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LETTER

Phosphorus alleviation of nitrogen-suppressed methane sink in global grasslands

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Abstract

Grassland ecosystems account for more than 10% of the global CH₄ sink in soils. A 4-year field experiment found that addition of P alone did not affect CH₄ uptake and experimental addition of N alone significantly suppressed CH₄ uptake, whereas concurrent N and P additions suppressed CH₄ uptake to a lesser degree. A meta-analysis including 382 data points in global grasslands corroborated these findings. Global extrapolation with an empirical modelling approach estimated that contemporary N addition suppresses CH₄ sink in global grassland by 11.4% and concurrent N and P deposition alleviates this suppression to 5.8%. The P alleviation of N-suppressed CH₄ sink is primarily attributed to substrate competition, defined as the competition between ammonium and CH₄ for the methane mono-oxygenase enzyme. The N and P impacts on CH₄ uptake indicate that projected increases in N and P depositions might substantially affect CH₄ uptake and alter the global CH₄ cycle.

Keywords

Grassland, methane, nitrogen, phosphorus, substrate competition theory.

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INTRODUCTION

Methane (CH₄) is an important greenhouse gas, contributing c. 20% to the anthropogenic climate warming (IPCC 2013). Grasslands cover more than 20% of the Earth's land surface and typically serve as a net sink for atmospheric CH₄ (Aronson & Helliker 2010; Dijkstra *et al.* 2013). However, this CH₄ sink may be altered by the growing input of nutrients, particularly nitrogen (N) (Galloway *et al.* 2008), due to anthropogenic activities. While previous studies have reported various impacts of N (particularly NH₄⁺) addition on the oxidation of atmospheric CH₄ in soil (Bollag & Czlonkowski 1973; Liebig *et al.* 2008), it has become clear that N addition typically suppresses this oxidation process, with few exceptions (Aronson & Helliker 2010; Liu *et al.* 2017). This N-induced suppression of CH₄ oxidation has been attributed to two mechanisms. First, CH₄ and NH₄⁺ compete for the same methane mono-oxygenase (MMO) enzyme (Gulledge *et al.* 2004), which can oxidise both CH₄ (to CH₃OH) and NH₄⁺ (to NO₂⁻) due to the similar molecular structure of CH₄ and NH₄⁺ (Dunfield & Knowles 1995; Gulledge *et al.* 2004). Second, the intermediates and end products (primarily nitrite)

of methanotrophic NH₄⁺ oxidation can be toxic to methanotrophic bacteria, explaining the inhibition of CH₄ consumption under increased N inputs (Schnell & King 1994; Bodelier & Laanbroek 2004). It should be noted that one distinction between CH₄ and NH₄⁺ is that methanotrophic bacteria grow on CH₄ (Davidson & Schimel 1995), but not on NH₄⁺ as the oxidising NH₄⁺ does not support CO₂ fixation for cell growth (Bédard & Knowles 1989; Gulledge & Schimel 1998).

While the negative impact of N addition is thus well understood, phosphorus (P) addition may alter this N-induced suppression on CH₄ oxidation. In a recent study, P addition alone was reported to enhance CH₄ oxidation, likely due to the alleviation of P limitation on methanotroph (Veraart *et al.* 2015). Given that P-fertiliser inputs and atmospheric P deposition are also increasing globally (Mahowald *et al.* 2008; Penuelas *et al.* 2013), investigating P impacts on CH₄ uptake rate, and especially its interactions with the increasing N inputs, is important for better understanding the role that aerobic soils play in the global atmospheric CH₄ cycle.

Atmospheric deposition and fertiliser inputs of N and P have increased across the globe since the industrial revolution (Mahowald *et al.* 2008; Penuelas *et al.* 2013; Brahney *et al.*

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2015). Increased inputs of N may reduce the CH₄ sink strength of grassland ecosystems, with potentially important feedbacks to the global climate system (Templer *et al.* 2012; Tian *et al.* 2015). The interactive effect of N + P additions on CH₄ oxidation has been reported as neutral (Lund *et al.* 2009) or positive (Zhang *et al.* 2014), and P has been found to alleviate the N suppression on CH₄ oxidation (Zhang *et al.* 2014); however, the mechanisms by which P alters the N suppression of atmospheric CH₄ oxidation in soils remain elusive. We therefore integrated a 4-year field manipulation experiment, a meta-analysis and an empirical modelling approach to investigate the effects of N + P additions on CH₄ flux in grasslands and the underlying mechanisms.

MATERIALS AND METHODS

Field experiment

The field experiment was conducted in Duolun County (42°02' N, 116°70' E, 1324 m a.s.l.), a semi-arid temperate steppe in Inner Mongolia, northern China. The multiple-year mean annual precipitation and mean annual temperature are *c.* 385 mm and 2.1 °C respectively. The topography is characterised by low foothills at elevations of 1150–1800 m. Soil is Calcis-orthic Aridisols (the US Soil Taxonomy classification), with $62.75 \pm 0.04\%$ sand, $20.30 \pm 0.01\%$ silt and $16.95 \pm 0.01\%$ clay (Wu *et al.* 2010). The dominant plant species include *Stipa krylovii*, *Artimesia frigida*, *Potentilla acaulis*, *Cleistogenes squarrosa*, *Allium bidentatum* and *Agropyron cristatum*.

