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Phosphorus availability as a primary constraint on methane emission from a freshwater wetland

Changchun Song, Guisheng Yang, Deyan Liu, Rong Mao*

Northeast Institute of Geography and Agroecology, Chinese Academy of Sciences, 3159 Weishan Rd, Changchun 130012, China

HIGHLIGHTS

- ▶ The effect of P enrichment on CH₄ emission is unclear in wetlands.
- ▶ 4-year P addition decreased CH₄ emissions in a temperate freshwater marsh.
- ▶ P addition's effect on CH₄ emissions was time-dependent.
- ▶ P addition's effect on CH₄ emissions did not vary with fertilization rates.
- ► Elevated P loading would suppress CH₄ emissions in freshwater wetlands.

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ABSTRACT

Human activities have increased phosphorus (P) loading in wetland ecosystems worldwide. However, little is known about the effect of P enrichment on CH₄ emissions from these ecosystems. In this study, a 4-year P addition experiment was conducted to examine the effects of increased P availability on CH₄ emission in a *Deyeuxia angustifolia*-dominated freshwater marsh in the Sanjiang Plain in northeastern China. Phosphorus was added at four rates (0, 1.2, 4.8 and 9.6 g P m⁻² year⁻¹). We investigated CH₄ emission during the growing season (early May to late September) using opaque chamber and gas chromatography method. Our results indicated that the effect of P enrichment on CH₄ emission was time-dependent. Increased P availability did not affect CH₄ emission in 2007 and 2008, but decreased in 2009 and 2010. Notably, four years of P addition decreased cumulative CH₄ emission during the growing season in the freshwater marsh, and the effect did not change with fertilization rates. From 2007 to 2010, P additions of 1.2, 4.8 and 9.6 g P m⁻² year⁻¹ caused a decline in growing-season CH₄ emissions by averages of 23%, 38% and 26%, respectively. Our results suggest that long-term P enrichment driven by agricultural activities would reduce CH₄ emission from temperate freshwater wetlands. This study also highlights the impact of experimental duration on accurate assessments of the effect of P addition on wetland CH₄ budget.

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1. Introduction

Methane (CH₄) is the second-most important anthropogenic greenhouse gas in the atmosphere after carbon dioxide (CO₂) and has 25 times the global-warming potential of CO₂ over a 100-year horizon (IPCC, 2007). Global atmospheric CH₄ concentration has increased from 700 ppb in pre-industrial times to 1782 ppb in 2006 (Wuebbles and Hayhoe, 2002; Fowler et al., 2009). Wetlands, excluding non-rice agriculture, are the largest natural source of CH₄ to the atmosphere, and they contribute approximately 20% of the global CH₄ budget (IPCC, 2007; Fowler et al., 2009). Of this

contribution, one-third is derived from temperate and boreal northern wetlands (Gauci et al., 2004). In wetland ecosystems, CH₄ emission generally exhibits drastic temporal and spatial variations (Fowler et al., 2009; Song et al., 2009) and varies with water table level (Christensen et al., 2003; O'Connor et al., 2010), temperature (Christensen et al., 2003; Sun et al., 2011), substrate C availability (Bubier and Moore, 1994; O'Connor et al., 2010), pH (Dunfield et al., 1993), redox state (Bubier and Moore, 1994), nutrient availability (Keller et al., 2006) and vegetation community and production (Whiting and Chanton, 1993).

Phosphorus (P) is one of the most important elements limiting plant primary production and other ecosystem processes in most terrestrial and aquatic ecosystems (Elser et al., 2007; Filippelli, 2008). In recent decades, anthropogenic P enrichment in most ecosystems worldwide has been driven by P mining and P transportation in

^{*} Corresponding author. Tel.: +86 431 85542204; fax: +86 431 85542298. E-mail address: maorong@neigae.ac.cn (R. Mao).

