the lower KCl-extractable inorganic N content in soil and lower flux of N leaching at Dinghushan. This point is now clearly made in the discussion.

3) We improve our explanation of N inhibition effects on CH_4 uptake by elaborating on known CH_4/NH_4^+ substrate competition for methane monooxygenase (line 372-373) and NO_3^-/NO_2^- toxicity for methanotrophs (line 377-382). In the discussion of P effect on CH_4 uptake, we focus mainly on the direct

P effects on methantrophs and indirect P effects by reducing inorganic N levels in soils (line 386-398). The indirect mechanism seems more likely, as support by our observation.

Line 363: I question that the study actual demonstrates that high deposition has shifted these soils to regional hotspots for N2O and CH4 production – this was not tested. Also, this is difficult to say without historical data or somehow experimental removing/reducing N deposition. Yes, adding P has reduced NO3 concentration, but this result is fairly modest, especially at 20 cm.

R: We agree with the referee that historic data on the relationship between atmogenic N deposition and N concentrations in soil water as well as emission rates of N_2O and CH_4 would be needed, to ultimately prove the connection between elevated N deposition and N_2O and CH_4 emissions in the sub-tropical forest region. As such data do not exist for TSP, our conclusions are solely based on comparison with similar research sites and on manipulation experiments at TSP. For example, Huang et al (2015) showed that a doubling of N deposition at TSP caused a doubling of N leaching (as nitrate), simultaneously in increasing N_2O emission (Liu WJ, personal communication). Our manuscript adds further evidence to this, as we find that P addition causes a significant decline in NO_3^- concentrations in soil water in the surface horizons (O/A and AB; Fig. 1), while emission rates of N_2O and CH_4 decrease (Figs. 4 and 6). Also other studies have reported strong correlation between NO_3^- concentration in soil water and N_2O emission rates (Gundersen et al., 2012).

Earlier studies at the TSP site indicate chronically elevated N deposition levels, high inorganic N concentration in soil water and strong soil acidification (Zhu et al., 2013; Larssen et al., 2011; Huang et al., 2015). These factors contribute to enhanced N_2O emission and reduced CH_4 uptake in soils (Liu et al., 2010; Le Mer and Roger, 2010). Among other studies in Southern Chinese forests (Tang et al., 2006, Fang et al., 2009; Zhang et al., 2008), our study reports the highest N_2O fluxes and lowest CH_4 uptake rates (or even net emission).

In addition, our recent ^{15}N tracer study at TSP (Yu et al., 2017), has shown that the enriched ^{15}N signals in emitted N_2O is identical to those in soil NO_3^- extracted from O/A horizon rather than AB horizon. This denotes the importance of NO_3^- availability in surface soils to N_2O emission. Also for the CH_4 oxidation, the N inhibition effect should be most important in surface soil (Bodelier and Laanbroek, 2004), where methanotrophic activity is more active with more aerobic condition. Therefore, our observation of significant decreases in NO_3^- concentration from O/A and AB horizons do support that P addition migrates the N_2O and CH_4 emissions from soil.

Comments from Reviewer #3

The manuscript by Longfei Yu and others presents a replicated forest fertilization experiment in an acidified and N-saturated Masson pine-dominated forest at TieShanPing, SW China. The experiment tests the role of mineral P fertilization in regulating nitrous oxide and methane emissions (uptake). Researchers measured soil water NO3- concentrations, N2O emissions, CH4 emissions (uptake), forest productivity, litter fall, litter chemistry, and soil characteristics prior to and after a one-time fertilization with 79 kg P ha-1 (NaH2PO4).

The Author's found that P fertilization results in declines in soil NO3- and suppressed emissions of N2O, and CH4 (not immediately, but over the long term). With P addition TSP soils switched from a CH4 source to a sink. Elevated biomass production was not observed over the 18-month experimental period. However, understory biomass was not assessed. Based on these results, Authors hypothesized that P additions resulted in increase NO3- uptake by plants and microbes leaving less for denitrification. Also, P addition was thought to lessen the NH4+ inhibition of methane oxidation.

Overall comments:

This manuscript is well written and presents a topic that is of interest generally to the readers of Biogeosciences. I have a couple of concerns that should be addressed prior to publication. First, I feel that these results and their interpretation would be easier to follow if there were a set of explicitly stated hypotheses. There is one hypothesis stated in the Abstract (that concerns the results), but not in the main body of the manuscript.

R: Following the reviewer's recommendation, we have changed our objectives to more explicit hypotheses. We now present three key hypotheses: 1) P addition stimulates tree growth; 2) P addition decreases soil inorganic N availability and 3) P addition reduces N_2O and CH_4 emissions (line 105-109).

Regarding the description of the experimental design and sampling, the description in the methods doesn't seem to reflect the data that is presented for N2O and CH4 (L167-179). From the methods, I gather that N2O and CH4 were measured a total of four times, but clearly more data points are presented. Please clarify in the text how frequently measurements were made over the entire experiment. Other clarifying points are made below in the line-by-line comments.

R: Thanks for the suggestion. To avoid confusion, we now describe the frequency of gas emission sampling together with the soil pore water sampling. In line 178-180, we add "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". In line 181, we put "also" before the description of "short-term" intensive sampling.

With the Results, at times the text is confusing because the treatment effects of P additions and the seasonal/temporal patterns are explained simultaneously. I would recommend some minor reorganizing of this information. Perhaps start with the overall seasonal patterns and then state the treatment effects or the opposite.

R: We appreciate reviewer #3's advice. We have now reorganized the results of N_2O and CH_4 fluxes, by starting with a paragraph for seasonal/temporal patterns of fluxes from Reference plots (line 243-255). The P effect on N_2O and CH_4 fluxes are then presented in another two paragraphs.

Line-by-line comments:
L16: Change GHG to green house gas
R: OK.

L18: If this is a single fertilizer event is it necessary to have the unit yr-1?
R: We have revised it as "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)" (line 17-19).

L20: Rephrase this sentence to read "We observed a significant decline in soil water NO3- concentrations (5 and 20 cm depths) and in soil N2O emissions following P addition."
L21: It is unclear if this number is the amount of reduction or if it represents the total emission. Please clarify
R: Agree. This number for emission reduction is now deleted. See line 20-22.