To assess the nitrogen (N) and phosphorus (P) impacts on CH₄ flux, a block design experiment with different combinations of N and P additions was established in early 2013 and run to 2016. A complete random design, with three block replicates, was adopted to address the high spatial heterogeneity. There were four experimental treatments (i.e. control (CK), N addition (N), P addition (P) and both N and P additions (N + P)), which were randomly assigned to four 6 × 6 m plots in each block. Two chambers for greenhouse gas measurement were set up in each plot. All blocks were separated with a 3-m buffer zone. The N and P were applied twice per month from May to September during 2013–2016; we sprayed the fertiliser solution to ensure that the application was evenly distributed in the plots. The dose of N addition was 100 kg N ha⁻¹ y⁻¹ as NH₄NO₃ solution; this dose was selected because it meets the N required to sustain the local maximum vegetation productivity (Bai *et al.* 2010). We monitored the atmospheric N deposition and estimated it to be 20.4 kg N ha⁻¹ y⁻¹ in our experimental site in 2012. The dose of P was 100 kg P ha⁻¹ y⁻¹, equivalent to previous nutrient addition experiments in grasslands (Phoenix *et al.* 2003). The plots with both N and P additions received the same amounts of N and P as in the N-only or P-only addition treatments. Control plots were not fertilised, but rather watered with the same amount of water as used in the fertiliser solutions; the water used to dissolve N and P was *c.* 800 ml, which is equivalent to 0.8 mm for a 1 m² plot, a pre-treatment field experiment found that this small amount of water addition did not cause any significant changes in

water supply, thus unlikely to have altered the ecosystem functions.

Over the study period, soil and plant properties were measured (Supplementary Online Materials). Soil samples were collected to a depth of 10 cm once per year during 2013–2016. Six soil cores were randomly taken in each plot and mixed completely. The measured soil variables included ammonium (NH₄⁺) and nitrate (NO₃⁻), soil pH, soil organic carbon (SOC), soil total nitrogen (STN), soil total phosphorus (STP), soil microbial biomass carbon (MBC) and soil microbial biomass nitrogen (MBN). It should be noted that no nitrite was detected at our field site. The measured vegetation variables included aboveground biomass, plant total carbon (PTC) and total nitrogen (PTN).

Field measurements of CH₄ Flux

A static chamber technique was used to measure the CH₄ flux (Song *et al.* 2009; Zhang *et al.* 2017). Stainless-steel permanent bases (50 × 50 × 12 cm) with a 3-cm-deep groove for water sealing were inserted into soil down to a depth of 12 cm in the plots *c.* 1 month before the experiment started in the first year. The chamber base was left in the field for 1 month before any flux measurement, avoiding potential disturbances. The CH₄ flux was measured weekly from May to September in 2013, 2014, 2015 and 2016 respectively. Detailed information for the chamber setup, CH₄ concentration measurement and flux calculation can be found in the supplementary online material.

The CH₄ fluxes were linearly interpolated and sequentially cumulated to estimate the total flux over growing seasons (Zhang *et al.* 2017). The linear interpolation was adopted due to two reasons: (1) it has been widely used in previous studies and has been suggested to be effective for seasonal interpolation of ecological variables (Song *et al.* 2009; Nikiema *et al.* 2011); (2) the auxiliary data to assist annually interpolation were lacking. Throughout this analysis, positive fluxes represent gas uptake by the grassland ecosystems.

Meta-analysis

A meta-analysis was conducted to investigate the N and P impacts on CH₄ across global grasslands. We collected publications by searching for “nitrogen and phosphorus”, “methane”, “grassland” and “upland” in Google Scholar in July 2016, and later updated in September 2018. The search returned 6230 publications. A few criteria were then used to screen these publications for our purpose. The criteria applied to determine whether or not to use the data were as follows: (1) it must be manipulation experiments with either external N and/or P addition; (2) the field measurements must cover at least one full growing season, which makes the estimation of annual budget more reliable and (3) the studies report clear information for the field site that is useful when extracting edaphic and meteorological data from global datasets. For sites without clear latitude and longitude, we Googled and found their geographic coordinates with country and site names. Finally, 35 papers were selected for data extraction. When the data were presented in figures, we extracted mean

values and standard errors using GraphClick (<http://www.ari-zona-software.ch/graphclick/>). For studies with measurements from different N and P addition levels in one paper, they were extracted and treated as independent data points.

Finally, we compiled a comprehensive dataset of 382 data points for meta-analysis (Figs S4 and S5). The dataset covers the major grassland types across the globe and all experiments were carried out between 1980 and 2017. For all field experiments, the atmospheric depositions of N and P from the global dataset were treated as background rate and were added to the reported N and/or P addition rates. As a majority of the data points from Yu *et al.* did not contain N or P deposition (Yu *et al.* 2017), we used the extracted contemporary nutrient deposition (Mahowald *et al.* 2008), based on latitudes and longitudes, as nutrient inputs. Every CH₄ flux rate corresponds to one site with auxiliary information including latitude, longitude and factors such as N deposition, P deposition, soil temperature (ST), soil moisture (SM), soil pH, soil organic carbon (SOC), bulk density (BD) and clay content (CL). Across the studies in our dataset, the N and P treatments fell within the ranges 0 ~ 200 kg N ha⁻¹ yr⁻¹ and 0 ~ 200 kg P ha⁻¹ yr⁻¹ (Mori *et al.* 2013; Zheng *et al.* 2016) respectively. The non-growing season CH₄ flux is normally not available for most field experiments, therefore, the annual rate of CH₄ uptake (kg C-CH₄ ha⁻¹ year⁻¹) thereafter was calculated by the ratio of the growing season to non-growing season CH₄ uptake as reported by a few studies (Li *et al.* 2012; Yue *et al.* 2016).

The soil properties for each point data site were extracted from global datasets according to their latitudes and longitudes. Specifically, the soil pH, SOC, BD and clay were retrieved from the Re-gridded Harmonized World Soil Database v1.2 in the Oak Ridge National Laboratory Distributed Active Archive Center for Biogeochemical Dynamics (available online: <https://daac.ornl.gov/SOILS/guides/HWSD.html>); soil temperature and moisture for the top 10 cm were from the NCEP/DOE AMIP-II Reanalysis (Reanalysis-2): <http://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.derived.surfaceflux.html>.

