Topography-related controls on N_2O emission and CH_4 uptake in a tropical rainforest catchment

Longfei Yu^{1,2,3,4}, Jing Zhu^{1,5*}, Hongli Ji⁶, Xiaolong Bai^{1,2,7}, Youxing Lin^{1,2,7}, Yiping Zhang^{1,2,7}, Liqing Sha^{1,2,7}, Yuntong Liu^{1,2,7}, Qinghai Song^{1,2,7}, Peter Dörsch⁸, Jan Mulder⁸, Wenjun Zhou^{1,2,7*}

¹Key Laboratory of Tropical Forest Ecology, Xishuangban ia Tropical Botanical Garden, Chinese Academy of Sciences, 666303, Mengla, Yunnan China

²Center of Plant Ecology, Core Botanical Gardens, Chinese Academy of Sciences, 666303, Xishuangbanna, Yunnan, China

³Institute of Groundwater and Earth Sciences, Jina Viniversity, 510632, Guangzhou, China

⁴Laboratory for Air Pollution & Environmenta! Technology, Empa, Swiss Federal Laboratories for Materials Science and Technology, Ueverlandstr. 129, CH-8600 Duebendorf, Switzerland

⁵Key Laboratory of Ecology of Rare and Endangered Species and Environmental Protection (Guangxi Normal University), Minist y of Education, 54004, Guilin, China

⁶Lushan Botanical Garden, Jiangxi Prevince and Chinese Academy of Sciences, 332900, Jiujiang, China

⁷University of Chinese Academy of Sciences, 100039, Beijing, China

⁸Faculty of Environmatal Sciences and Natural Resource Management, Norwegian University of Life Sciences, Postbox 3003, N-1432 Aas, Norway

*Correspondence: Jing Zhu (zhuj@gxnu.edu.cn) and Wenjun Zhou (zhouwj@xtbg.cn.cn)

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Abstract

Forest soils in the warm-humid tropics significantly contribute to the regional greenhouse gas (GHG) budgets. However, spatial heterogeneity of GHG fluxes is often overlooked. Here, we present a study of N₂O and CH₄ fluxes over 1.5 years, along a topographic gradient in a rainforest catchment in Xishuangbanna, SW China. From the upper hillslope to the foot of the hillslope, and further to the flat groundwater discharge zone, we observed a decrease of N₂O emission associated with an increase of soil water-filled-1 ore-space (WFPS), which we tentatively attribute to more complete denitrification to N at larger WFPS. In the well-drained soils on the hillslope, denitrification at anaerobic microsites or under transient water-saturation was the potential N₂O source. Negative CH₄ fluxe, a ross the catchment indicated a net soil CH₄ negatively related to WFPS, reflecting the to ographic control. Our observations also suggest that during dry seasons N_2O emissio (v/a) significantly dampened (< 10 $\mu g N_2O$ -N m⁻² h⁻¹) and CH₄ uptake was strongly enhance. (83 µg CH₄-C m⁻² h⁻¹) relative to wet seasons (17 µg N₂O-N m⁻² h⁻¹ and 56 μg CH₄-C m⁻² ½⁻¹). In a post-drought period, several rain episodes induced exceptionally high N₂? emissions (450 µg N₂O-N m⁻² h⁻¹) in the groundwater discharge zone, likely driven by flushing of labile organic carbon accumulated during drought. Considering the global warming potential associated with both GHGs, we found that N₂O emissions largely offset the C sink contributed by CH₄ uptake in soils (more significant in the groundwater discharge zone). Our study illustrates important topographic controls on N₂O and CH₄ fluxes in forest soils. With projected climate change in the tropics, weather extremes may interact with these controls in regulating forest GHG fluxes, which should be accounted for in future studies.

1 Introduction

Nitrous oxide (N₂O) and methane (CH₄) are potent greenhouse gases (GHGs) with global warming potentials (over 100 years) 28 and 265 times of that of carbon dioxide (CO₂), respectively (Myhre et al., 2013). Recent records have indicated enhanced growth of both N₂O and CH₄ levels in the atmosphere (Nisbet et al., 2019; Thompson et al., 2019). N₂O and CH₄ emissions may significantly offset the worldwide efforts in mitigating CO₂ emissions to counteract climate change and thus challenge the Paris agreement which aims at limiting global warming to 1.5 - 2°C (Millar et al., 2017). Soil emissions play a major role in global GHG budgets (Davidson and Kanter, 2014; Tian et al., 2015). Among soils with natural vegetation, warm and humid tropical forest soils are believed to be the most important N₂O source (Matson and Vitousek, 1990; Werner et al., 2007) and active CH₄ sink (Dutaur and Verchot, 2007). Globally, tropical forests are estimated to emit 1.03 Tg N₂O yr⁻¹ (Zhuang et al., 2012) and to take up 6.66 Tg CH₄ yr⁻¹ (Dutaur and Ve ch. 2007). However, current estimates for regional N₂O and CH₄ budgets show large urcartainties, mostly due to spatial heterogeneity of GHG fluxes and limited studies of biogenche nical controls on N₂O and CH₄ production/consumptions in soil (Dutaur and Verchot, 2067: Verner et al., 2007).

N₂O is produced from both biotic and abiotic processes in forest soils (Butterbach-Bahl et al., 2013). Biological production of N₂O in soils generally involves nitrification and denitrification processes, which predominate in aerobic and anaerobic conditions, respectively (Weier et al., 1993). Abiotic production of N₂O occurs by chemical reactions of biological nitrogen (N) cycling intermediates (nitrite or hydroxylamine) (Liu et al., 2019), and has been shown to potentially play a role in acidic forest soils (Wei et al., 2017). Aside from production, the

reduction of N_2O to N_2 by denitrification is important in controlling N_2O fluxes, and this step is sensitive to oxygen availability, controlled by soil moisture (Weier et al., 1993), and to pH (Liu et al., 2010). Hence, variability of N₂O emission may be largely explained by soil carbon (C) and N substrates, temperature, moisture and soil pH. In (sub)tropical forest soils, N₂O emissions show a seasonal pattern with episodically high fluxes in the wet seasons (Zhu et al., 2013a), while the effect of soil temperature seems to be less important (Gütlein et al., 2018; Kiese et al., 2003; Kiese and Butterbach-Bahl, 2002). Lowland tropical forests usually have fast N cycling rates under warm climate, thus continuously providing inorganiant as substrate for N₂O emission (Müller et al., 2015; Vitousek and Matson, 1988). Howeve, microbial production of N₂O in soil may depend on biologically available C and other nutric as such as phosphorus (P), which are relatively limited compared to N. Two studies which manipulated litter inputs to tropical forest soils, found that dissolved organic C derived from litter decomposition drives N₂O production through denitrification (Gao et al., 2012: Wieder et al., 2011). Other studies have applied P fertilizer to the forest floor, and observed mitigation of N₂O emissions, which they attributed to enhanced N uptake in soils (Müller et al., 2015; Yu et al., 2017b; Zheng et al., 2016).