L23-24: The "As for N2O" is a confusing way to begin this sentence. Can you revise to something like "P addition significantly decreased CH4 emissions, turning TSP soils from a net source to a net sink." I'm sure the Authors will have a more eloquent way of conveying that message.
R: It is now changed to "P addition significantly decreased CH_4 emissions and turned the soil from a net source into a net sink" (line 24-26)

L26-27: It's my preference to put this caveat in the discussion or that it's rephrased. The current wording suggests that you measured understory and that there was an increase in understory biomass.
R: This sentence is now rephrased, as "Within the 1.5 years after P addition, no significant increase of forest growth was observed and P stimulation of forest N uptake by understory vegetation remains to be confirmed" (line 27-30)

L48-49: 'frequently shifting aerobic conditions' is awkward please revise. Perhaps this is better put in terms of aerobic and anaerobic?
R: OK (line 50-53).

P4 L56: Consider changing 'mineral' to 'inorganic'
R: Done.

L100: The hypothesis is stated in the abstract but not the main text. Please include in text prior to the objectives.
R: Please refer to our earlier reply to the overall comments. See also line 105-109.

L105: It's unclear why the study site name is in quotations. L116: TSP hilltop is not intuitive. Please explain in text
R: The quotation is removed now. "TSP hilltop" refers to upland soils from the hillslopes at TSP, which has been previously documented by Zhu et al., 2013b. Here, we rephrase "at TSP hilltop" as "on the hillslopes" (line 123-124).
L121-122: Can you state over what time period the decline in growth has occurred here?

R: Wang et al. (2007) and Li et al. (2014) reported the decline in forest growth at TSP during 2001-2004 and 2004-2012, respectively. So we rephrase the sentence as “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)” (line 129-131).

L128: Rather than an *, please use ×
R: OK. Change is made in line 136.

L141: Can you report the Na+ concentrations of the Reference plots? L157: Change (2 mm) to (2mm × 2mm)
R: We add “(0.52-1.31 mg L^{-1})” for the reference plots (line 149). More specific values for different soil depths could be found in Table S2 of the support information.

L165: I think part of the instrument name is missing. Should this be ‘inductively coupled plasma atomic emission spectroscopy’? For all makes/models of equipment here and throughout, please add the location information.
R: OK. See line 174-175.

L171: Change to “...into 12 mL pre-evacuated glass vials... (Chromacol, UK).” L172: I would recommend splitting this into another sentence: “Vials were over pressurized to avoid contamination during sample transport.”
R: It is modified as “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”, as shown in line 182-184.

L173: Is ‘Mixing ratio’ what you mean or should this be ‘Fluxes of ...’ or ‘Concentrations of...’
R: Yes. Mixing ratio is the direct result that we obtain from gas analysis on gas chromatograph coupled with ECD. Mixing ratios of N_2O and CH_4 are commonly expressed as ppb or ppm, similar to “concentration”. For fluxes, we need further calculation based on the change of gas mixing ratio with time, as described later in the same paragraph.

L190: Please specify if the same trees were measured at each time point, this is critical to the interpretation of these data.
R: Yes, they had been marked since the first measurement. This is now explained (line 204).

L194: Rather than ‘sum of precipitation’ can this be termed ‘daily total precipitation’? Please provide the time period over which precipitation and temperature were measured.
R: OK (line 207-208).

L198: I gather from the methods that gas samples were collected from August 2013 forward, but only during the month of May (2, 7, 10, and 12). This doesn’t reflect all of the data points that are shown in Figures 2 and 5. I would insist that the Authors add clarity to the methods or only show data that were collected in this study.

R: Please refer to the earlier reply to the overall comments of the reviewer. Changes are made accordingly in line 178-180.

L206: It is unclear if fluxes of 'litterfall' nutrients were scaled to the biomass production. Or was litter biomass a component of the overall biomass calculation? L210-213: For tree growth, how were the 3 different time points treated? Please be specific.
R: The fluxes of litterfall were not used for tree biomass evaluation. Instead, we used allometric models (specifically for masson pine; Li et al. 2011 and Zeng et al. 2008) to estimate the tree biomass, based on measured diameters at breast height (DBH). We specify now that DBH is used for tree biomass estimation (line 205).
The data for tree growth refer to Table S3, which includes tree biomass, 500-needle weight and needle nutrient contents. The three samplings were tested for treatment effect separately with one-way ANOVA (line 223-226).

L226: The phrase 'sum of charge of dissolved base cations is unclear', at any rate, it would be more appropriate to say that charge was significantly different between fertilized and unfertilized. I am curious if the 'charge' decreases in the P treatment because of the increase of Na+. Can you please address?
R: We thank the reviewer for the suggestion. This phrase is changed to "the overall cationic charge" (line 240-241). The increase in total cationic charge due to Na^+ addition should be modest, while the charge of dissolved of Ca^{2+} is far more important. P addition stimulated the uptake of N, thus resulting in a decline in soil NO_3^- concentration. As NO_3^- is the major anion in soil water, the decrease of NO_3^- concentration leads to a decline in cation concentration (Figure S3). Thus, the observed decrease in cationic charge is a direct effect of the decline in mobile anions (line 409-411).

L232-234: Please report the block effect here. L240: Was there a significant block effect that could be reported here?
R: The block effects were clearly described in the sentences that follow, in line 247-249 for N_2O and in line 250-254 for CH_4.

L236-238: Rephrase to read: The P addition resulted in a 50% (average 3 kg N ha-1 yr-1) reduction of cumulative N2O emissions (Fig. 3). Please add +/- Stderror if it is available
R: We have revised it to "The P addition resulted in a 50% (3 kg N ha^{-1} yr^{-1} on average) reduction of cumulative N_2O emission (Fig. 3)" (line 257-259).

L238: Change was to were.
R: "effects" is changed to "effect" (line 259).

L245: Should this unit be CH4-C here and throughout? Also can you add +/- Stderror here?
R: The units for N_2O and CH_4 fluxes have been checked throughout the manuscript and corrected as g N_2O-N m^{-2} hr^{-1} and g CH_4-C m^{-2} hr^{-1}, respectively. For annual emission, they are described as kg N_2O-N ha^{-1} yr^{-1} and kg CH_4-C ha^{-1} yr^{-1}. Here, the CH_4 flux data show a skewed distribution, thus having large standard deviations. Since the treatment effect on CH_4 fluxes had been tested for significance with mixed-effect models, it is not necessary to include standard error here.

L250: What does 138 t ha-1 represent? Is it an average across both years and both treatments? I'm not sure how informative that is. Based on your supplemental data, it looks as if biomass was actually lower in the P addition treatment compared to the Reference treatment.
R: Agree. This value denotes the mean for both years and in both treatments. Based on statistical results, P treatment exhibited no difference from the reference. In the text, we have now described the change as "insignificant" and deleted the value for tree biomass (line 271-272).

L252: The 500g needle weight does not need to be reported here.
R: Agree.

L253: This sentence needs to be clarified to indicate the mechanism responsible for differences in needle chemical composition. "Linked" is vague.
R: It is rephrased as "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" (line 273-275).

