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Methane uptake in forest soils along an urban-to-rural gradient in Pearl River Delta, South China

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We investigated soil CH₄ fluxes from six forests along an urban-to-rural gradient in Guangzhou City metropolitan area, South China. The most significant CH₄ consumption was found in the rural site, followed by suburban, and then urban forest sites. The rates of CH₄ uptake were significantly higher (by 38% and 44%, respectively for mixed forest and broadleaf forest) in the rural than in the urban forest site. The results indicate that soil water filled pore space (WFPS) is the primary factor for controlling CH₄ consumption in subtropical forests. The reductions of soil CH₄ uptake in urban forests were also influenced by the higher rates of atmospheric nitrogen (N) deposition and increases in soil nitrate (NO₃⁻) and aluminum (Al³⁺) contents as a result of urbanization. Results from this work suggest that environmental changes associated with urbanization could decrease soil CH₄ consumption in subtropical forests and potentially contribute to increase of atmospheric CH₄ concentration.

Methane contributes up to 30% to total net anthropogenic radiative forcing of the atmosphere¹. Atmospheric CH₄ has increased 151% since 1750 as a result of an imbalance between increasing sources and decreasing sinks². The largest biological sink of CH₄ is consumption by soil aerobic bacteria³. Upland soils (e.g., forest, grassland, and desert) have been shown to be significant sinks for CH₄⁴, contributing approximately 6% of the total consumption⁵. Among upland soils, forest is probably the most efficient CH₄ sink⁴. Urbanization has increased rapidly worldwide and 70% of the world population will be urban by 2050⁶. The urban population of China rose from 18% in 1978 to 45% in 2005, and is projected to be 60% by 2030⁷. Urbanization is generally accompanied with environmental changes in chemistry and climate, which consequently impact the capacity of CH₄ consumption in urban forests^{8,9}.

Previous studies showed that CH₄ uptake was commonly lower in urban than in suburban and rural forests^{8,10,11}. However, there was disagreement regarding the mechanisms underlying the reductions of CH₄ uptake. Goldman et al. ascribed the suppressed CH₄ uptake to lower rates of organic matter degradation and nutrient cycling caused by air pollution (especially as O₃)⁸. Groffman and Pouyat reported that increases in N deposition and CO₂ levels might be the possible mechanisms¹⁰. Costa and Groffman found that the differences in N cycling associated with urbanization led to a reduction in the microbial populations responsible for CH₄ uptake¹². So far, the effects of urbanization on CH₄ uptake in tropical and subtropical forest soils remain unclear.

The objective of this study was to investigate how CH₄ fluxes from subtropical forests varied along an urban-to-rural gradient in Guangzhou City metropolitan area, and to determine what factors underlie urbanization effects on CH₄ uptake. We hypothesized that CH₄ consumption would be lowest in urban forests, followed by the suburban and rural forest sites, due to the exposure to urbanization-induced environmental changes of the former.

Results

Soil CH₄ uptake. Heishiding (HSD, rural site) forests had the highest rates of CH₄ uptake, followed by Dinghushan (DHS, suburban site) and then Maofengshan (MFS, urban site) forests (Fig. 1 a, b, all *P* < 0.05). For pine and broadleaf mixed forests (MF), the mean rate of CH₄ uptake in HSD was higher by 38% than in MFS