Most well-drained fores soi's constitute a net CH₄ sink, i.e. take up and oxidize CH₄ from atmosphere (Le Mer and F.oger, 2001). High-affinity methanotrophs mediating this uptake utilize CH₄ as the only C and energy source (Kolb, 2009). The activity of these methanotrophs strongly depends on the diffusion of atmospheric CH₄ into soils, which is related to soil porosity (thus related to soil moisture and clay content) (Le Mer and Roger, 2001; Smith et al., 2003). By contrast, soil temperature and pH play less important roles for CH₄ oxidation. In forest soils with excessive N, CH₄ uptake is often found to be inhibited (Aronson and Helliker, 2010; Kolb, 2009; Liu and Greaver, 2009). One possible mechanism is that increased NH₄⁺ in soil competes with

CH₄ for the reactive site of methane monooxygenase, the key enzyme for CH₄ oxidation (Bedard and Knowles, 1989). In water-saturated forest soils, net CH₄ emissions have been reported, pointing to a role of methanogenesis for net CH₄ flux in forest soils under a wet climate (Itoh et al., 2009; Kaiser et al., 2018; Yu et al., 2019b). Soil CH₄ production, i.e. methanogenesis, is mediated by a group of microbes (methanogens) commonly found in strictly anoxic environment (Conrad, 2009), which may become active during anoxic spells. As methanotrophs and methanogens have been reported to coexist in variable environments (Angle et al., 2017; Cai et al., 2016), both production and consumption processes need to be considered when investigating CH₄ turnover, particularly in soils with fluctuating water content (Kolb and Horn, 2012).

Tropical forests are typically under warm and hamid climate, thus having large potentials to become regional hotspots of GHG emissions. And et al., 2012; Keller et al., 1986; Werner et al., 2007). Recent studies to predict and mitigat. GHG emissions associated with climatic changes and human activities in tropical for sts are commonly based on climate and fertilizer manipulation experiments (Hall et al., 2012; Müller et al., 2015; Steudler et al., 2002; Werner et al., 2006), or on comparing GAG fluxes among different land-use types (Gütlein et al., 2018; Zhou et al., 2016). However, spatial heterogeneity of tropical forest soils, which is often linked with topography, has been generally overlooked when considering ecosystem-level GHG balances. Topography controls soil moisture levels, hydrological transport of nutrients and vegetation type in tropical forests, thus exerting a strong effect on forest N cycling and N gas fluxes and their responses to environmental changes (Anderson et al., 2015; Enanga et al., 2016; Weintraub et al., 2015). Fang et al. (2009) measured GHG fluxes along a steep but short slope in a broad-leaved forest of South China and found an increase of N₂O emission together with increasing soil moisture along the slope. By contrast, in a subtropical forested catchment in SW

China, Zhu et al. (2013a) reported a different relationship with N₂O emissions being much larger at the upper hillslope than in a groundwater discharge zone that is occasionally water-logged. Such discrepancy may be explained by more complete denitrification to N₂ in periodically submerged soils (Koehler et al., 2012) and illustrates the importance of topographic positions on N cycling and thus N₂O emission (Wexler et al., 2014; Yu et al., 2016). Previous works have also revealed topographic control on CH₄ fluxes (Kaiser et al., 2018; Warner et al., 2018; Yu et al., 2019b). For example, Warner (2018) identified the transition zone between upland and valley bottom as a hotspot of CH₄ uptake in a temperate forests land coape. Wetland/riparian soils in the lowland subtropical-tropical forests have recently been found to be potentially important CH₄ sources which could alter the ecosystem CH₄ budgets (Salape et al., 2018).

To explore topographic controls on N₂O and CH₄ fluxes in tropical rainforest catchments, we conducted *in-situ* measurements over 1.5 years at three topographic positions along a catchment located in SW China. This site has *e* p phounced monsoonal climate with distinct wet and dry seasons and provides the oppor unity to examine the effect of dry-wet transition on soil-atmosphere GHG exchange (C'Connell et al., 2018). We hypothesized that: 1) N₂O emission increases and CH₄ uptake cecreases from the drier upper to the wetter lower topographic positions; 2) warm-hum²J summers promote N₂O emissions but suppress CH₄ uptake; 3) topographic controls on N₂O and CH₄ fluxes are less significant in the dry seasons.

2 Materials and Methods

2.1. Site description

The study site is a 19.7-ha tropical forested catchment, located in Xishuangbanna (XSBN), SW China (Fig. 1). XSBN is covered by rainforests with a mean canopy height of 35 m. With a mean elevation of 750 m a.s.l., XSBN has a mean annual temperature of 21.4 °C and a mean annual precipitation of 1415 mm (Cao et al., 1997). The climate shows a distinct seasonal pattern, with a cool-dry season between November and April and a warm-wetter about between May and October (Fei et al., 2018). Generally, XSBN receives more than 80% of the annual precipitation during the wet season. The annual N deposition in canopy throughfall is about 10 kg ha⁻¹. XSBN is dominated by Oxisols developed from marine sen 'stone (Qiao et al., 2014). XSBN soils have a thin organic layer (~ 1 cm), and the top-so.¹ p⁻¹ ranges from 4 to 5 (Chan et al., 2006). Organic C and total N contents in soils (0-10 cn. depth) range from 24 to 28 and 1.6 to 2.8 g kg⁻¹, respectively (Chan et al., 2006; Qiao et al., 2014).

2.2. Experimental Design

N₂O and CH₄ fluxes we. • measured at three hydrologically connected topographic positions (A, B and C) in the XSBN rainforest catchment (Fig. 1). Position A is situated at the upper hillslope, position B at the foot of the hillslope (footslope; the transition point between hillslope and groundwater discharge zone) and position C in the groundwater discharge zone near a stream. The upper hillslope is relatively steep, with an inclination of 30°. The groundwater discharge zone flattens out, and is occasionally water saturated after heavy rainfall.

From July 2013 to December 2014, we measured soil-atmosphere N₂O and CH₄ exchange biweekly in triplicate plots at each of the three topographic positions. Fluxes were measured by static chambers (Yu et al., 2019b). Stainless steel frames were permanently installed in the soil weeks prior to our first sampling. During gas sampling, we used vented stainless-steel chambers (30-cm diameter and 30-cm height) deploying them against water-sealed frames. To measure fluxes, we collected 20-ml gas samples 0, 15, 30, 45 and 60 min after chamber deployment by 50-ml syringes equipped with three-way valves. Gas samples were injected into pre-evacuated 12-ml vials crimp-sealed with butyl septa (Chromacol, UV), and transported back to the laboratory at the XSBN Tropical Botanical Garden (Chine: Academy of Sciences) where they were analyzed for N₂O and CH₄ within 24 hours. Next to each static chamber plot, we installed ceramic lysimeters (P80; Staatlich Porzellanmanu faktur, Berlin) to collect soil water from O/A and AB soil layers. Triplicates of soil vater was collected bi-weekly together with the gas sampling during the wet season, but it was not possible to obtain enough soil water for chemical analysis during the dry seasons. Triplicates of water samples for each layer at each topographic position were pooled and stored to 4°C prior to analysis.