Global extrapolation

The data for the grassland distribution were derived from the Global Mosaics of the standard MODIS land cover-type data product (MCD12Q1) from <http://glcf.umd.edu/data/lc/index.shtml>. The spatial coverage is -180.0° ~ 180.0° in longitude and -64.0° ~ 84.0° in latitude. The global land cover datasets are available as ESRI ASCII Grid format files and were reprojected to be consistent with soil and climate datasets (Fig. S4). The global simulations were carried out at a spatial resolution of 0.5° × 0.5°. According to the definition of Food and Agriculture Organization of the United Nations (FAO), any geographic area dominated by natural herbaceous plants including grasslands, prairies, steppes and savannahs with coverage of at least 10% was designated as grassland (Latham *et al.* 2014). The annual CH₄ uptake was determined by summing up all grassland grids (eqn 1). For the model simulation, we set N and P depositions to pre-industrial level to represent ambient condition (90% range: 0.05 ~ 2.94 kg N ha⁻¹ year⁻¹ and

0 ~ 0.08 kg P ha⁻¹ year⁻¹), both N and P were set to contemporary level to represent N and P concurrent treatment (90% range: 1.07 ~ 13.06 kg N ha⁻¹ year⁻¹ and 0.003 ~ 0.24 kg P ha⁻¹ year⁻¹), N set to pre-industrial level and P set to contemporary level to represent P addition only, P set to pre-industrial and N set to contemporary to represent N addition only.

Both climate and soil data were resampled and re-projected to be consistent through NCL (NCAR Command Language, current version 6.4.0). The data with higher resolution were resampled to the lowest resolution at 0.5° × 0.5°. Based on the calculated CH₄ uptake rate in every grid and grid area of grassland, we scaled up the results from this analysis by multiplying the estimated CH₄ uptake with the corresponding grid areas:

$$F(\text{CH}_4) = \sum_{k=0}^n \binom{n}{k} \text{CH}_{4k} A_k \quad (1)$$

where $F(\text{CH}_4)$ is the sum of the global grassland CH₄ uptake expressed as Tg C-CH₄ year⁻¹, and CH_{4k} is the CH₄ uptake rate in k th grid as kg C-CH₄ ha⁻¹ year⁻¹, and A_k is the area of the k th grid (Fig. S8).

Statistical analysis and uncertainty analysis

We used a one-way ANOVA analysis (ANOVA; R package) followed by Duncan's *post hoc* test to examine the N and P impacts on annual CH₄ uptake rate, soil NH₄⁺ and NO₃⁻, and plant N content. A stepwise multivariate analysis was adopted to evaluate the response of the soil plant and microbial communities to environmental factors as well as their correlations with each other. A number of key variables, including N input rate, P input rate, soil temperature, soil pH, soil organic carbon density, bulk density and clay content, are kept in the multiple linear equation to quantify their impact on CH₄ flux (eqn 2).

A structure equation model (SEM) approach was used to explore how nutrient (N, P and N + P) additions alter the CH₄ uptake. The “lavaan” package (<https://cran.r-project.org/web/packages/lavaan/lavaan.pdf>) for R program (version 3.4) was used for the structural equation modelling. Data used in the SEM and linear regression analyses were calculated as the mean of every year during the 4 years (Figs S2 and 3). Prior to the SEM procedure, we conducted principal component analysis (PCA) to remove the redundant variables for meteorological factors, soil edaphic parameters, plant and microbial parameters. Meanwhile, due to the strong multicollinearity among other variables, the variance inflation factor (VIF) was used to quantitatively select the right factor for SEM model (VIF < 5). One SEM model (Fig. S3) was used to disentangle the direct and indirect environmental impacts on the CH₄ uptake, and the other (Fig. 3) was used to analyse the N and P impacts on CH₄ uptake. The fitted conceptual model was further modified with “mod_ind” (within the R package “lavaan”) to yield a reliable multivariate causal network. The SEM results were evaluated with the comparative fit index (CFI), the normed fit index (NFI) and the chi-square test (χ^2). The chi-square value is the traditional measure for evaluating the overall model fit and accessing the magnitude of discrepancy between the sample and fitted covariance matrices; NFI is an incremental fit index that assesses the model by

comparing the chi-square value of the model to the null model that assumes that all variables are uncorrelated; CFI is a revised form of the NFI that takes into account sample size; the model fits with NFI > 0.95 and CFI > 0.95, indicating a good SEM (Hooper *et al.* 2008).

The Latin Hypercube Sampling based Monte Carlo method was adopted to quantify the uncertainties of the global grassland CH₄ uptake under different treatments (Control, N, P and N + P conditions) estimated by the empirical models. The LHS approach has been used in our previous study (Xu 2010); detailed procedure can be found in Supplementary Online Material.

RESULTS

A field experiment for N and P impacts on CH₄ uptake

The field experiment showed that the N-only and concurrent N + P additions treatments significantly suppressed CH₄ uptake over the entire duration of the experiment (2013–2016, Fig. 1a). Suppression of CH₄ oxidation by N addition and N + P additions amounted to −16.3% ($P < 0.01$) and −7.9% ($P = 0.04$) relative to the control plots in 2013, −7.8% ($P < 0.01$) and −1.8% ($P = 0.10$) in 2014, −12.3% ($P < 0.01$) and −6.6% ($P < 0.01$) in 2015, and −18.3% ($P < 0.01$) and −12.5% ($P < 0.01$) in 2016 respectively (Fig. 1d). Over the study period, the average annual CH₄ uptake rates were 2.94 ± 0.07 kg C-CH₄ ha^{−1} in control plots, 2.54 ± 0.04 kg C-CH₄ ha^{−1} in N addition plots and 2.73 ± 0.06 kg C-CH₄ ha^{−1} in N + P addition plots (Fig. 1a). In comparison with the ambient treatment, the P-only treatment slightly and not statistically significantly stimulated CH₄ uptake throughout the experiment: +7.4% ($P = 0.12$) in 2013, +4.0% ($P = 0.09$) in 2014, +1.2% ($P = 0.2$) in 2015 and +2.3% ($P = 0.16$) in 2016 respectively (Fig. 1d). Overall, P addition alone thus did not affect CH₄ uptake, N addition alone suppressed CH₄ uptake and concurrent N + P additions suppressed CH₄ uptake to a lower degree, suggesting that P addition alleviates the N suppression of CH₄ uptake in semi-arid grasslands (Fig. 1a and d).