L253: 'hardly' is a vague word, please replace. L273: Change mineral to inorganic
R: OK. "hardly" is changed to "not" now (line 276).

L275-279: This sentence is complex and confusing. Please revise, as it seems to contradict your former statement.
R: OK. We have simplified our discussion, with additional details for support. "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." (299-303).

L273: This paragraph is long and difficult to follow. I believe that the Authors could find a way to make it more streamlined and easier to follow.
R: See the next comment.

L283-292: I think it would be better to put your study into context of others that used similar additions. Perhaps the reference to moderate P additions is a bit of a distraction. I would recommend revising to focus on more similar studies.
R: This paragraph has now been divided into two paragraphs. The first one has been rephrased and focuses only on the key points of our experiment (296-310). The second one compares the fertilization effects of N, P and N+P on N_2O emissions among forests with different N status, involving a space-for-time comparison. While our main discussion still addresses the comparison to similar studies in Southern China, we have modified the comparison with the Ecuadorian study, and focus on different responses of N_2O emissions to P addition in forests with different N status (315-345). See more details in our response to comments from Reviewer #2.

L306: Likewise, the point of this paragraph is not entirely clear. As well, it is unclear if the referenced studies are also covering the short-term (~10day) span of time that is referenced in this manuscript.

R: To make it more concise, we have rewritten this paragraph, starting with studies that found that P addition increases N_2O emission. Then we discuss the possible mechanisms for P stimulation of N_2O emissions, and emphasize that the P effect on denitrifier activity could be rather fast (Mori et al., 2013c). However, we did not see any P effect on N_2O emission from the intensive observation shortly after P application at TSP. This may be attributed to large denitrification potentials in TSP soils, supported by a lab-incubation study (Zhu et al., 2013c). See the changes in line 346-359.

L324: Change production to 'CH4 production' just to be clear this isn't primary production
R: OK. See line 370.

L353: Change apparently to 'may have'
R: OK. See line 407.

L355: I liked the nice flow and organization of this paragraph!
R: Much appreciated.

L375-376: Is there a citation from your previous work that you can add here? As is, these data don't provide obvious evidence for this.
R: We have added Zhu et al. (2013b) as a reference (line 429). For more details, see our reply to reviewer#2's comments.

L377: Can this statement be qualified by stating 'to overall reduce'
R: Yes, it can. See the changes made in line 430.

L379: GHG is used here and in the abstract, but is not explicitly defined. Please do so. L388: References are not alphabetized consistently.
R: For GHG, please refer to our previous response to the comment on line 16. References have been checked and corrected as requested.

Tables and Figures:
L603: Please change 'Background' to 'Ambient'
R: OK. See line 675.

Table 1. Was 5.0 mg kg-1 the detection limit of the instrument for PH2O? If so, please just indicate this in the footer of the table rather than dedicating an entire column to the < 5.0 information.
R: OK. This is now changed (line 675-681).

Table 2. Here and throughout, please be consistent that the P treatment is '+P'. Also it is unclear what the letters indicate in terms of significance. Should they not indicate significant differences among the Ref and +P treatment? Or is this across all time points? If so, the analysis should more appropriately be a repeated measures analysis.
R: The different letters indicate significant difference between the Reference and P treatment (line 682-685). This is tested by one-way ANOVA for each sampling, which has been specified in statistics (line 223-226).

Figure 4 and Figure 6: The letters indicating significance are somewhat unnecessary here. The point could be made in either the figure legend or with an asterisk centered above the two boxes. In both figures, I would recommend adding the statistical test that you used.
R: We thank the reviewer's suggestion. We would prefer keeping both the letters indicating significance and adding statistical methods to the figure captions. See line 699 and line 707.

Figure S6: Litter is spelled incorrectly in the axis title
R: OK. Revised.

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

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Article type: Research Article

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 ~~GHG~~ greenhouse gas emissions remain ~~understudied~~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} ~~yr^{-1}~~, ~~applied~~ as NaH_2PO_4 powder) to an N-saturated, Masson pine-dominated forest soil at TieShanPing (TSP), Chongqing, SW China. We observed a significant decline in both ~~in~~ NO_3^- concentrations in soil water (~~at~~ 5- and 20-cm depths) and in soil N_2O emissions, following P application~~, the latter by 3 kg N ha^{-1} yr^{-1}~~. We hypothesize that enhanced N uptake by plants ~~and soil microbes~~ in response to P addition, ~~results~~ resulted in less available NO_3^- for denitrification. By contrast to most other forest ecosystems, TSP is a net source of CH_4. ~~As for N_2O,~~ P addition significantly decreased CH_4 emissions~~,~~ and ~~turning~~ 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 ~~at TSP, but~~ and P stimulation of forest N uptake by understory vegetation remains to be confirmed~~we cannot exclude that understory vegetation increased~~. Our study indicates that P fertilization of N-saturated, subtropical forest soils may mitigate~~s~~ N_2O and CH_4 emissions, in addition to alleviating nutrient imbalances and reducing losses of N through NO_3^- leaching.

Key Word: N_2O and CH_4 emission, N saturation, Phosphate fertilization, soil CH_4 uptake, acid forest soil.

1 Introduction

Anthropogenic activities have transformed the terrestrial biosphere into a net source of CH_4, N_2O and 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, 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 South 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~~ly shifting~~ shifts between aerobic and anaerobic conditions in soils ~~aeration conditions~~ during monsoonal summers, promoting ~~both~~ alternating nitrification and denitrification (Zhu et al., 2013b) and ~~to~~ causing 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 ~~constrains~~ limits its capability to retain N (Huang et al., 2015; Li et al., 2016), resulting in ample amounts of ~~mineral~~ inorganic N (NH_4^+ and NO_3^-) being present in the soil solution. Accordingly, Hall & Matson (1999) observed larger N_2O emission in P-limited than in N-limited tropical forests ~~after~~ one year ~~of~~ after repeated ~~N~~ addition of N. Likewise, previous N manipulation studies in forests of South 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 two years. However, N_2O emission was smaller when P was added together with N (50 kg N ha^{-1} yr^{-1}) than in treatments with N addition alone (Martinson et al., 2013). After continued fertilization for three years, also P addition alone reduced N_2O emissions at these sites (Müller et al., 2015). In tropical China, with high N deposition (~ 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 South 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 South 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 oxidize 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 ~~the~~ 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 CH4 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 ~~in soils~~ 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 South 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 ~~mineral~~ inorganic N concentrations for soil-atmosphere exchange of

CH_4, and how this is affected by P fertilization 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 SW China under ambient N deposition and studied the effects of P addition on emission rates, nutrient availability and tree growth. ~~The objectives were i)~~We hypothesized that i) P addition stimulates forest growth; ii) stimulated forest growth results in increased N uptake by trees and understory vegetation, and thus decreases the soil inorganic N concentration; iii) P addition reduces~~to quantify~~ soil N_2O ~~and CH_4~~ emission and promotes CH_4 uptake~~s, ii) to investigate the effect of P addition on N_2O and CH_4 emission~~.