We monitored precipitat on every half an hour with a rain gauge coupled with a HOBO data logger (RG3-M, Onset, US) attached to the top of an eddy flux tower near our sampling plots. Throughfall volume was measured with a tailor-made V-shape container (2.0 m*0.3 m and 11 replicates) installed under the forest canopy. Throughfall volume was calculated from the water levels recorded with a HOBO data logger (U20-001-04, Onset, US). At the watershed outlet, we installed a 90° V-notch weir instrumented with a water-level recorder (U20-001-04, Onset, US), which was set to take average discharge measurements at 5-min intervals. Water samples of throughfall and runoff were additionally collected bi-weekly for chemical analyses except for the

period of November 2013 to January 2014. In parallel with the gas sampling, soil temperature and volumetric soil moisture were measured at 10-cm depth using a hand-held time domain reflectometer (TDR100, Campbell Scientific, Logan, Utah, USA) close to the flux sampling plots.

2.3. Chemical analyses and data calculation

Gas samples collected from the static chambers were analyzed for N₂O and CH₄ mole fractions with a gas chromatograph coupled with an electron capture refector (ECD) and a flame ionization detector (FID) (Agilent GC- 7890A, USA). For ECD, a makeup gas of Ar/CH₄ (90/10 vol%) was used. We calculated N₂O and CH₄ fluxes by innear or polynomial regression (estimating time-zero fluxes in case a concave downward curve in concentration change was seen) of gas mole fractions over time (Yu et al., 2013a).

In water samples (including precipitation, 5.1 water and runoff), pH was measured with a pH electrode (Orion SA720, ThermoFishc. USA). We measured NH₄⁺ and NO₃⁻ concentrations in water samples with a continuous (10w autoanalyzer (Auto Analyzer 3; Bran and Luebbe GmbH, Germany). Total organic carbon (SOC) and total nitrogen (TN) concentrations were determined with LiquiTOC II Elementa. Analyzer System (Langenselbold, Germany). Additionally, soil water collected with lysin eter samplers had been already filtrated, so the TOC concentrations determined for soil water mostly represent dissolved organic carbon (DOC). The dissolved organic nitrogen (DON) concentration in water samples was calculated as the difference between TN and total inorganic N (NH₄⁺ and NO₃⁻).

We calculated soil water-filled pore space (WFPS) from the TDR-measured volumetric soil moistures (VM, cm³/cm³), soil bulk density (BD, 1.0 g/cm³ for Plots A and B; 1.2 g/cm³ for Plot C) and soil particle density (PD, 2.65 g/cm³) (Linn and Doran, 1984) as below:

WFPS (%) = VM /
$$(1 - BD/PD) * 100$$
 (1)

2.4. Estimates of annual gas budgets

We estimated annual N₂O and CH₄ fluxes based by linear interpolation of fluxes between subsequent sampling dates (Yu et al., 2019b). Annual cumulative fluxes computed for each replicate sampling plot were averaged to obtain mean annual fluxes, and uncertainties were determined as 1 SD. Considering the exceptionally high N₂O fluxe on 6/17/2014, our estimates were made by two scenarios: Scenario 1 took all flux data into account and Scenario 2 replaced fluxes on 6/17/2014 from the previous and the next samplings. To examine the combined contribution of N₂O and CH₄ fluxes across three topographic positions of the XSBN catchment to global warming, we normalized the N₂O and CH₄ luxes to CO₂-equivalents (kg CO₂) based on their global warming potentials (GW's) Tian et al., 2016) and calculated the net GWP contributions of three plots in the catchia ent as below:

$$GWP_{net} = F_{N2O} * GWP_{N2O} + F_{CH4} + GVP_{CH4}$$
 (2)

where GWP_{net} refers to GWF cor tributions equivalent to kg CO_2 ha⁻¹ yr⁻¹ equivalent as a result of both N_2O and CH_4 each arge; F_{N2O} and F_{CH4} refer to fluxes of N_2O and CH_4 (kg ha⁻¹ yr⁻¹), respectively; GWP_{N2O} and GWP_{CH4} refers to 100-year GWPs for N_2O (265) and CH_4 (28), respectively.

2.5. Statistical analyses

All statistical analyses for this study were performed with R 3.3.1 (R Core Team, 2016). We checked all data for normality (Kolmogorov- Smirnov's test) and homoscedasticity (Levene's test) before testing for statistical differences. Logarithmic or square root transformation was

applied to data which were not normally distributed. We used linear mixed-effect models to test differences in N₂O and CH₄ fluxes among three topographic positions (Yu et al., 2017b). For this model, we took the difference of topographic positions as a fixed effect and sampling time and replicates within each plot as random effects. We applied stepwise linear regressions of N₂O and CH₄ fluxes against different biogeochemical factors. Additionally, Pearson correlation tests were applied to test individual correlations between gas fluxes and soil parameters. We used repeated analysis of variance (ANOVA) to compare soil temperature, WFP₃, pH and C and N contents in soil water, and estimated annual gas budgets across topographic positions. If not specified otherwise, the level of significance is set to P < 0.1.

3. Results

3.1. Climatic factors

From 2011 to 2015, XSBN received annual precipitation ranging from 1060 to 1419 mm (Fig. 2a). In 2014, annual precipitation declined by ~30%, indicating a drier year. If compared with the other years, 10 months in 2014 had negative rainfall anomalies which were particularly large in the cool-dry season in the beginning of 2014, suggesting drought condition (Fig. 2b). Daily precipitation and throughfall records show that most rainfall (Including rainstorms) occurred in the wet seasons (May to October; Fig. 3a). By contrast, during the dry season starting from January 2014, almost no rainfall had occurred until Line end of March 2014, suggesting a significant drought. The variation of runoff was perenally in accordance with that of rainfall, displaying high discharge in the wet seaso, s (Lig. S1a).

Over the observational period of 1.5 'each temporal variation of WFPS followed that of rainfall, showing small values < 20% in March 2014 and large values up to 80% in August and September 2014. Along the topographic gradient, WFPS significantly increased in the order position A < B < C (Figs. 1 and 3a). The difference of WFPS among topographic positions was smaller in dry seasons than in wet seasons. Soil temperature at 10 cm depth was consistently > 20°C during the warm-wet season and varied between 15 and 20°C in the cool-dry season (Fig. 2b), without showing any apparent spatial pattern.

3.2. Water chemistry in throughfall, runoff and soil water

Of TN in throughfall, DON concentration was highest (up to 10 mg N L⁻¹), while NO₃⁻¹ concentration ranked second (Fig. S1b). The concentrations of TN were significantly smaller in

runoff, and NO₃⁻ and DON contributed equally (Fig. S1c). No apparent temporal pattern of N concentrations in throughfall and runoff was found, except for elevated NO₃⁻ concentration in throughfall at the end of 2014 (Fig. S1b). Most likely this was due to enhanced dry deposition in the dry season of 2014, given that little rainfall occurred in this period (Fig. 3a). Furthermore, TOC concentration in throughfall spiked at the transition from dry to wet season in 2014, up to 100 mg C L⁻¹ (Fig. S1b). A possible explanation is the accumulation of TOC in the canopy during the dry season without rainfall. The variations of TOC in runoff were generally in accordance with those in throughfall, but differed in magnitudes cannot TOC in throughfall).