fertilizers, animal feed, agricultural crops and other products (Bennett et al., 2001; Filippelli, 2008). Similar to other natural ecosystems, wetlands are currently experiencing increased P loading as a result of human activities (Keller et al., 2005; Song et al., 2011). In wetland ecosystems, P enrichment is assumed to stimulate plant growth and increase plant production, enhancing substrate availability for methanogens through root exudates and litter turnover, which increases CH₄ fluxes to the atmosphere. However, previous studies have found that the response of plant productivity to P addition is often species-specific and that this response varies by wetland type (Verhoeven and Schmitz, 1991; Chapin et al., 2004; Keller et al., 2006). Moreover, increased P loading to wetlands may change soil microbial community and activity (Keller et al., 2006) and alter plant-mediated transport of CH₄ (Lu et al., 1999). Given that emission of CH₄ from the soil to the atmosphere is dependent on the balance among CH₄ production, transport and oxidation, the effect of P enrichment on CH₄ emission is still uncertain in wetlands. An improved understanding of the response of CH₄ flux rates to P enrichment in specific wetland ecosystems is essential for accurately assessing the global CH₄ budget from wetlands and projecting the future climate through global climate models.

The Sanjiang Plain, located east of Heilongjiang Province in northeastern China, is an alluvial floodplain that includes one of the largest freshwater wetland areas in China (Zhao, 1999). In the past 50 years, marshes have been extensively drained and used for agriculture in the Sanjiang Plain (Yan et al., 2001). Therefore, the formerly pristine wetlands in this region generally have been subjected to increased P loading, mainly due to fertilization application in the adjacent agricultural lands. Will increased P input alter CH₄ flux rates in the wetland ecosystems in the Sanjiang Plain? To answer this question, we established a P fertilization experiment in 2007 in a *Deyeuxia angustifolia*-dominated freshwater marsh in the Sanjiang Plain of northeastern China. We subsequently collected one 4-year (from 2007 to 2010) continuous observational dataset to assess the effect of P enrichment on wetland CH₄ emission during the growing season (early May to late September).

2. Material and methods

2.1. Site description and experimental design

The study was conducted in a D. angustifolia-dominated freshwater marsh located near the Sanjiang Mire Wetland Experimental Station (47°35′N, 133°31′E, 56 m a.s.l) in the Sanjiang Plain, Heilongjiang Province, northeastern China. D. angustifolia-dominated freshwater marsh is one of the main wetland types in the Sanjiang Plain, as this ecosystem type accounts for approximately 31% of the wetland area in this region (Zhao, 1999). The study site has a temperate continental monsoon climate with a mean annual (1990-2010) temperature of 2.53 °C (month range -20.4 to 21.6 °C), a mean annual precipitation of 566 mm (approximately 50% fall in July and August) and a frost-free period of approximately 125 days per year. The soil in the study site is a typical meadow marsh soil and is classified as an Inceptisol in the US soil taxonomy classification system. Mean soil organic C at 0-15 cm depth is 95 mg g⁻¹, total N concentration is 8.5 mg g⁻¹, bulk density is 0.67 g cm⁻³ and pH is 5.35. The dominant plant species at this site are D. angustifolia and Glyceria spiculosa, which comprise more than 90% of the total aboveground biomass. Detailed information regarding the study site was described by Song et al. (2009).

We used a complete randomized block design with four P addition treatments (control, 0 g P m $^{-2}$ year $^{-1}$; P1, 1.2 g P m $^{-2}$ year $^{-1}$; P2, 4.8 g P m $^{-2}$ year $^{-1}$; P3, 9.6 g P m $^{-2}$ year $^{-1}$) that was replicated three times, resulting in 12 plots of 1 m \times 1 m. During autumn 2006, plastic (PVC) frames (1 m \times 1 m, 0.5 m in depth) were installed to

prevent horizontal movement and lateral loss of the added P, and each plot was separated by a 1-m buffer zone. Meanwhile, board walks giving access to the whole experimental area were established to minimize further disturbance on the plots. Phosphorus was added as NaH₂PO₄. Phosphorus fertilizer was dissolved in 1 L distilled water and applied at 2-week intervals from early May to late August. At the same time, the control plots were treated with the same volume of distilled water. We chose a P amendment of 1.2 g m⁻² year⁻¹ because it was comparable to the amount of the background increased P input in wetlands during the growing season in this region. To examine the probable increases in P input to these wetlands in the future, we experimentally increased the ambient growing season P loading rate by factors of 4 and 8.