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 northeast 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 ^{o}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. The soil bulk density of the O/A horizon (~ 5 cm) is about 0.75 g cm^{-3}. Generally, ~~S~~soil water-filled pore space (10 cm) ~~at TSP hilltop~~on the hillslopes ~~generally~~ 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 ha^{-1} (dominated by NH_4^+; Yu et al., 2016), while the annual bulk N deposition is from 20 to 30 kg 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 ~~at TSP~~ has been reported to ~~resulted in~~cause severe decline in forest growth at TSP since 2001 (Li et al., 2014; Wang et al., 2007), and ~~in~~ a decrease~~d~~ 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 ~ 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 fertilizer 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 (Supplementary Materials, Table S1 and Figure S1), to ensure significantly increase in soil available P. To apply P fertilizer evenly, we divided each plot into a 5 m * 5 m grid and broadcasted the powdered fertilizer 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 South China.

The addition of $NaH_2PO_4 \cdot 2H_2O$ at the P-treated plots also resulted in an input of 59.0 kg ha^{-1} of sodium (Na). One month after the fertilizer 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~~ 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 ~~strong~~ significant negative impact on plant and microbial activities.

2.3 Sample collection and analyses

153 Within each plot, three ceramic lysimeters (P80; Staatliche Porzellanmanufaktur, Berlin) were
154 installed at 5- and 20-cm soils near the plot centre in August 2013. To obtain water samples,
155 350-ml glass bottles with rubber stoppers were pre-evacuated, using a paddle pump, and
156 connected to the lysimeters for overnight sampling. Between November 2013 and October 2015,
157 we sampled soil pore water bi-monthly in the dry and dormant season and monthly during the
158 growing season. All water samples were kept frozen during storage and transport. Concentrations
159 of NH_4^+, NO_3^-, potassium (K^+), calcium (Ca^{2+}), and magnesium (Mg^{2+}) in soil water were
160 measured at the Research Center for Eco-Environmental Sciences (RCEES), Chinese Academy
161 of Sciences, Beijing, using ion chromatography (DX-120 for cations and DX-500 for anions).

162 In August 2013, soils from the O/A (0-3 cm), AB (3-8 cm) and B (8-20 cm) horizons were
163 sampled near the lysimeters for soil analysis. Total P and plant-available P contents were
164 monitored in samples collected from the O/A horizons every six months, starting two days
165 before P addition. Soil samples were kept cold (< 4 ^oC) during transport and storage. Before
166 analysis, soil samples were air dried and sieved (2 mm). Soil pH was measured in soil
167 suspensions (10 g dry soil and 50 ml deionized water) using a pH meter (PHB-4, Leici, China).
168 Total soil C and N contents were determined on dried and milled samples, using a LECO
169 elemental analyzer (TruSpec$^@$CHN, USA). To measure total P, 1 g dry soil was digested with 5
170 ml of 6 M H_2SO_4 (Singh et al., 2005) and measured as ortho-phosphate by the molybdenum blue
171 method (Murphy and Riley, 1962). Ammonium lactate (0.01 M)-extractable P and H_2O-
172 extractable P (P_{Al} and P_{H2O}, respectively) were measured as ortho-phosphate after extraction (1.5
173 g dry soil in 50 ml solution) (Singh et al., 2005). Ammonium oxalate (0.2 M)-extractable Fe, Al
174 and P were measured by inductively coupled plasma optical emission spectroscopy~~inductive~~

~~coupled plasma~~ (ICP-OES~~7500;~~ , 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 three times (7, 10 and 12 May) after the P application. Gas samples ~~(20 ml)~~ were taken 1, 5, 15 and 30 minutes after chamber deployment. ~~and~~ 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 ~~sample transport~~shipment. Mixing ratios of N_2O and CH_4 were analyzed using a gas chromatograph (Model 7890A, Agilent, USA) at RCEES, equipped with an ECD for detection of N_2O (at 375 ^oC with 25 ml min^{-1} Ar/CH_4 as make up gas), a FID for CH_4 (250 ^oC; 20 ml min^{-1} N_2 as make-up gas) and a 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^oC. 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

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 analyzer (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 of 2015, we measured the height and the diameter at breast height (DBH) of 6 to 10 Masson pines (~~only those~~marked in November 2013; ~~with~~ DBH > 5 cm) at each plot. ~~These data were~~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 ~~sum of~~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, in 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 normalized 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~~, Turkey post-hoc test~~) was conducted to examine the treatment effects on soil pH, nutrient contents in organic matter, tree biomass, 500-needle weight and needle nutrient content for each sampling~~and data of tree growth~~. 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~~ 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). In general, the concentration of NH_4^+ in soil water was small (< 0.5 mg 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~~ Reference plots, and the ~~sum of charge of dissolved base~~overall cationic charge~~s~~ 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~~During the experimental period~~, N_2O fluxes varied seasonally (Fig. 2), showing a significant relationship with daily precipitation (Fig. S4a), but not with daily mean temperature (Fig. S4b). ~~In the Reference plots, m~~Mean N_2O fluxes were generally below 50 µg N_2O-N m^{-2} hr^{-1} in the dry, cool season, but reached values of up to 600 µg N_2O-N m^{-2} hr^{-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 ~~T~~the greatest annual N_2O emission ~~was observed in the Reference plot~~ (7.9 kg N ha^{-1}) ~~of block 2~~. CH_4

fluxes in the Reference plots also varied greatly among blocks (Fig. 5). Net-emission of CH_4 was observed in summer 2013 (~ 80 µg CH_4-C m^{-2} hr^{-1}) in blocks 1 and 2, whereas block 3 showed CH_4 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 (Figs. 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) ~~decrease~~ reduction ~~in~~ of cumulative N_2O emission~~ by about 3 kg N ha^{-1} yr^{-1} on average, which is a 50% reduction~~ (Fig. 3). No immediate effect~~s~~ (within days) of P addition on N_2O emission was observed (Fig. S5).

~~CH_4 fluxes varied greatly between blocks (Fig. 5). Net-emission of CH_4 was observed in summer 2013 (~ 80 µg C m^{-2} hr^{-1}) in blocks 1 and 2, whereas block 3 showed CH_4 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 either precipitation or air temperature (Fig. S5c&d).~~ In the 1.5 years following P addition, mean CH_4 fluxes indicated net CH_4 emission (~ ~~+~~3.8 µg CH_4-C m^{-2} hr^{-1}) in the Reference, whereas net CH_4 uptake (~ ~~-~~6.5 µg CH_4-C m^{-2} hr^{-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.