Due to drought during the dry seasons of 2013-2014, we only collected soil water from the wet seasons for chemical analyses (Figs. 4 and S2). In soir water collected from the organic horizon (O/A, Figs. 4 and 1), little NH₄⁺ was detected, NO₃⁻ and DON concentrations increased from position A to B and then decreased from position B to C. Both NO₃ and DON concentrations were smaller in soil water than in the orghiall. Significantly higher NO₃ concentrations in soil water were found in the wet sear on of 2013 than in 2014 (except for position C; Fig. 4). Soil water DOC concentrations were generally higher and more variable in the wet season of 2014 than in 2013; a strong increase in DOC concentration was found at position B in the beginning of the wet season of 2014 S il water pH, varying from 5 to 7 and was lower at position A than at down-slope topographic positions (Fig. 4), and its variability may have been largely linked with that of throughfall (Fig. S1). In soil water from the mineral horizon (AB, Fig. S2), variations of N concentration and pH were mostly similar to those found in the organic horizon. However, the NO₃ concentration in the AB horizon was about twice as high as in the O/A horizon at positions A and B. In addition, at position C, we observed an unexpected increase of DOC concentration in the fall of 2013 in the AB horizon, but not in the O/A (Fig. S2).

3.3. Spatiotemporal patterns of N₂O and CH₄ fluxes

Comparison of linear mixed effect models fitted to N₂O fluxes at the three topographic positions indicated that N₂O emission was overall highest at position A (Fig. 5a). Over the entire observation period, N₂O fluxes varied from 0 to 713 µg N m⁻² h⁻¹, exhibiting larger variability among replicates at higher fluxes (Figs. 3c and S3). On 6/17/2014, 1.5 months after the drought period, an exceptionally high mean flux of 450 µg N₂O-N m⁻² h⁻¹ was observed at position C. Although less pronounced, this was also seen at position B but not at A. WFPS was mostly below 50%, but several rain episodes had occurred before an 1 at his date (70 mm in total during the week before and 30 mm on 6/17/2014), thus representing a rewetting event after drought. Emission rates of N₂O averaged for the entire observation period (as shown by box-whisker plots) decreased from the hillslope (A) to the foots one (B) and to the groundwater discharge zone (C), if the exceptionally large fluxes measured of 6/17/2014 at position C were excluded (Fig. 5a). The effect of topography on N₂O emissions became less obvious when segregated for individual seasons (Fig 5b). N₂O emission was generally lower in the dry season than in the wet season (Fig. 5b). If compared for observations during the same months, N₂O fluxes in wet season of 2014 were larger than in 2013 of the topographic positions on the hillslope (Fig. 5c).

CH₄ fluxes were mostly negative, indicating that soils at the XSBN catchment acted as a sink for CH₄ most of the time (Fig. 3d). According to comparison of linear mixed effect models, position A showed significantly larger CH₄ uptake than positions B and C overall (Fig. 5d). When segregating CH₄ fluxes for season, it can be seen that this was mainly due to very high uptake rates at position A during the dry season (Fig. 5e). CH₄ uptake rates among triplicates varied between 6.3 and 141.4 µg C m⁻² h⁻¹ and were significantly larger in the dry season than in the wet season (Fig. 5e). In the second wet season, soils took up significantly more CH₄ than in the

first dry season (Fig. 5f). However, on 6/17/2014 when exceptionally high N_2O fluxes were found, CH_4 uptake was somewhat suppressed, but not significantly distinguishable from other fluxes recorded in the wet season of 2014 (Fig. 3d).

3.4. Factors controlling N₂O and CH₄ fluxes

Stepwise linear regression including all ancillary variables (soil temperature and WFPS, soil water pH, DOC and mineral N concentrations) revealed that soil N2O fluxes in the XSBN catchment are positively related to soil temperature and negativaly related to WFPS (Table 1). By looking into individual topographic positions, we also found positive relationship between soil temperature and N₂O flux (Fig. 6a). However, or positions A and B (Fig. 6b), linear regressions showed significantly positive correlation between N₂O flux and WFPS, which is opposite to the relationship shown by stopy se linear regression (Table 1). Pearson productmoment correlation suggested that the 150 flux was significantly negatively correlated with soil water pH when using data from all three rositions (Table S1), whereas temporal pH variability at single positions did not seer to influence N₂O flux. Such relationship may reflect the topographic control on soil water pH (wetter soils with higher pH; Figs. 4 and S2). DOC in the soil water had a significant positive effect on N₂O emissions at positions A and C but not for the entire transect (Table S1), which is in line with the results of multiple linear regression (Table 1). At position A, the N₂O flux was also significantly influenced by DON (positive) and NH₄⁺-N (negative) concentration in the soil water.

CH₄ fluxes across the catchment were largely (56%) explained by soil WFPS and temperature (Table 1). Positive relationships of CH₄ fluxes with WFPS (i.e. a negative relationship between CH₄ uptake and WFPS) and temperature were also observed at individual topographic positions

(Figs. 6c&d). At position A (Table S1), Pearson correlation showed a positive effect of NO₃⁻-N concentration and a negative effect of DON concentration on CH₄ fluxes.

3.5. Annual N₂O and CH₄ budgets

Annual N_2O fluxes across different topographic positions in the XSBN catchment varied from 0.76 to 1.94 kg N ha⁻¹ yr⁻¹, among which the groundwater discharge zone (C) contributed the most, albeit that its uncertainty was also largest (Table 2). However, if the exceptionally high fluxes on 6/17/2014 at position C (Fig. 3c) were replaced with the average of fluxes from the adjacent dates, the computed annual N_2O flux would be smallest (0.41 kg N ha⁻¹ yr⁻¹).

Annual CH₄ uptake rates ranged from 5.27 to 7.47 k₂ C ma⁻¹ yr⁻¹ and were higher at the upper hillslope (A) than at the other two topographic positions (Table 2). We calculated net global warming potential (GWP) based on the annual fluxes of N₂O and CH₄. The results suggest that soil N₂O emission at the XSBN catchment was largely offset the GWP resulting from CH₄ uptake, thus turning the catchment into a net source of radiative forcing based on N₂O and CH₄ which offsets the CO₂ sink of this forest. Net GWP contribution increased downslope the catchment. However, if excluding the exceptionally high N₂O flux from 6/17/2014, the lower topographic positions exhibited a small sink of GWP.

4. Discussion

Previous catchment studies have shown that topography affects N₂O and CH₄ fluxes in forest soils (Fang et al., 2009; Kaiser et al., 2018; Warner et al., 2018; Yu et al., 2019b; Zhu et al., 2013a). Our results, covering 1.5 years of N₂O and CH₄ fluxes at three distinct topographic positions of the XSBN headwater catchment (Figs. 3 and 5) suggest that soil moisture gradient (Fig. 1) associated with topography is an important driver for the spatial variabilities observed for N₂O and CH₄ fluxes (Fig. 6 and Table 1). At the hillslop (position A) where soils were relatively dry, we identified hotspots of both N₂O emission and CH₄ uptake, while the footslope (B) and the groundwater discharge zone (C) appeared to contribute less to the N₂O source or CH₄ sink, particularly during the wet season.