2.2. CH₄ flux measurement

CH₄ emission was measured at weekly to biweekly intervals using an opaque static chamber (constructed from stainless steel, $0.5 \text{ m} \times 0.5 \text{ m} \times 0.5 \text{ m}$) during the growing seasons of 2007, 2008, 2009 and 2010. Before sampling, the chambers were placed into collars (also made from stainless steel) that were permanently installed in the soil. Inside the chamber, two fans were used to keep the air homogenized during gas sampling, and a thermometer sensor and a trinal-venthole were installed. Gas sampling was usually carried out at 9:00 AM local time and lasted half an hour; four gas samples were collected using polypropylene syringes at 10-min intervals. Meanwhile, air temperatures inside and outside the chamber were recorded using a thermometer sensor, and the surface water table was measured using a ruler near the chamber in each plot. When the water level was below the soil surface, the water table was determined by digging a small well near the chamber in each plot. The gas samples were stored in syringes less than 12 h before being measured, and CH₄ concentration was analyzed by a gas chromatogram (Agilent 4890D, Agilent Co., Santa Clara, CA, USA) using flame ionization detection. CH₄ emissions were calculated from the linear changes in chamber gas concentration with an average chamber temperature and were expressed as mg C m⁻² h⁻¹. Sample sets were rejected when the R^2 of the linear regression of CH₄ concentration over time was lower than 0.9. Total annual CH₄ emissions were linearly and sequentially calculated from the emissions between each pair of adjacent measurement intervals during the growing season and expressed as g C m⁻² year⁻¹. Detailed information about gas sampling using an opaque static chamber was described by Song et al. (2009).

2.3. Statistical analyses

Data were tested for normality using the Levene's test, and all non-normal data were transformed prior to analysis. Repeated measures analysis of variance tests (ANOVAs) were used to examine inter-annual variability in the CH₄ budget during the growing season in combination with P addition treatments. If there were significant interactions between time and P addition levels on annual CH₄ budget, we conduct further tests for significant differences in CH₄ budget among all treatments using a Fisher's least significant difference test. For each year, we tested the effect of P addition level on seasonal CH₄ fluxes using repeated measures ANOVAs. All statistical analyses were conducted with SPSS 11.5 for Windows, and the accepted significance level was $\alpha = 0.05$.

3. Results

Precipitation in the growing season (May—October) changed substantially over the 4-year study interval with 529 mm in 2007, 372 mm in 2008, 529 mm in 2009 and 504 mm in 2010. Mean air

temperatures were 18.0, 17.7, 17.2 and 18.6 °C in 2007, 2008, 2009 and 2010, respectively (Fig. 1).

Temporal dynamics of CH_4 emissions in the freshwater marsh followed a one-peak pattern in 2007 and 2009 and a two-peak pattern in 2008 and 2010 (Fig. 2). During the growing season, peak CH_4 emissions occurred in June in 2007 and 2008, but in August in 2009 and 2010 (Fig. 2).

During 4 years of growing seasons, the P1, P2 and P3 treatments decreased CH₄ emissions by 23%, 38% and 26%, respectively (all P < 0.05, Fig. 3). However, there was a significant interaction between P addition level and year on CH₄ emission (P < 0.001, data not shown). Over the growing season, P addition had no effect on CH₄ emissions in 2007 and 2008, but decreased in 2009 and 2010 (Fig. 3). However, there was no significant difference in CH₄ emissions among the P1, P2 and P3 treatments (Fig. 3).