3.3 The effect of P addition on tree growth

Throughout the 2-year experimental period, we observed no significant change in tree biomass ~~(138 t ha^{-1})~~ in response to P addition (Table S3). Likewise, there was no effect of P treatment on the 500-needle weight ~~(13 g on average)~~. Between the two samplings in 2013 and 2014, we found differences in chemical composition of the pine needles, but ~~this effect was not linked to P addition~~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 ~~hardly~~ not affected 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

~~Background~~ 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} hr^{-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 South China. For instance, N_2O emission rates averaged to 37 µg N_2O-N m^{-2} hr^{-1} in unmanaged sites at Dinghushan (Fang et al., 2009; Tang et al., 2006) and 50 µg N_2O-N m^{-2} hr^{-1} in N-fertilized 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)~~,~~ ~~as suggested by the~~A similar trend~~s~~ of increasing N_2O emissions with increasing N deposition rates has been reported for ~~indicated by data from~~ 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~~ed~~ 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 ~~mineral~~ inorganic N in soil water (predominantly NO_3^-; Fig. 2), particularly during summers, when NO_3^- concentrations were relatively ~~high~~ 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. ~~decreased mineral N content, most likely as a consequence of~~The attenuation of soil NO_3^- by P addition at TSP may reflect ~~-~~stimulated ~~plant~~ N uptake by plants and/or soil micro~~bial assimilation~~organisms. In a similarly N-rich, tropical forest in South China, Chen et al. (2016) reported a stimulation of net N mineralization and nitrification after six 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 immobilization by soil microbes. However, during our study period of two 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.~~It is noteworthy that there was no significant correlation between N_2O emission rates and soil water NO_3^- concentration in our study (Figs. 2 and S2), suggesting that the suppressing effect of P on N_2O emissions was indirect, probably by affecting the competition for mineral N between plant roots and microbes (Zhu et al., 2016).~~

In contrast to our ~~1.5-year~~ study, P-addition experiments in South Ecuador (Martinson et al., 2013) and South China (at Dinghushan Biosphere Reserve (Zheng et al., 2016) found no effect of a single P addition on N_2O emission during the first two years after application. However, significant reduction in N_2O emission was observed after three to five years ~~with~~ of continuous P addition, both at the Ecuadorian and the Chinese site (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 moderate amount of P added (10 kg P ha^{-1} yr^{-1}; Martinson et al., 2013). Moreover, the experiments were conducted in a forest with~~the relatively low ambient N deposition (~ 10 kg N ha^{-1} yr^{-1}) and small N_2O fluxes (~ 0.36 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 kg P ha^{-1} yr^{-1}; Martinson et al., 2013) could have resulted in an insignificant P effect in the first two years. By contrast, t The Dinghushan site in South China receives 28 36 kg inorganic N ha^{-1} yr^{-1} through wet inorganic N depositionby throughfall (Zheng et al., 2016Chen et al., 2016; Fang et al., 2008), which is similar to the inorganic N deposition at our siteTSP (Chen and Mulder, 2007b; Huang et al., 2015). However, soil KCl-extractable inorganic N (~ 40 mg N kg^{-1}; Zheng et al., 2016) and NO_3^- leaching (~ 20 kg N ha^{-1} yr^{-1}; Fang et al., 2008) at the Dinghushan site are several-fold smaller than at our site (~ 100 mg N kg^{-1} and ~ 50 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 µg m^{-2} h^{-1}) at Dinghushan were smaller than at TSP (> 50 µg m^{-2} h^{-1}; Fig. 4). These indicate that Dinghushan forest has stronger N assimilation and is thus less N-rich than TSP forest. They also observed larger N_2O emission rates (~ 0.88 kg N ha^{-1} yr^{-1} in the Reference plots) than in the Ecuadorian site. However, forests do not always display a straightforward relationship between N deposition and N_2O emissions. Manipulation experiments in the European NITREX project, for instance, revealed a much stronger correlation of N_2O emissions with soil NO_3^- leaching than with N deposition (Gundersen et al., 2012). Indeed, KCl-extractable mineral N at the Dinghushan site (~ 40 mg kg^{-1}; Zheng et al., 2016) is several-fold smaller than at our site (> 100 mg kg^{-1}; Zhu et al., 2013b), indicating that Dinghushan is less N-rich than TSP. This Therefore, we suggests that the response of N_2O emission to P addition might 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).

~~Other studies have~~Another study in a secondary in South China reported~~observed~~ increased N_2O emissions during two years ~~upon~~ after P addition, in a secondary mixed forest (~~Mori et al., 2013c;~~ Wang et al., 2014). ~~In an *Acacia mangium* plantation, fertilized with P, Mori et al. (2013b&c) found that N_2O emissions were stimulated in the short term but reduced in the long term.~~ 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~~are~~ generally explained by enhanced microbial ~~biomass~~ 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 (one week) incubation experiment with soils from an *Acacia mangium* plantation. Unlike Mori et al. (2013c), we did not find increased N_2O emissions ~~measured shortly~~ within a week after P addition at our site ~~in May 2014 were not different from fluxes in untreated reference plots~~ (Fig. S5). This may ~~indicate that~~suggest that denitrifier community at TSP was not responsive to the P applied, probably because TSP hillslope soils have large denitrification potentials (Zhu et al., 2013c). ~~plant uptake at TSP is more important for the effect of P addition on N_2O emissions than changes in microbial activity, which are expected to occur more rapidly.~~

The Reference plots at TSP showed net CH_4 emission for extended periods ~~of the year~~ (Figs. 5 and 6). Also, long-term CH_4 fluxes sampled between 2012 and 2014 on hillslope soils~~s~~ 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 ~~uptake rate~~fluxes reported for well-drained, forest soils in South ~~Chinese~~ China ~~forest~~ range from -30 to -60 μg CH_4-C m^{-2} hr^{-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 ~~by~~ due to supressed CH_4 consumption. One ~~reason~~ general explanation for the net CH_4 emission at TSP could be inhibition of CH_4 oxidation by NH_4^+, ~~as reported previously~~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 g $L^{-1;}$ Fig. 1), which does not preclude, however, that NH_4^+ availability from the soil exchangeable pool ~~is~~ 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, 2013b; Zhang et al., 2011), and has been attributed to ~~lessening the NH_4^+~~alleivating N inhibition of methane oxidation. ~~Unfortunately, we did not measure KCl-extractable NH_4^+ in our study, but a decline of available NH_4^+, which is the substrate for nitrification, is likely as NO_3^- concentrations in soil water were significantly smaller with in the P treatments (Fig. 1).~~ P addition may also result in a change of the taxonomic composition of the

methane oxidizing 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 fertilization is due to alleviating direct NH_4^+ inhibition of methane monooxygenase (Veldkamp et al., 2013), rather than to P-stimulation of methanotrophic activity (Veraart et al., 2015).