In non-fertilized soils, N₂O emission is often primarily associated with soil moisture content (Van Lent et al., 2015); with soil moisture contents below and near field capacity, N₂O fluxes display a positive relationship with roi. Phoisture. At the well-drained hillslope of XSBN, where WFPS was mostly below 60%, we observed significant positive correlations of N₂O emission rates with WFPS (Figs. 6b). Both nitrification and denitrification pathways can contribute to N₂O production in soils (Firescene and Davidson, 1989), and their relative importance largely depends on oxygen availability. Positive correlation of N₂O emissions with WFPS has been taken as indicative for denitrification being the dominant source (Davidson et al., 2000). In warm and well-drained soils with abundant mineralization, however, nitrification may support denitrification directly by producing NO₃ and indirectly by consuming oxygen at significant rates. At the hillslope of XSBN, we observed vertical increases of NO₃ concentration within soil profiles (Figs. 4 and S2), suggesting net production of NO₃ (i.e. nitrification). Small changes in

soil moisture around 60%-WFPS may thus result in coupled nitrification-denitrification (Zhu et al., 2013a). Yu et al. (2017a) conducted a short-term *in situ* ¹⁵N tracing experiment at the hillslope in a subtropical forest in SW China and found that denitrification was the dominant N₂O source during a period with high N₂O fluxes (WFPS: 60 to 75%). At moderate WFPS values, small increases in soil moisture may stimulate nitrification, which could be a direct source of N₂O, particularly during the dry season at XSBN, when WFPS dropped below 40% (Fig. 3a). In addition, a short-term *in situ* ¹⁵N labeling experiment performed in the XSBN forest in late April 2016 indicated that nitrification was the predominant N₂O source during the first 27h after adding ¹⁵NO₃ (WFPS: 20 to 60%; Zhou et al., Fersonal Communication). However, it is beyond the scope of this study to partition N₂O source—solely based on the gas flux and soil measurements. More field experiments with isotor accertain poth wet and dry seasons are needed to fully elucidate the major N₂(*pr. duction processes at XSBN.

Surprisingly, opposite to our hypothes: 142O fluxes at the footslope and in the groundwater discharge zone, where WFPS w s consistently higher, were lower than at the drier hillslope during the wet season (Figs. 3a, and 5b). Zhu et al. (2013a) observed a similar spatial pattern of N2O fluxes during the vet eason along a hydrological flow path in a subtropical forested catchment in SW China, showing on average lower N2O emission rates in the wetter groundwater discharge zone than on the drier hillslope. They attributed this to lower NO3 availability and more complete denitrification to N2 in the more permanently saturated groundwater discharge zone, as opposed to the well-drained hillslope, which showed a flashier hydrology. Together with a significant decrease of NO3 in the soil solution along the hydrological flow path (Figs. 1, 4 and S2), this would support the idea that denitrifier communities in the groundwater discharge zone express N2O reductase more constitutively and

hence have inherently lower $N_2O/(N_2O+N_2)$ ratios. This was proven for the catchment in SW China by standardized laboratory incubations (Zhu et al., 2013b). Yu et al. (2016) later showed in situ that denitrification in the groundwater discharge zone indeed was an important N sink in this catchment. Cui et al. (2018) reported *ex situ* denitrification product ratios ($N_2O/(N_2O+N_2)$) for surface soils from the XSBN catchment. Integrating over 60 h of denitrification, the $N_2O/(N_2O+N_2)$ ratios significantly decreased from hillslope to groundwater discharge zone, which was a common pattern for a number of soils from (sub)tropic. I forest sites in South China.

Apparent seasonal patterns of N₂O fluxes were observed it all opographic positions (Figs. 3c and 5b), showing significantly higher N₂O emission ir the warm-wet season. Such patterns are consistent with a number of studies in the subtropice/tropics (Fang et al., 2009; Gao et al., 2018; Hall et al., 2012; Kiese et al., 2003; Zhu et al., 2013a). As shown by Table 1 and Fig. 6a&6b, both soil WFPS and temperature seemed to relay roles in regulating temporal variations of N₂O fluxes. However, it is difficult to exentangle their relative importance, given that soil temperature was also positively corrected with WFPS (significant for the whole catchment and for Position C; Fig. S4). Xu-Ri et al. (2012) summarized N₂O fluxes across climatic zones with a large range of temperature; their results indicated that temperature sensitivity of N₂O emission is mostly apparent when comparing different climatic zones. Moreover, a number of (sub)tropical studies of N₂O fluxes have reported significant impacts from soil moisture, while temperature is not a limiting factor (Gütlein et al., 2018; Kiese et al., 2002; Tang et al., 2006; Werner et al., 2006). Therefore, we consider soil moisture as a more important driver for temporal variability of N₂O emissions at XSBN.

Although mean N_2O fluxes at XSBN were small (< 20 μ g N m⁻² h⁻¹, Fig. S3) and not different from the mean rates from other unmanaged soils in the tropics (Gao et al., 2018; Hall et al., 2012;

Kiese et al., 2003; Werner et al., 2006), we observed exceptional peaks of N₂O emission on 6/17/2014 at the footslope and in particular in the groundwater discharge zone (Fig. 3c). Since such N₂O flux peaks were not seen in late summer 2013, the high N₂O emission rates in June 2014 may have been due to the dry-to-wet transition at the start of the rainy season. After the drought period between January and April 2014 at XSBN, several rain episodes occurred in May and June, of which the rain event on 6/17/2014 was largest (30 mm, Fig. 3a). In a water manipulation experiment in a tropical forest of Costa Rica (Nob.) et al., 2001), a significant increase of soil N₂O flux was observed 8 h after a rain episode. L. a review, Kim et al. (2012) found that the change of N₂O fluxes in response to soil rewetting is highest in forest ecosystems and suggested that this is due to enhanced microlal metabolism after rewetting and displacement of gas from the soil pore space through water. Furthermore, monthly precipitation anomalies in 2014 indicate that its dry sea on was drier while June and July were wetter than the long-term average (Fig. 2b); hence, the contrasting climatic conditions between dry and wet seasons in 2014 may explain the relatively large N₂O fluxes found in wet season 2014 compared to 2013 (Figs. 3c and 5c).

In addition to rain episoces, enhanced nutrient inputs in the post-drought period may drive high N_2O fluxes. We observed significant increases in TOC or DOC concentration in throughfall and soil water in the wet season 2014 compared with 2013 (Figs. 4 and S1b). The peak TOC concentration in throughfall in the middle March of 2014 (Fig. S1) may be due to the accumulation of organic matters in the canopy during the drought period. Therefore, we hypothesize that, at the start of wet season, accumulated organic matters from the canopy was flushed by rainfall to the soil and then transported laterally to the lower topographic positions, fueling microbial activities and thus N_2O production. Previous work has highlighted the

importance of lateral nutrient transport (including NO₃) along hydrological gradients driven by summer rainstorms for denitrification (Yu et al., 2019a) and N₂O emission (Enanga et al., 2016). On the other hand, studies in tropical forests have suggested that drought promotes release of organic matter to soils (e.g. root mortality) and aerobic decomposition of organic matters, which would result in more biologically available carbon in soils (Cleveland et al., 2010; Wieder et al., 2011). This is in line with the enhanced DOC availability observed in soil water at XSBN catchment after drought (Fig. 4). To confirm the importance of post drought rewetting effects for annual N₂O budgets of tropical forest soils, N₂O emission and soil moisture would have to be measured at a higher frequency (e.g. daily; Barton et al., 2015).