A Meta-analysis of N and P Impacts on CH₄ Uptake in Grasslands

To investigate the universality of these N and P effects on CH₄ uptake in grassland soils, we carried out a global meta-analysis to quantify the responses of CH₄ uptake rates in grassland ecosystems to nutrient addition treatments (N-alone, P-alone and concurrent N + P additions) (Fig. 2). This global meta-analysis showed that, relative to the control treatments, N addition and concurrent N + P additions cause significant declines in annual CH₄ uptake: −27% ($P < 0.01$) and −14% ($P < 0.01$) respectively (Fig. 2). The smaller inhibition effect of concurrent N + P additions than the N-only addition on CH₄ uptake ($P = 0.02$) (Fig. 2) is consistent with the results of our field experiment (Figs 1a and 2).

Mechanisms of N and P impacts on CH₄ uptake

Based on various statistical analysis, we hypothesise that the following mechanisms underpin the N and P impacts on CH₄

uptake. Whereas experimental N addition leads to an accumulation of mineral N (NH₄⁺ and NO₃[−]) in the soil (N-only compared with control), P addition enhances plant N uptake and N accumulation in vegetation biomass (N + P treatment compared with N-only addition) (Figs 1b and c, 3a1, 3d; Fig. S1). In our field experiment, P-only addition significantly decreased soil NH₄⁺ content ($P < 0.01$, Fig. 1b), but did not significantly affect plant N content (P-only addition compared with control) ($P = 0.35$, Fig. 1c). The concurrent N + P additions, in contrast, did stimulate N accumulation in vegetation to a much higher degree than N addition alone ($P < 0.01$, Fig. 1c). This increase in vegetation N accumulation significantly reduced soil NH₄⁺ content under the N + P additions treatment compared with the N-only treatment ($P < 0.01$, Fig. 1b). Therefore, the P addition substantially stimulates plant growth and plant uptake of NH₄⁺ (Dijkstra *et al.*, 2013), which reduces the NH₄⁺ concentration in soils and thereby alleviates the N suppression of CH₄ uptake (Fig. S1).

To disentangle the contribution of the various potential drivers, we applied structural equation models (SEM) to quantify the environmental controls of CH₄ uptake (all factors were classified into four groups: meteorology, microbes, plant and edaphic factors, Figs S2 and S3) using the field experiment's data (Fig. 3a–d). Our models considered how nutrient additions directly or indirectly affects CH₄ uptake (Fig. 3 and Fig. S3). The SEM results suggest that under control (non-fertilised) conditions, the impact of meteorology on CH₄ uptake was directly ($\beta = -0.33$, standardised coefficient) or indirectly mediated through microbes ($\beta = -0.8$, standardised coefficient) and plants ($\beta = -0.21$, standardised coefficient) (Fig. S3a). N and P additions affected CH₄ uptake indirectly through edaphic factors (N treatment: $\beta = 0.5$, standardised coefficient, Fig. S3b; N + P treatment: $\beta = 0.37$, standardised coefficient, Fig. S3d). Soil NH₄⁺ had a direct negative effect and plant N content had a positive effect on the CH₄ uptake under control conditions and in all nutrient addition treatments (Fig. 3a–d). Meanwhile, added N stimulated the accumulation of soil NH₄⁺ (N treatment: $\beta = 0.45$), added P had a positive impact on plant P and soil P, but had a negative yet insignificant impact on soil NH₄⁺; while the N + P treatment had a negative impact on soil NH₄⁺ ($\beta = -0.18$). Compared with the control, the N addition strengthened the negative effect of NH₄⁺ on CH₄ uptake (β ranging from −0.57 to −0.77) (Fig. 3a and b); P addition did not change the negative effect of NH₄⁺ ($\beta = -0.57$) and the positive effect of plant N ($\beta = 0.34$) (Fig. 3a and c); N + P additions alleviated the suppression effect of soil NH₄⁺ (β changing from −0.77 to −0.72) (Fig. 3b and d). These results showed that CH₄ uptake is highly associated with soil N and P contents in semi-arid grasslands.

Global Estimation of P alleviation of N-suppressed CH₄ Sink in Grasslands

We further developed an empirical model to quantify the N and P impacts on soil oxidation of atmospheric CH₄ across global grasslands, using existing global datasets of soil properties and meteorology (Methods). Two thirds of the compiled data were used for model fitting, whereas the remaining one third of the data were used for model validation (Fig. S7).