Shortly after fertilizer 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, ~~the occurrence of~~ Na^+ toxicity ~~at the treated plots,~~to a degree affecting N turnover processes in our plots~~,~~ is unlikely, as Na^+ concentrations in soil water, within one month after application (Table S2), did not exceed 5 mg L^{-1}, which is far smaller than the values commonly assumed to cause~~indicate~~ toxicity (40 to 100 mg L^{-1}) (Bernstein 1975). Frequent precipitation at TSP (Yu et al., 2016), both prior and following the addition of $NaH_2PO_4.2H_2O$ (Fig. 2), ~~apparently~~ 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 ~~plants~~ trees within the relatively short period of our study (Table S3 and Fig. S6), making it difficult to link the observed reduction in ~~mineral~~ 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. Firstly, two years of observation may be too short to detect any significant increase in tree growth, due to NO_3^- uptake ~~by plants~~, given the commonly large variabilities in tree biomass estimates (Alvarez-Clare et al., 2013; Huang et al., 2015). Secondly, a significant proportion of the added P, and of excess N, may have been assimilated by the understory ~~biomass~~vegetation, which was not assessed in this study. Previously, understory biomass~~vegetation~~ has been reported to quickly respond to P addition (Fraterrigo et al., 2011). Thirdly, 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 understory growth ~~and/or soil microbial biomass~~ may have been the main mechanisms responsible for observed NO_3^- decline in the P-treated soil (Hall & Matson 1999).

~~Overall, o~~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 fertilization was shown to significantly decrease NO_3^- concentrations in soil water and to overall reduce ~~both~~ N_2O and CH_4 emissions. These findings provide a promising starting point for improving forest management towards GHG abatement targets, taking into account the P and N status of subtropical soils in the region.

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Table 1 ~~Background~~ Ambient soil properties of the experimental plots at Tieshanping (TSP). Values are means and standard deviations in parenthesis (n = 6)[ϕ]. 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)	-*	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_{H2O} = Water-extractable P,~~ P_{Al} = Ammonium lactate-extractable P, Al_{ox} = Oxalate extractable Al, Fe_{ox} = Oxalate extractable Fe, P_{ox} = Oxalate extractable P.

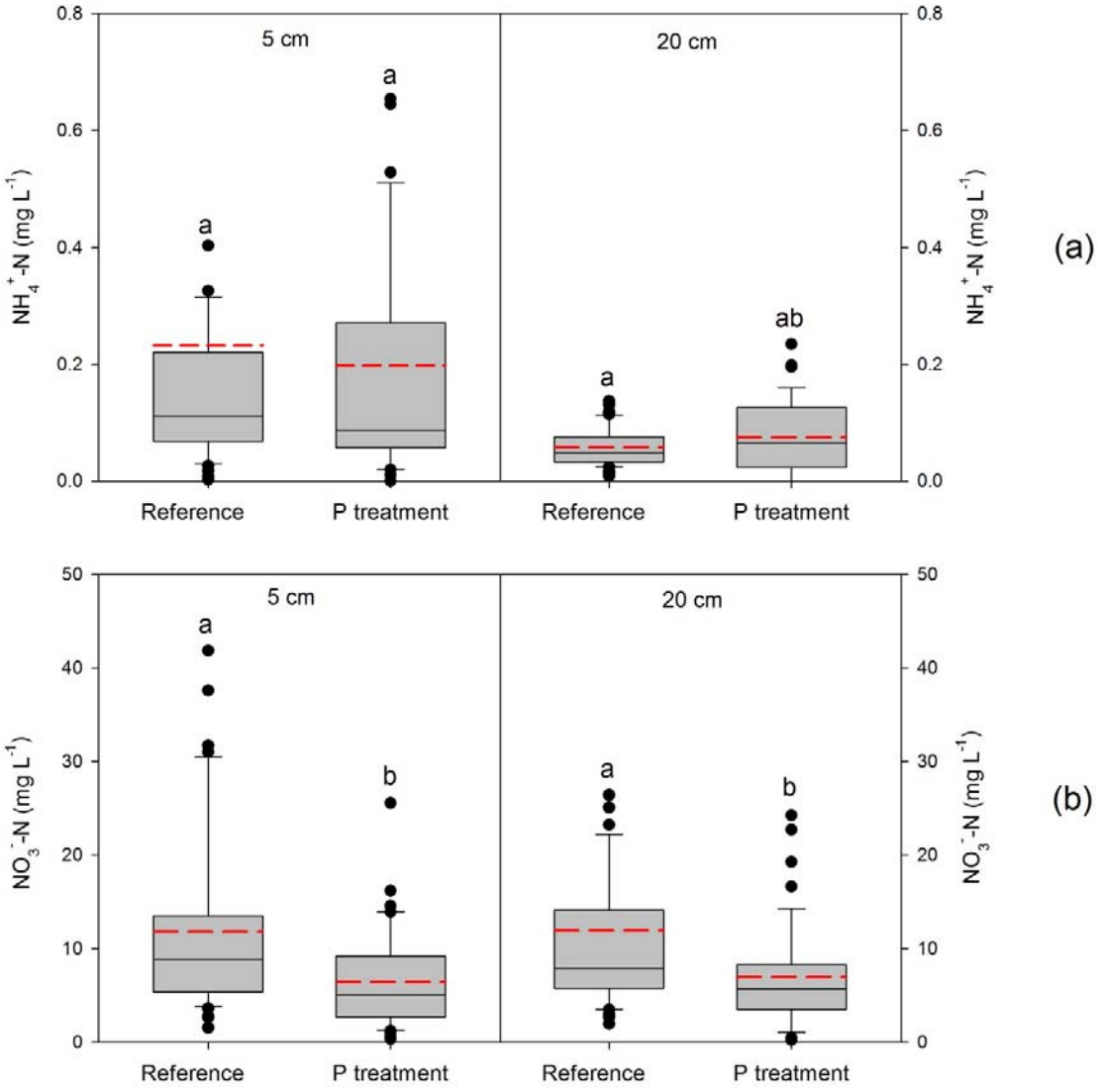
[ϕ] Water-extractable P was below a detection limit of 5 mg kg^{-1}, thus not presented in table,

[*] Data not available

682 **Table 2** Soil pH, C, N and P contents in the O/A horizon (0-3 cm) in the References (Ref) and P
683 treatments. Values are means and standard deviations in parenthesis (n = 9). P addition was
684 conducted on 14/05/04, after the first two sampling dates.