The observed, predominantly negative CH₄ fluxes indicate that XSBN forest soils act as a net CH₄ sink (Figs. 3d and 5d). CH₄ fluxes from soils depend on the balance between CH₄ production by methanogens and consumption by methanotrophs (Le Mer and Roger, 2001; Smith et al., 2003). Methanotrophic activity in the aerobic surface soil horizons of XSBN surpassed CH₄ production (if active) our ing most of the time. Net CH₄ uptake decreased from the hillslope to the footslope and the groundwater discharge zone (Figs. 3d and 5d), along with increasing WFPS (Figs. and 2a). Faster diffusion of atmospheric CH₄ into the soil at the drier hillslope soil may have been the most important driver for the observed enhanced uptake (Veldkamp et al., 2013; Zhang et al., 2011). The strong impact of soil moisture on CH₄ uptake is also reflected by the seasonal variation of CH₄ fluxes, showing significantly larger CH₄ uptake rates in the dry season than in the wet season (Fig. 5e). In the meanwhile, the positive correlation between soil temperature and CH₄ fluxes as shown by Table 1 and Figs. 6c&6d, may rather reflect the covariation of temperature with WFPS (Fig. S4) than indicate temperature controls on methanotrophic activity, as methanotrophs are often reported to be insensitive to temperature,

especially in tropics (Le Mer and Roger, 2001). Spatiotemporal variability of methanotrophic activity may additionally be regulated by soil inorganic N (Aronson and Helliker, 2010; Kolb, 2009). For example, soil nitrifier activity could compete with methanotrophs for O₂ and thus inhibit CH₄ oxidation. At the footslope, where CH₄ uptake rates were smaller than at the hillslope (Fig. 5d), we observed higher NO₃⁻ concentrations in soil water (Figs. 1, 4 and S2), which may reflect a higher nitrification activity there. The significant negative effect of NO₃⁻ concentration on CH₄ uptake rates at the hillslope as indicated by reason correlation (Table S1) supports such regulation.

The topographic gradient of CH₄ fluxes observed at YSBN is similar to findings from several other studies (Itoh et al., 2009; Kaiser et al., 2018; Yu et al., 2019b). However, while previous studies reported net uptake at the higher and ne. emission at the lower topographic positions of the catchments, we rarely observed net CH₄ emission at XSBN catchment, even in the groundwater discharge zone where Wr. Swas highest in the wet season (Figs. 3d and 5e). This most likely indicates inadequate chaer biosis (non-saturated soils) and/or biologically available carbon for methanogenesis in swils (Le Mer and Roger, 2001). In other catchments reporting net CH₄ sources, wetland or ipar an soils with high groundwater table levels were identified (Itoh et al., 2009; Kaiser et al., 2018), which also provide biodegradable organic matter (Pacific et al., 2011) for methanogenesis. Another possible explanation is that, in the deep soils of groundwater discharge zone, most newly produced CH₄ was consumed by the methanotrophs in the surface and sub soils, under fluctuations between oxic and anoxic conditions (Conrad, 2002; Kolb and Horn, 2012).

The mean CH₄ sink strength over 1.5 years at the three topographic positions varied from 60 to 80 µg C m⁻² h⁻¹, which is at the higher end of what is commonly reported for humid tropical

forest soils (Courtois et al., 2018; Hall et al., 2012; Kiese et al., 2003; O'Connell et al., 2018; Werner et al., 2006; Wood and Silver, 2012). Highest CH₄ uptake rates at XSBN were observed during the drought period (Figs. 3d and 5e). Wood et al. (2012) excluded throughfall from a humid tropical forest soils and found that drought enhanced CH₄ uptake by up to 5 times. Likewise, other studies reported significant increase in CH₄ consumption during dry seasons (Kiese et al., 2003; O'Connell et al., 2018; Tang et al., 2006). In the post-drought period, one study in a humid tropical forest of Puerto Rico looked into the shifts of CH₄ fluxes (O'Connell et al., 2018). They discovered a dramatic increase in net emission of CH₄ from soils in all topographic positions, fueled by release of biodegradable of anic matter during rewetting, which largely offset the CH₄ sink from the well-drained soils juring drought. Also, at our site, we observed a significant increase in DOC concentration in soil water during the post-drought period (Figs. 4 and S2). Although N₂O em. ssions increased noticeably (June 2014) relative to the previous wet season (Figs. 3c and 5f), CH₄ fluxes did not differ from those in the previous drought period (Figs. 3d, 5e and 5f). It is noteworthy that WFPS in the post-drought period was always below 80% (Fig. 3b), in licating that soils in the groundwater discharge zone were not saturated during our observation. This suggests again the importance of soil water saturation for net CH₄ emissions, and cyptains why rewetting after drought did not stimulate CH₄ emission at XSBN (Fig. 3d). In addition, our observation may be explained the suppression of methanogenesis due to competition with denitrification for organic carbon in the post-drought period (Conrad, 2002; Le Mer and Roger, 2001).

We estimated annual N_2O and CH_4 budgets with cumulative fluxes between September 2013 and August 2014 (Table 2) and tested two scenarios, including one considering the N_2O flux anomalies on 6/17/2014. The full dataset (Scenario 1) shows that the groundwater discharge zone

contributed the highest N₂O source (1.94±1.25 kg N ha⁻¹ yr⁻¹) among the three topographic positions, which was largely due to the exceptionally high flux (450 μg N₂O-N m⁻² h⁻¹) on 6/17/2014. Even though the groundwater discharge zone accounts for only < 1% area of this catchment, it may be a hotspot for elevated N₂O emissions during post-drought, adding uncertainty to the catchment N₂O budget. If the flux anomalies were not considered (Scenario 2), the annual N₂O flux was largest on the hillslope. In both scenarios, there was a variable CH₄ sink across different topographic positions in the catchment, ranging from 5.27 to 7.47 kg CH₄-C ha⁻¹ yr⁻¹ (Table 2). This CH₄ sink is clearly larger than the global avalage sink strength (Yu et al., 2017), which can be largely attributed to the stimulated Couptake during drought, particularly in the well-drained soils of the hillslope (Figs. 3d and 5e). Considering the net GWPs associated with the two GHGs, we found that N₂O emissior s² an icantly surpassed CH₄ uptake (Scenario 1, Table 2); by contrast, if N₂O flux an ma'res were excluded, the CH₄ sink at the lower topographic positions of the catchment yould well balance the GWP of N2O (Scenario 2, Table 2). A recent report has suggested that Yunnan region (SW China), where XSBN is located, is experiencing more extreme weight dry events, i.e., wet seasons become wetter and dry seasons become drier (Liu et al., $2^{0.1}4$). As a result, seasonal variations of N₂O and CH₄ fluxes would be further enlarged; in we seasons, the hillslopes which account for more than 90% of the catchment area, may act as an enhanced N₂O source with a weakened CH₄ sink, while the groundwater discharge zone may contribute a notable CH₄ source (Yu et al., 2019); in dry seasons or even drought, CH₄ uptake would substantially rise and N₂O emission would decline, but such C sink might be largely offset by stimulated N₂O emission in the post-drought periods (this study). Overall, as budgeted in Table 1, future climate change in this region may further

stimulate N_2O emissions, and such positive contribution of GWP is unlikely to be significantly alleviated by the soil CH_4 sink.