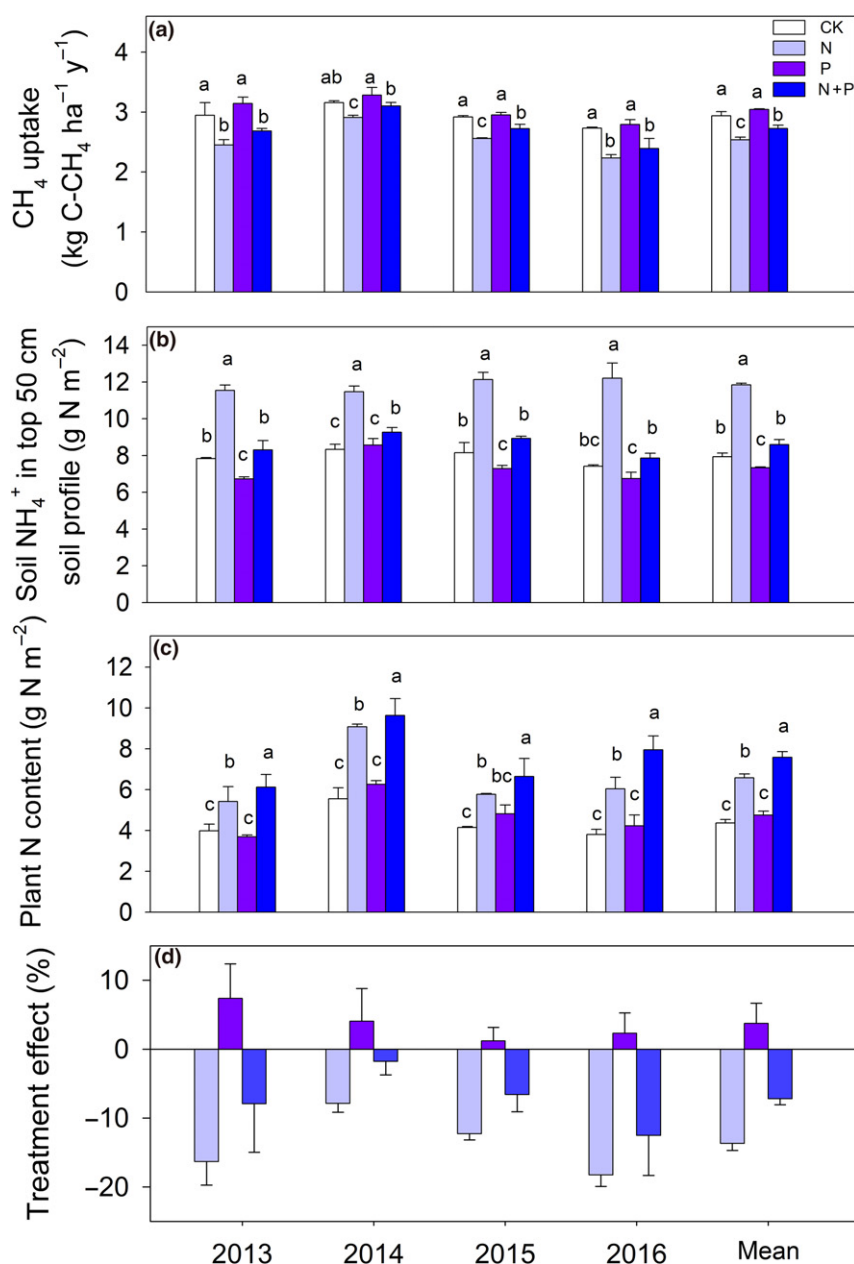


Figure 1 Differential effects of N, P and concurrent N and P additions on surface CH₄ flux, soil NH₄⁺ and plant N content in semi-arid grassland from 2013 to 2016. (a) Net nutrient effects on mean CH₄ uptake; (b) Net nutrient effects on soil NH₄⁺ content in top 50 cm and (c) Net nutrient effects on plant N content. The error bars indicate the standard error of means ($n = 3 \times 2$). Different letters represent significant differences among treatments ($P < 0.05$) by the one-way ANOVA. (d) N and P effects on CH₄ uptake as normalised to the control treatment in semi-arid grassland from 2013 to 2016; error bars indicate the standard error (note that the N + P all have suppression impact but in a lesser degree).

The best fitting equation obtained with the stepwise regression procedure was as follows:

$$F_{\text{CH}_4} = m + a \times N + b \times P + c \times \ln(N) \times \ln(P) + d \times \text{ST} + e \times \text{pH} + f \times \text{SOC} + g \times \text{BD} + h \times \text{CL} \quad (2)$$

where F_{CH_4} is the annual CH₄ uptake rate; N is the nitrogen input in g ha⁻¹ year⁻¹; P is the phosphorus input rate in g ha⁻¹ year⁻¹; ln represents the natural logarithm; ST is soil temperature (K); pH is the soil pH value; SOC is soil organic carbon content (in %); BD is bulk density (g cm⁻³); CL is

clay content (in %); m is the intercept of the function and a, b, c, d, e, f, g and h are coefficients. The coefficients and key parameters for the regression are listed in Table S1. The model explained more than 37% of the variation in CH₄ uptake rate across the globe (Fig. S7).

We then applied the empirical model to estimate the CH₄ sink strength of global grasslands (Fig. S8), under four treatments: ambient (i.e. pre-industrial) levels of N and P depositions, elevated N deposition (contemporary N deposition and pre-industrial P deposition), elevated P deposition

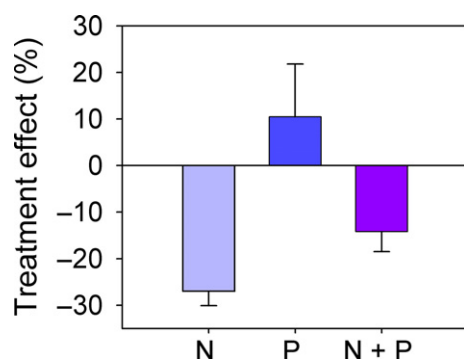


Figure 2 A meta-analysis of N and P effects on CH₄ uptake in grassland ecosystems (percentage is used to report the N and P impacts).