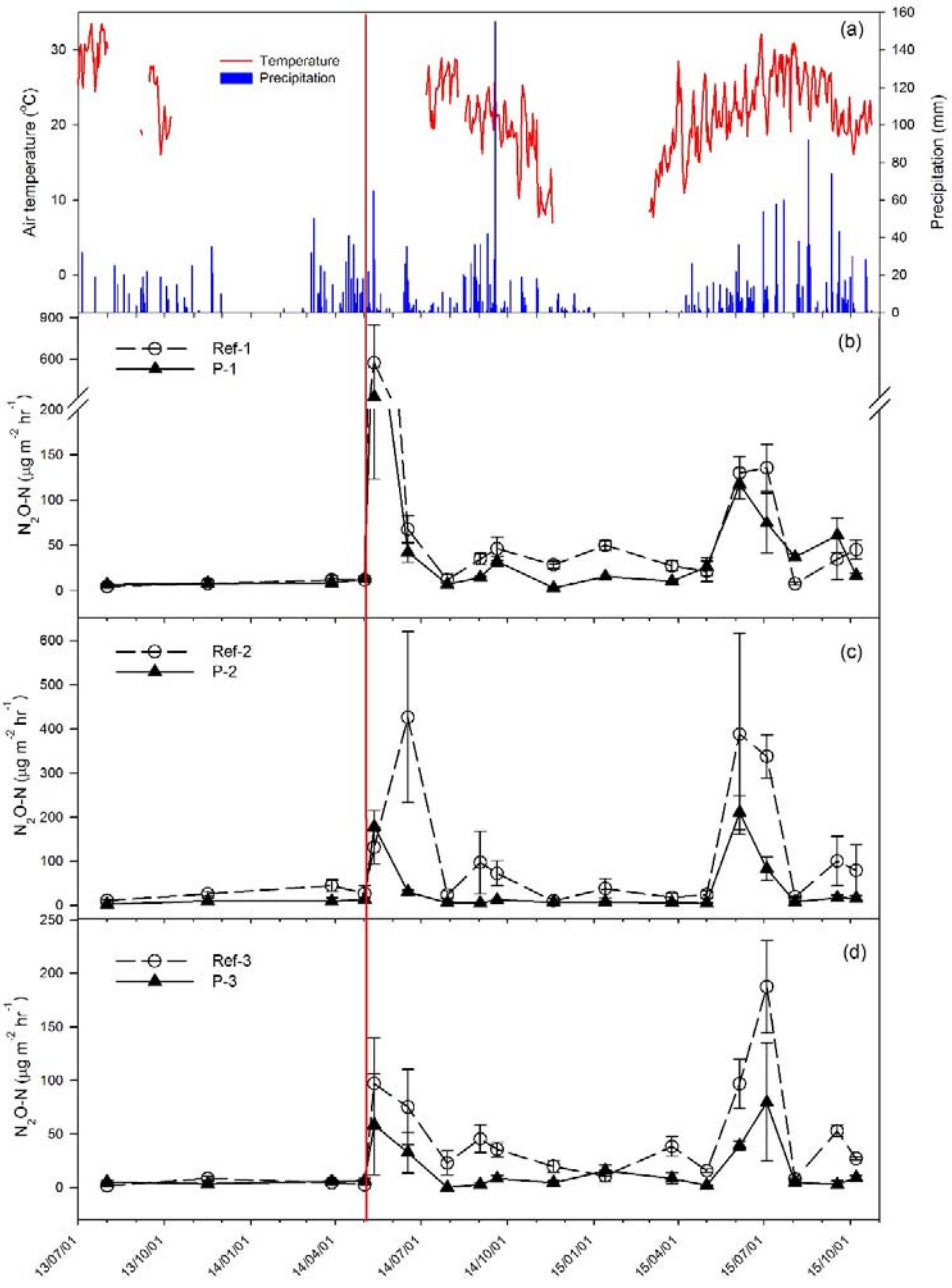
		pH	Total C	Total N	C/N	P_{Al}	Total P
			g kg^{-1}	g kg^{-1}		mg kg^{-1}	mg kg^{-1}
13/08/02	Ref	3.7 (0.1)^{ab†}	8.3 (2.3)^{ab}	0.5 (0.1)^{ab}	16.9 (1.1)^{b}	5.4 (1.4)^{a}	292 (46)^{ab}
	P	3.6 (0.1)^{b}	6.7 (2.0)^{b}	0.4 (0.1)^{b}	17.1 (2.1)^{ab}	5.1 (1.3)^{a}	260 (70)^{b}
14/05/02	Ref	3.7 (0.1)^{a}	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}
14/05/10	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}
14/12/02	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}
15/08/02	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}

685 † Different letters indicate significant differences between References and P treatments ($p < 0.05$).