5. Conclusions

Observing N₂O and CH₄ fluxes over 1.5 years at different landscape positions in the XSBN tropical forested catchment, we found pronounced topographic control on N₂O emissions and CH₄ uptake. Hydrologically driven variability in WFPS was identified as the dominant factor controlling N₂O and CH₄ fluxes, both in space and time: N₂O emission and CH₄ uptake decreased significantly from the drier hillslope soils to the witter soils in the groundwater discharge zone; by contrast, soil N₂O source was dampened in the any season relative to the wet season while the soil CH₄ sink was enhanced. In response to rain episodes, hydrological transport of labile organic carbon might have played a role in fulling episodically large post-drought N₂O emissions at the lower topographic positions of the catchment. However, such "post-drought" effects were not found for CH₄ fluxes, most miles, due to inadequate anoxia (low WFPS) despite rewetting. By evaluating the GWPs associated with these two GHGs, we found that the contributions from N₂O emission were significantly larger than CH₄, but the quantification of their relative importance across to ographic positions requires better constraints of fluxes. With projected climate change for the tropical region in South China (Liu et al., 2014), topographic patterns of N₂O emission and CH₄ consumption in tropical rainforests may be significantly altered, further adding uncertainty to regional estimates of GHG budgets.

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Figures and Tables

Figure 1 Schematic illustration of topography and sampling locations in the studied rainforest catchment at XSBN. Plots A, B and C denote the three topographic positions upper hillslope, foot of the hillslope and groundwater discharge zone, respectively. Right insert: box and whisker plots of soil WFPS (at 10 cm depth), NH_4^+ -N and NO_3^- -N and DON concentrations in soil water (O/A layer) measured along a hydrological continuum given by throughfall \rightarrow position A \rightarrow position B \rightarrow position C \rightarrow outlet. N concentrations were derived from measurements during the wet seasons of 2013 and 2014 only; no samples were obtained during the dry seasons. Mean values are indicated by red dashed lines.

Figure 2a Annual precipitation at XSBN during 2011-2015 (the lowest value found for 2014 is marked in red). **2b** Monthly precipitation anomalies in 2014 (vs. 2) 11-2015).

Figure 3 Time-series of climatic and soil factors as well as GHC theres measured between July 2013 and December 2014 at topographic positions A, B and C Paralla shows daily precipitation, throughfall, and soil WFPS at 10-cm depth; panel b shows soil temperature at 10-cm depth; panels c and d show mean N₂O and CH₄ fluxes, with error cars indicating 1 SD. Note the logarithmic scale for N₂O emission rates in panel c. The same data plotted on a linear scale can be found in the supplement (Fig. S3). All data except precipitation were collected bi-weekly. Shaded area indicates the dry seasons.

Figure 4 N concentrations (NH₄⁺, NO₃⁻ and DC N), COC and pH of soil water sampled from the O/A horizon at three topographic positions (to) panel: A; middle panel: B; bottom panel: C). In the dry seasons, the collected volumes of so. water were insufficient for chemical analyses.

Figure 5 Box and whisker plots of N_2O (> **b** and **c**) and CH_4 (**d**, **e** and **f**) fluxes between July 2013 and December 2014 at topographic positions A, B and C. Panels **a** and **d** compare fluxes at three topographic positions (for N_2O fluxes at C, datasets with and without the exceptional flux on 6/17/2014 were included); panels **b** and **e** compare average fluxes between wet and dry seasons; panels **c** and **f** compare Tuxes between the wet seasons in 2013 and 2014 (fluxes between July and October were elected for comparison between two wet seasons, according to the availability of data). Mean values are indicated by red dash lines. Different letters indicate significant difference (n < 0.1).

Figure 6 Relationship of Γ_2 O (logarithmically transformed; sub-figures **a** and **b**) and CH₄ (**c** and **d**) fluxes with soil temperature and WFPS at three topographic positions along the rainforest catchment XSBN. Refer to Figure 1 for plot codes. Soil WFPS and temperature were measured at 10 cm depth. Linear regression lines (only shown if significant; p < 0.1) with linear polynomials are shown in each figure.