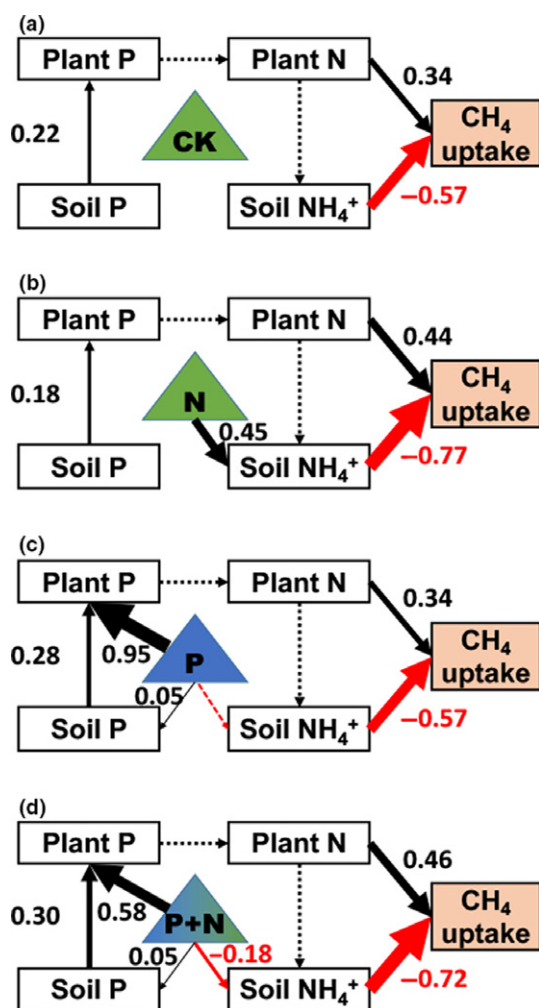


Figure 3 Structural equation modelling (SEM) for the relative controls of soil NH₄⁺ content, soil P content, plant N and plant P on CH₄ flux under various treatments; (a) ambient ($n = 96$, $\chi^2 = 1.389$, $DF = 0$, $CFI = 0.986$, $NFI = 1$), (b) N addition ($n = 96$, $\chi^2 = 5.671$, $DF = 0$, $CFI = 0.968$, $NFI = 1$), (c) P addition ($n = 96$, $\chi^2 = 1.644$, $DF = 0$, $CFI = 0.993$, $NFI = 1$) and (d) concomitant N + P additions ($n = 96$, $\chi^2 = 10.347$, $DF = 0$, $CFI = 0.943$, $NFI = 1$) (black lines represent positive paths; red lines represent negative paths; the solid lines represent the path is statistically significant [$P < 0.05$]; the dotted lines represent non-significant effects [$P > 0.05$]).

(contemporary P deposition and pre-industrial N deposition), and concurrently elevated N + P depositions (contemporary N and P depositions) (Fig. 4). Simulated global grassland CH₄ sinks amounted to 4.43 ± 0.20 Tg C-CH₄ year⁻¹ for the ambient scenario, 3.92 ± 0.16 Tg C-CH₄ year⁻¹ for the elevated N scenario, 4.60 ± 0.22 Tg C-CH₄ year⁻¹ for the elevated P scenario and 4.18 ± 0.18 Tg C-CH₄ year⁻¹ for the N + P scenario (Fig. 4). Addition of N only thus suppressed the global grassland CH₄ sink by ~ 0.50 Tg C ($\sim 11.4\%$), whereas concurrent P deposition alleviated this suppression to a c. 5.8% reduction in CH₄ sink.

Conceptual model for P alleviation of N-suppressed CH₄ sink

Based on our field experiments, the global meta-analysis, and the empirically derived insights of N and P impacts on CH₄ uptake, we here propose a conceptual framework that summarises the possible mechanisms underlying the interactive impacts of N and P additions on CH₄ uptake (Fig. 5). In this conceptual model, MMO represents the whole group of enzymes responsible for CH₄ oxidation under aerobic conditions (Dunfield & Knowles 1995) (Fig. 5a). Under ambient conditions (e.g. pre-industrial N and P deposition), grassland productivity is generally limited by low soil N availability (Ladwig *et al.* 2012) and by low soil P availability in more than half of the grasslands (Fay *et al.* 2015) and grassland plant species have therefore optimised their N uptake and allocation processes during ecological succession (Bai *et al.* 2004). Soil N does not leach under these low-N conditions and available N is either assimilated by plants or immobilised by soil microbes. Because soil mineral N (particularly NH₄⁺) is maintained at a low level, competition with CH₄ for the MMO enzyme is weak (Fig. 5a). In contrast, sustained or high N addition will push the system out of N limitation and result in NH₄⁺ accumulation in the soil (Fig. 5b). This in turn strengthens the competition with CH₄ for the MMO enzyme, thereby suppressing the oxidation of atmospheric CH₄ in grassland soils (Fig. 5b). If N and P are concurrently added, the added P stimulates vegetation growth and uptake of mineral N, especially NH₄⁺, and thus partially alleviates the N-induced suppression of CH₄ oxidation. (Fig. 5c). This theoretical framework emphasises the substrate competition theory when explaining the P alleviation of N-suppression on CH₄ uptake.

DISCUSSION

Results from both the field experiments and the global meta-analysis support the substrate competition theory as a predominant mechanism explaining P alleviation of N-suppression of CH₄ uptake in global grassland. A number of studies have concluded that soil NH₄⁺ content is a major driver of CH₄ uptake suppression in drylands (Dunfield & Knowles 1995; Gullledge & Schimel 1998). The toxic impact of nitrite on methanotrophic activity might be another reason for this suppression (Dunfield & Knowles 1995). In our field site, however, the toxic impact of nitrite on methanotrophic bacteria (Schnell & King 1994) most likely did not contribute to the inhibited CH₄ uptake because no nitrite was detected in