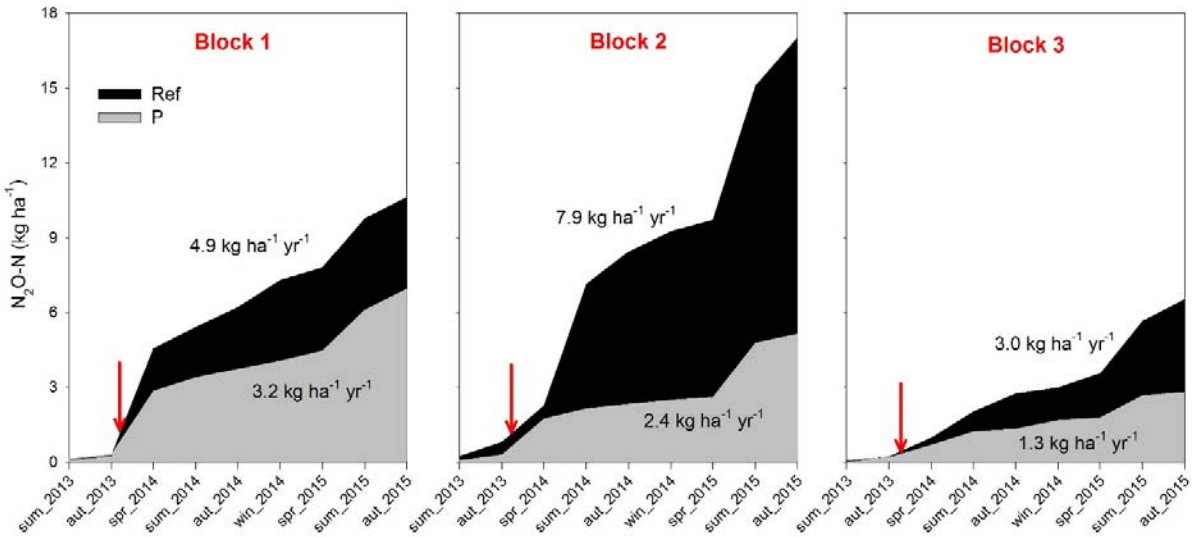
686

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



690

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



694

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

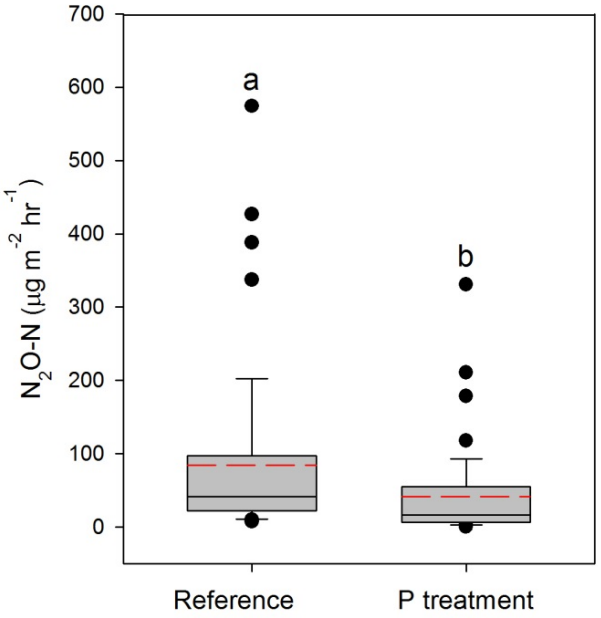
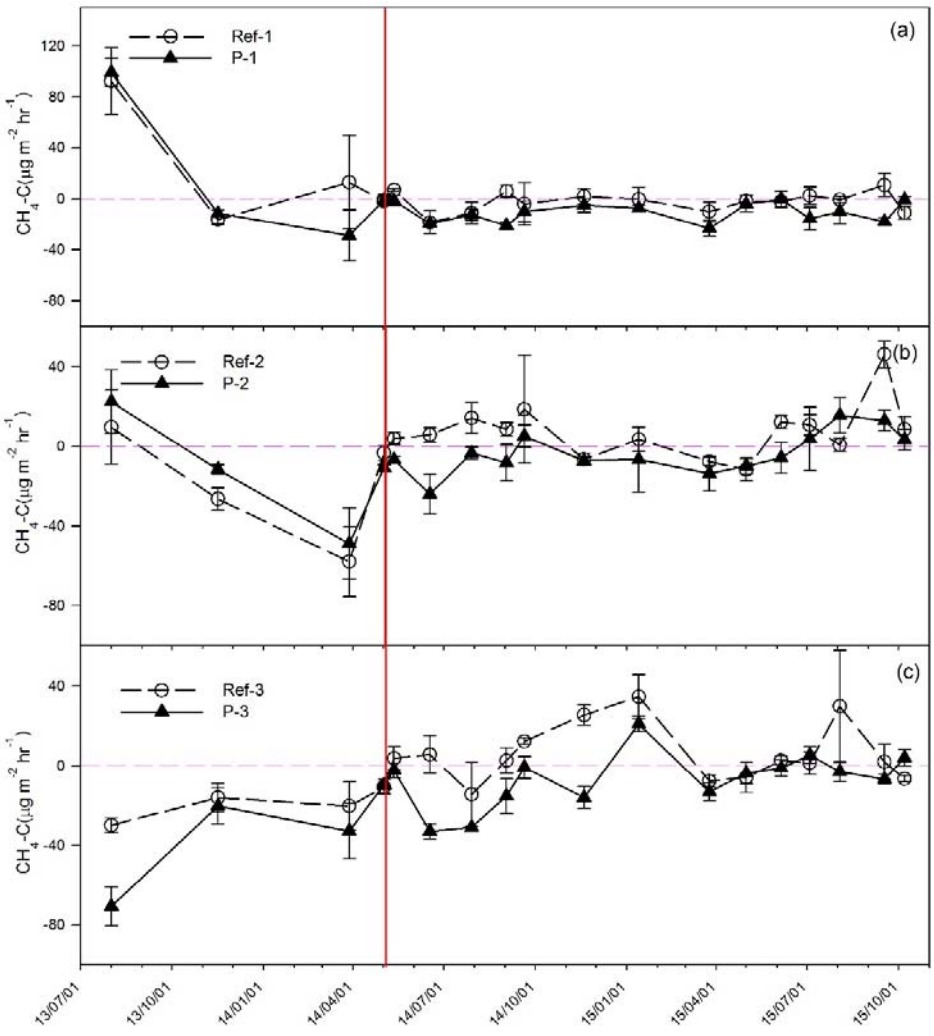
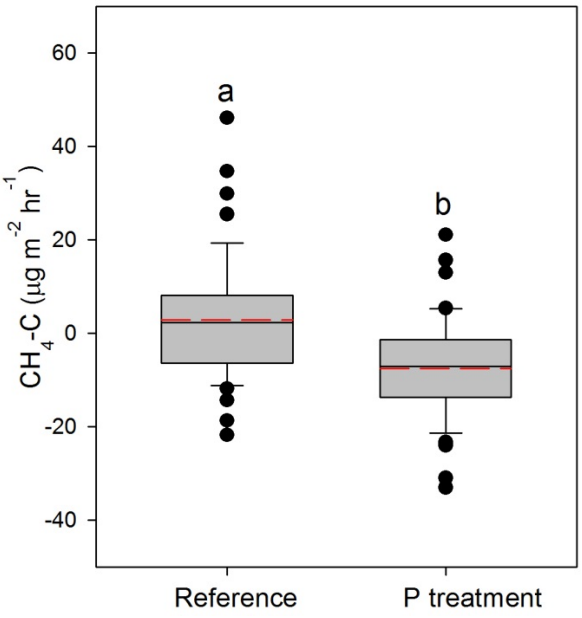


Fig. 4 Box whisker plots for N_2O fluxes in the Reference and P treatment throughout 1.5 years after the P addition; red dashed lines indicate mean values; linear mixed-effect models were used to test the P treatment effect; different letters indicate significant difference ($p < 0.05$).



701

702 **Fig. 5** Monthly mean CH_4 fluxes (±SE) in the References (Ref) and P treatments for three blocks
703 (a-c); the horizontal broken line indicates zero flux the red vertical line refers to the date of P
704 addition (4 May, 2014).



705

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