4. Discussions

Notably, four years of P addition decreased CH₄ emissions during the growing season in the freshwater marsh in the Sanjiang Plain in northeastern China (Fig. 3). Lu et al. (1999) also found that increased P availability caused a decline in CH₄ emission in paddy soils. Moreover, Zhang et al. (2011) observed that three years of P addition increased CH₄ uptake in an old-growth tropical forest in southern China. However, previous studies found that P addition caused a short-term increase in CH₄ emissions from peat soils (Aerts and Toet, 1997) or paddy soils (Han et al., 2002). In addition, Keller et al. (2005) found that six years of P fertilization had

a limited effect on rates of CH₄ production and consumption and thus CH₄ emission in a Minnesota fen. Lund et al. (2009) also found that a two-year P fertilization experiment had no effect on CH₄ exchange in two northern peatlands with contrasting N deposition rates. The inconsistent impacts of P enrichment on CH₄ emission may be explained by differences in experimental duration and ecosystem type (Keller et al., 2006). Indeed, we also found that the impact of P addition on CH₄ emission varied with experimental duration, although the cumulative relationship was negative (Fig. 3). Therefore, long-term field experiments would need to be conducted to examine temporal changes in the P addition effect on CH₄ emission and accurately predict the response of CH₄ emissions to P enrichment in wetland ecosystems. However, our results to date suggest that increased P loading caused by agricultural activities constrains CH₄ emissions in temperate freshwater wetlands.

In the present study, we inferred that the decrease in CH₄ emission under P addition treatments may be driven by several related mechanisms. First, increased P availability may reduce root biomass and exudation, reducing the substrate for methanogenesis (Lu et al., 1999; Mer and Roger, 2001). In the adjacent freshwater marshes, Song et al. (2011) found that P fertilization did not affect plant aboveground biomass. Considering that P addition generally increased the plant shoot/root ratio (Macek and Rejmánková, 2007), we speculated that increased P availability would decrease root biomass in this freshwater marsh. Second, P enrichment may inhibit methanogenesis in soils (Conrad et al., 2000; Keller et al., 2006) and increase soil methanotrophic potential (Joulian et al., 1998). Third, P fertilization may decrease root porosity and inhibit

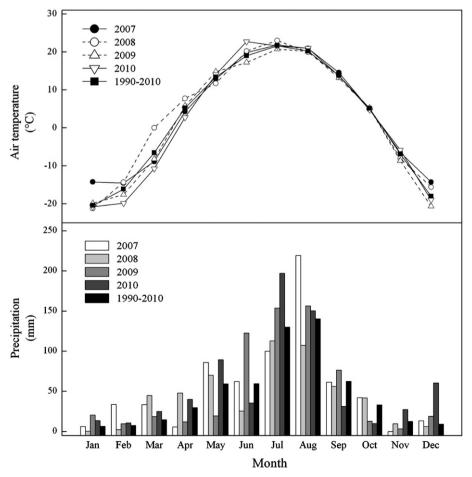


Fig. 1. Temporal changes in monthly mean air temperature (lines) and precipitation (bars) from 2007 to 2010 in the study site.

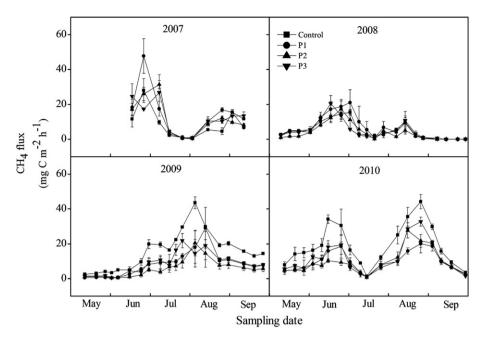


Fig. 2. Temporal dynamics of the CH₄ flux in the control and phosphorus addition treatments during the growing seasons from 2007 to 2010. Error bars represent \pm SE (n=3). Control, 0 g P m⁻² year⁻¹; P1, 1.2 g P m⁻² year⁻¹; P2, 4.8 g P m⁻² year⁻¹; P3, 9.6 g P m⁻² year⁻¹.

the development of root aerenchyma (Lu et al., 1999). In addition, changes in plant community structure induced by nutrient addition may alter soil carbon quality and potentially influence CH₄ flux from wetlands (Keller et al., 2006).