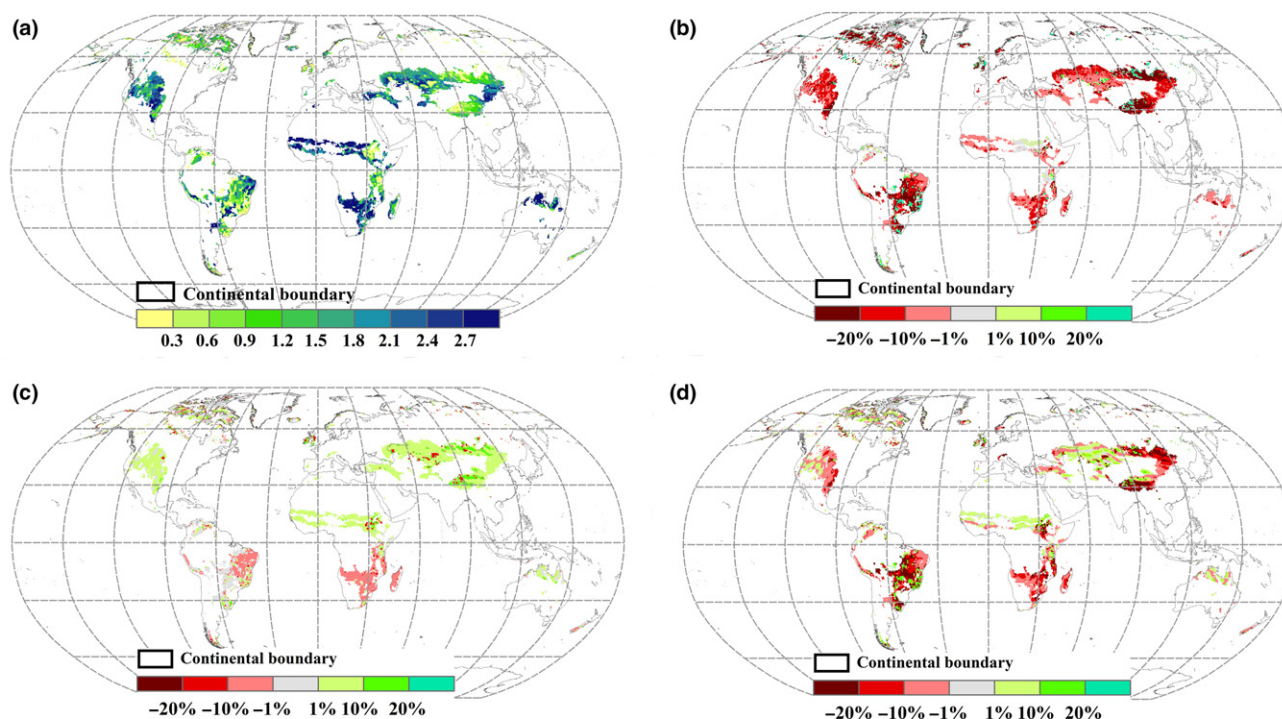


Figure 4 The model simulated contemporary (2005–2014) CH₄ oxidation and N and P impacts across global grasslands; (a) absolute CH₄ flux (Kg C ha⁻¹ yr⁻¹) under ambient condition (pre-industrial N + P deposition); (b) relative impact of N deposition (contemporary N deposition and pre-industrial P deposition) compared with the ambient condition; (c) relative impact of P deposition (pre-industrial N deposition and contemporary P deposition) compared with the ambient condition and (d) relative impact of N + P depositions (contemporary N + P depositions) compared with the ambient condition.

the soil, a widespread phenomenon in semi-arid grasslands (Holst *et al.* 2007; Giese *et al.* 2013; Zhang *et al.* 2017). Therefore, we conclude that substrate competition remains the best theory to explain the P alleviation of N-suppression of CH₄ oxidation.

A recent study in tropical forest postulated that P mitigation of the N-suppressed CH₄ oxidation might due to P stimulation of methanotrophic activities, although it further concluded it might be a minor contribution due to the trivial fraction of microbial biomass taken in by CH₄ oxidation bacteria (Zhang *et al.* 2011). This mechanism was postulated but not supported by empirical data (Zhang *et al.* 2011). Enhanced methanotrophic activity by P enrichment is thus one possible mechanism, yet it remains to be empirically confirmed as a predominant mechanism. More molecular and genomic analysis of the linkages between microbial mechanisms and ecosystem function remain needed to mechanistically elucidate and predict the CH₄ oxidation in a changing environment (Xu *et al.* 2016). Combined, the field experiment, meta-analysis, and global empirical modelling analysis unambiguously demonstrate that P-stimulated soil N depletion mitigate the N-induced reduction in CH₄ uptake.

Both the global meta-analysis and modelling results indicate a widespread impact of N and P on CH₄ uptake in grassland. Summarising all grids with treatment impacts compared with ambient grids, we found that N suppression of CH₄ uptake occurs in over 90% of global grasslands; the pervasive N suppression is consistent with reported N suppression of CH₄

uptake in many ecosystems (Mosier *et al.* 2003; Chen *et al.* 2013; Liu *et al.* 2017; Zhang *et al.* 2017). Model results suggest that P alleviation of N-suppression of CH₄ uptake occurs in over 89% of the global grasslands (Fig. 4). Considering the limited studies on P impacts on CH₄ uptake (Veraart *et al.* 2015), more field experiments on this aspect are needed. Estimates of the global grassland sink for CH₄ have been made in a large number of studies, yielding a broad range 1.9–9.3 Tg CH₄ yr⁻¹ (Potter *et al.* 1996; Ridgwell *et al.* 1999; Yu *et al.* 2017). Our result (5.9 Tg CH₄ year⁻¹) falls within the range of previous studies. According to our empirical model, increased N deposition reduced the grassland CH₄ sink by 11%, which was similar to another modelling study (10%) (Zhuang *et al.* 2013), but lower than the value reported in a data synthesis study (38%) (Liu & Greaver 2009).

The effects of N and P depositions on CH₄ uptake were quantified, yet a few issues should be paid attention to when interpreting the results. First, we quantified the potential CH₄ patterns under four N and P deposition scenarios, but did not consider the contributions of other changing environmental factors, such as increasing atmospheric CO₂ concentrations, and shifting land management practices, which have been shown to be important determinants of CH₄ flux (Xu *et al.* 2010). Second, our global-scale estimates of CH₄ uptake were based on a simple empirical model. Intensive data–model integration and model–model intercomparisons are still needed to better quantify the uncertainties in these CH₄ budgets. Third, the meta-analysis was carried based on the most