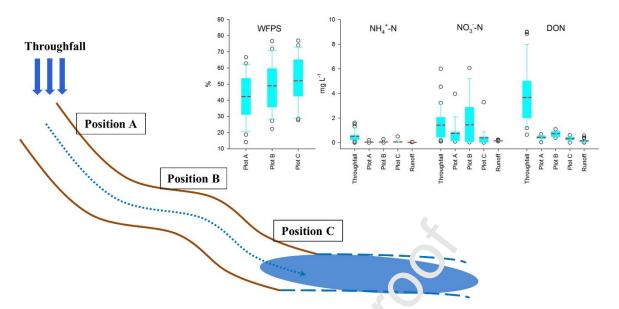


Figure 1

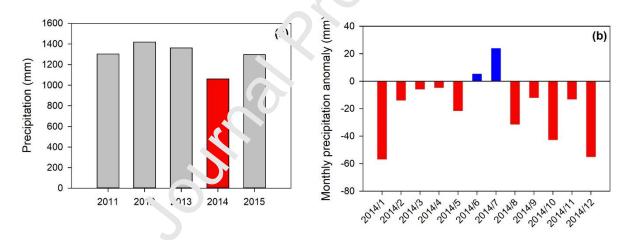


Figure 2

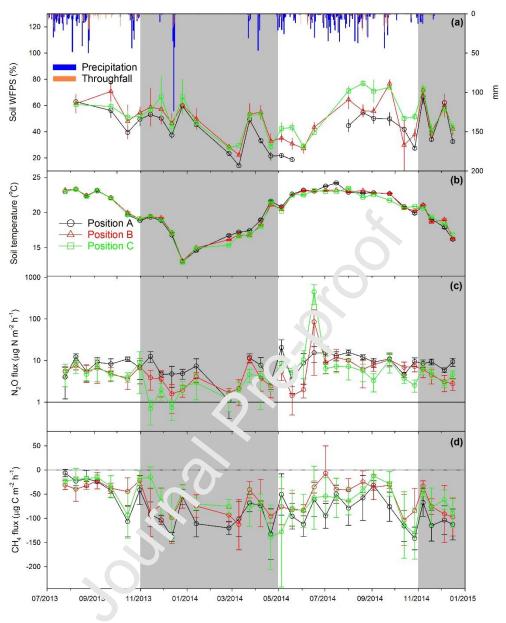


Figure 3

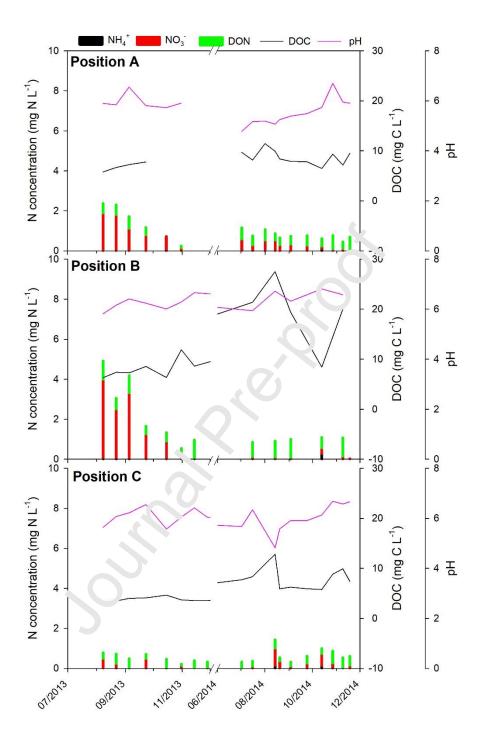


Figure 4

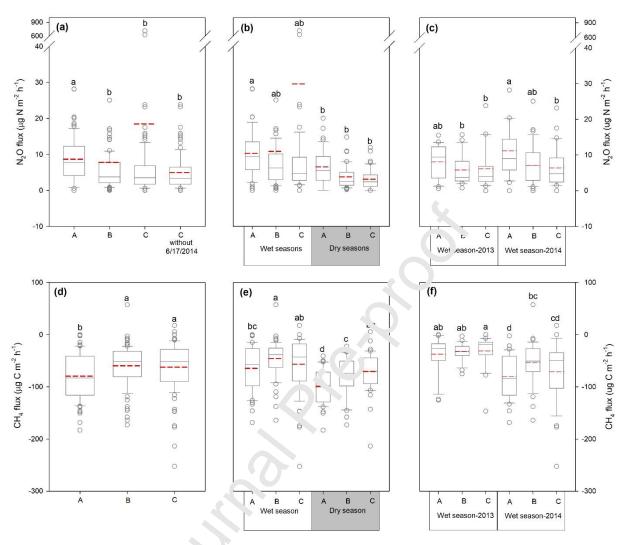


Figure 5

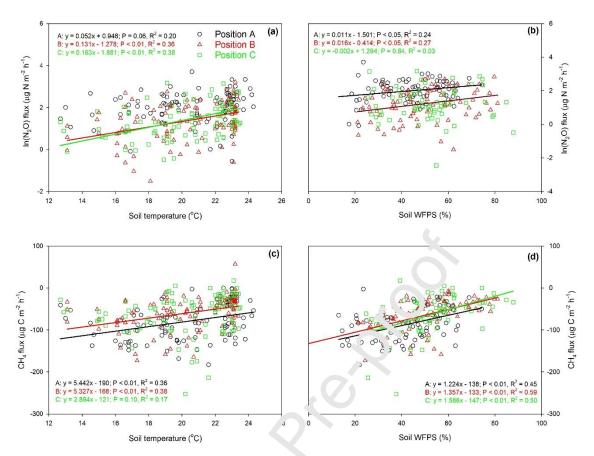


Figure 6

Tables

Table 1 Stepwise linear regression of N2O and CH4 fluxes against soil parameters§

	Soil WFPS		Soil Ten	nperature	Paradan	D ² A 1;
	Slope	p	Slope	p	Equation	R ² -Adjust
ln(N ₂ O)	-0.017	0.04	0.30	< 0.01	$ln(N_2O) = 0.30 \text{ Temp -} 0.017WFPS -3.66$	0.35
CH ₄	1.71	< 0.01	8.30	0.03	CH ₄ = 1.71WF 'S + \.30Temp - 334	0.56

[§] Soil parameters including soil WFPS and temperature (Temp), pH, DOC, NH_4^+ -N and NO_3^- -N were tested for stepwise linear regression. Soil WFPS and entire were measured at 10 cm depth. All other soil parameters were determined in the C/A layer. Parameters other than Temp and WFPS showed insignificant slopes (P > 0.1) in the regression and were excluded during stepwise regression.

Table 2 Annual fluxes of N_2O and CH_4^{θ}

		Upper hillslope Position A		Foot of hillslope Position B		Groundwater discharge zone Position C	
		Fluxes	Uncertainty	Fluxes	Uncertainty	Fluxes	Uncertainty
	N ₂ O (kg N ha ⁻¹ yr ⁻¹)	0.76	0.07	0.71	0.59	1.94	1.25
Scenario 1 [†]	CH ₄ (kg C ha ⁻¹ yr ⁻¹)	-7.32 [*]	0.97	-5.27	1.?9	-5.73	1.61
geenario 1	Net GWP (equivalent to kg CO ₂ ha ⁻¹ yr ⁻¹) [§]	43.2	46.5	9< 9	251	594	524
	N ₂ O (kg N ha ⁻¹ yr ⁻¹)	0.75*	0.05	0.43	0.20	0.41	0.16
Scenario 2 [†]	CH ₄ (kg C ha ⁻¹ yr ⁻¹)	-7.47*	ો.9ડ	-5.31	1.49	-5.78	1.53
Section 2	Net GWP (equivalent to kg CO ₂ ha ⁻¹ yr ⁻¹) [§]	33 1	40.5	-19.2	100	-45.1	87.8

⁶ Annual budgets were estimated be set on cumulative fluxes of N₂O and CH₄ from September 2013 to August 2014. Presented fluxes are no an values computed from cumulative fluxes at triplicates, with 1 SD representing uncertainties. A, R and C refer to three topographic positions upper hillslope (A), foot of the hillslope (B) and groundwater dust arge zone (C).

[§] Global warming potential of N₂O and CH₄ (over 100 years) are calculated as 265 and 28 times that of CO₂.

 $^{^{\}dagger}$ As exceptionally high N₂O fluxes occurred on 6/17/2014, N₂O and CH₄ fluxes were computed for two scenarios, of which the first took all flux data into account and the second replaced fluxes on 6/17/2014 with the average of the previous and the following sampling. Therefore, net GWP was also computed for two scenarios.

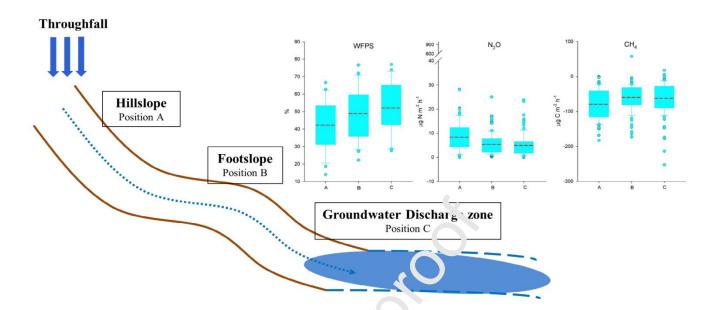
^{*} Significant difference among topographic positions.

Author contributions

J. Zhu and W. Zhou conceived this project and designed the field experiment. W. Zhou led the field sampling and laboratory analyses; H. Ji, X. Bai, Y. Lin, Y. Zhang, L. Sha, Y. Liu and Q. Song provided support during the whole sampling and data acquisition process. L. Yu, J. Zhu and W. Zhou performed data analysis and planed for the manuscript. L. Yu wrote the main manuscript; J. Zhu, W. Zhou, P. Dörsch and J. Mulder were involved in the revisions of the article. All authors were involved in the scientific discussions and commenting on the manuscript.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.



Graphical abstract

Highlights

- Significant controls of topography on N₂O and CH₄ fluxes in a tropical catchment
- Response of GHG fluxes to soil water contents at different topographic positions
- Weaker N₂O emission and stronger CH₄ uptake in dry seasons
- N₂O fluxes, but not CH₄ fluxes, respond significantly to post-drought rainfalls