Interestingly, the amount of P added had no additive effect on CH_4 suppression in the freshwater wetland, and adding P with different rates had the same negative effect on CH_4 emission (Fig. 3). In a previous study conducted in an adjacent freshwater marsh, Song et al. (2011) observed that aboveground biomass did not change in response to different levels of P fertilization, suggesting that P may not limit plant growth in these wetlands. These findings revealed that there was a threshold for P-induced effects on aboveground biomass and CH_4 emission, and the critical value for CH_4 emission was below 1.2 g P m⁻² year⁻¹ in this freshwater

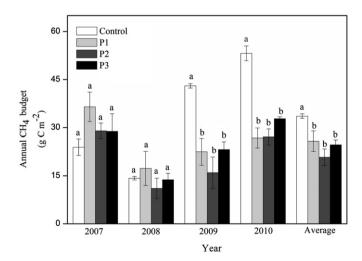


Fig. 3. CH₄ budgets during the growing season from 2007 to 2010 in the control and phosphorus addition treatments. Error bars represent \pm 5E (n=3). Means with different lowercase letters in the same column are significantly different at P<0.05. Control, 0 g P m⁻² year⁻¹; P1, 1.2 g P m⁻² year⁻¹; P2, 4.8 g P m⁻² year⁻¹; P3, 9.6 g P m⁻² year⁻¹.

wetland. This finding implies that elevated P loading would exert a consistent suppression of CH₄ emission in temperate freshwater wetlands.

The mean CH₄ budget in the control plots averaged 33.6 g C m⁻² during the growing season and ranged from 14.3 g C m $^{-2}$ in 2008 to 53.2 g C m $^{-2}$ in 2010 (Fig. 3). The mean CH₄ budget in this freshwater marsh was comparable to that in an Carex lasiocarpa-dominated adjacent freshwater $(36.6 \text{ g C m}^{-2} \text{ from } 2002 \text{ to } 2005, \text{ Song et al., } 2009) \text{ but was greater}$ than the value observed in the adjacent D. angustifolia-dominated freshwater marsh (3.15 g C m⁻² from 2002 to 2005, Song et al., 2009) and lower than the CH₄ budget in a Carex schmidtii and D. angustifolia-dominated freshwater marsh in the Xiaoxing'an Mountains of northeastern China (90 g C m⁻² from May to October in 2008, Sun et al., 2011). The magnitudes of our results during the growing season were similar to the previous studies performed in temperate wetlands in North America (Bubier et al., 1993; Treat et al., 2007). Moreover, CH₄ emissions exhibited large seasonal and inter-annual variations over the study period (Figs. 2 and 3) and were positively correlated with air temperature and the height of the water table (all P < 0.001, data not shown). These results imply that air temperature and the water table are important factors controlling CH₄ emissions in the study site and also indicate that CH₄ emissions in temperate freshwater wetlands are sensitive to climate change.

Although our study focused on the effect of P addition on CH₄ emission, the natural wetlands in this region of China also received elevated N input, due to fertilization application in the adjacent agricultural lands. Keller et al. (2006) found that N and P fertilization acted independently on CH₄ production and oxidation in peatlands. However, Zhang et al. (2011) observed that increased P availability mitigated the inhibition of N enrichment on CH₄ uptake in an old-growth tropical forest. Therefore, how increased N availability interacted with increased P availability to affect CH₄ emissions is still unclear, and therefore the net effect of nutrient enrichment on CH₄ emission remains uncertain in this ecosystem. Further study is needed to assess the combined effect of N and P additions on CH₄ emission in temperate freshwater wetlands.

5. Conclusions

In the present study, we have assessed the effect of P enrichment on CH₄ emission in a *D. angustifolia*-dominated freshwater marsh in the Sanjiang Plain in northeastern China. Notably, four years of P addition suppressed CH₄ emissions in this wetland, and the negative effect did not vary with fertilization rates. These results suggest that long-term increased P loading from agricultural activities reduces the emission of CH₄ from temperate freshwater wetlands. Given widespread eutrophication by anthropogenic P input for temperate freshwater wetlands, our results will help to develop an accurate C budget for wetland ecosystems.

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