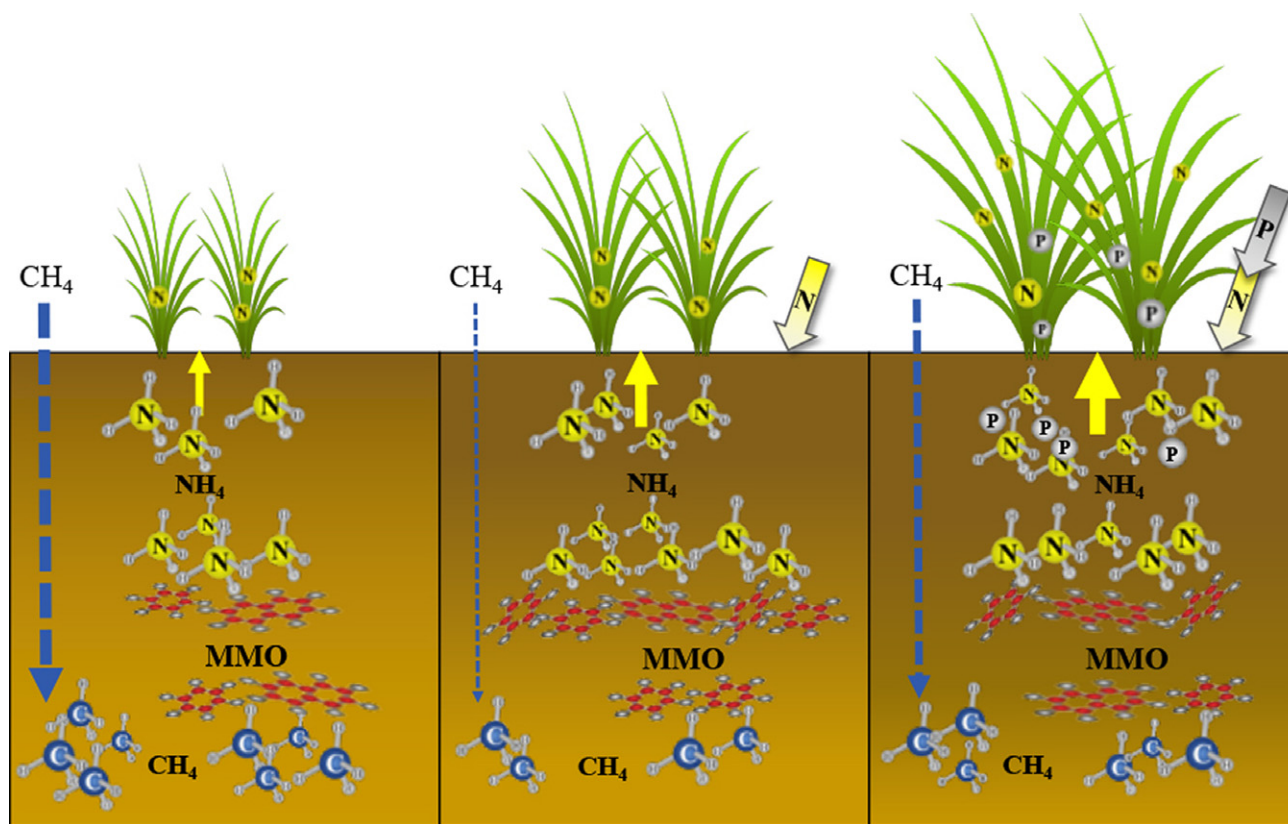


Figure 5 Graphic diagram showing substrate competition between CH_4 and NH_4^+ for methane mono-oxygenase (MMO); under ambient condition (left panel), N addition condition (middle panel) and concomitant N + P additions (right panel); the trivial soil NH_4^+ is in weak competitive equilibrium with CH_4 for MMO under ambient condition; and the added NH_4^+ shifts the competitive equilibrium and thus reduces the CH_4 oxidation; the P addition pushes the shifted equilibrium back and alleviates the N suppression on CH_4 oxidation by stimulating N cumulation in vegetation biomass.

comprehensive dataset for the impacts of N and P additions on CH_4 flux in global grassland; however, the uneven distribution of the field observational dataset might cause biases in the global analysis of nutrient impacts. This unequal spatial distribution of field observations exists for eddy covariance towers (Baldocchi *et al.* 2001), vegetation data (Kattge *et al.* 2011) and soil data (Xu *et al.* 2013). This problem needs to be resolved for reaching a more accurate global extrapolation and increasing our understanding of the functioning of terrestrial ecosystems. Deposition and addition of N and P take place in many forms, including inorganic and organic, dry and wet deposition. Due to limited data availability (Mahowald *et al.* 2008) and field experiments (Wang *et al.* 2015), the impacts of various N and P forms in atmospheric depositions could not be fully investigated in this study. Understanding the impacts of different N and P forms on CH_4 oxidation still remains to be fully investigated.

CONCLUSION

With a combination of a field experiment, a meta-analysis and an empirical modelling approach, we found that the substrate competition theory is most likely the predominant mechanism explaining the P alleviation of the N-suppression of CH_4 uptake. We further estimated that contemporary atmospheric N deposition suppresses the CH_4 sink by 11.4%, while P

deposition alleviates this suppression and leads to a 5.8% reduction in CH_4 sink in global grasslands.

This study is among the first attempts to quantify the P impacts on the N suppression of soil oxidation of atmospheric CH_4 in global grasslands. It advocates model development to incorporate P mechanisms not only for the nutrient impacts on carbon cycling but also for a more accurate projection of the CH_4 budget of terrestrial ecosystems.

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COMPETING INTERESTS

The authors declare no competing interests.

AUTHOR CONTRIBUTIONS

L.Z., J.B. and X.X. conceived the project. L.Z., X.G., L.H., L. Y. and Z. Z. performed the field experiments. J.H., Z. Z. and C.S. contributed datasets. L.Z., F.Y. and X.X. developed the empirical model and carried out the model simulation. L.Z., F.Y., D.A.L. and X.X., interpreted the results. L.Z., J.B. and X.X. wrote the manuscript with assistance from other co-authors.

DATA AVAILABILITY STATEMENT

The majority of the data used for analysis in this study are clearly documented in the materials and method section or in the supplementary information. All data used for analysis are archived at Dryad: <https://doi.org/10.5061/dryad.vt4b8gtms>

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