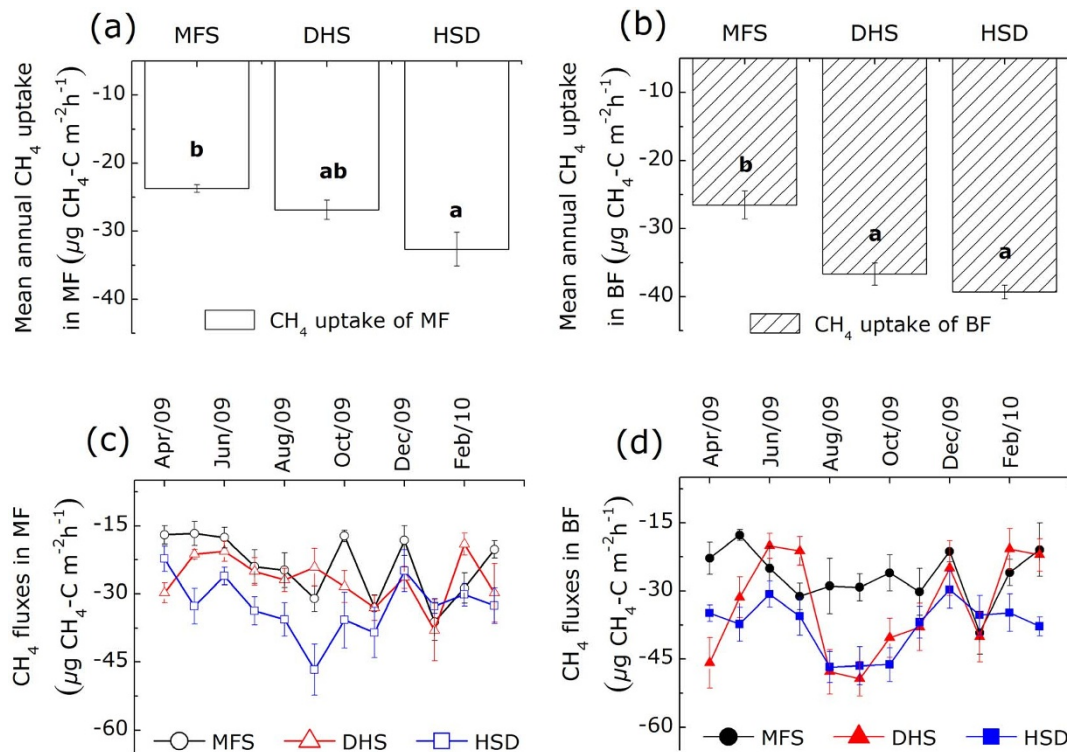


Figure 1 | Comparisons of the mean rates and seasonal patterns of CH₄ uptake. (a) and (b) depict the mean rates of CH₄ uptake, and (c) and (d) show the seasonal patterns of soil CH₄ uptake (Mean value \pm 1 standard error, $n = 5$); Different letter “a” and “b” denote significant differences ($P < 0.05$) for the same forest type. MF, mixed forest; BF, broadleaf forest; MFS, Maofengshan; DHS, Dinghushan; HSD, Heishiding.

site (Fig. 1 a). In broadleaf forests (BF), the average rate of CH₄ uptake was higher by 37% in DHS, and 44% in HSD than that of MFS site (Fig. 1 b, all $P < 0.05$). In DHS forest site, average CH₄ uptake for BF was 37% higher than that of MF ($P = 0.04$). However, there were no differences between MF and BF stands at other forest sites. Monthly mean CH₄ uptake in all forests showed a similar seasonal pattern, with the highest consumption occurring in the fall (August to October) (Fig. 1 c, d).

Biotic and abiotic variables. The amount of N deposition in rainfall was higher in MFS and DHS than in HSD site ($P < 0.05$), with no significant difference between MFS and DHS (Table 1). Soil temperature and WFPS exhibited clear seasonal patterns (Fig. 2 a, b), similar to those of air temperature and rainfall at each site (Fig. S1). Higher soil WFPS and temperatures were found in MFS forests than that of HSD site (Fig. 2 c, d, all $P < 0.05$).

For both forest types, soil NO₃⁻ contents were higher in HSD than that of MFS and DHS sites, whereas soil NH₄⁺ and total N (TN) showed an opposite pattern (Table 2). Soil bulk density and pH values were significantly lower in MFS and DHS than in HSD sites, conversely, soil Al³⁺ contents were higher in MFS and DHS sites (Table 2). Soil CH₄ uptake showed negatively related with soil NO₃⁻ and Al³⁺ contents (Fig. 3 a, c), and positively related to pH values (Fig. 3 b). Soil microbial biomass C (MBC) contents were lowest in MFS, followed by DHS and HSD for the same forest stands, whereas microbial biomass N (MBN) had no significant differences across the gradient sites (Table 2).

Discussion

Methane fluxes at all sites were predominantly negative during the study period, indicating CH₄ uptake from the atmosphere. While many studies have quantified differences in CH₄ cycling under the

Table 1 | Site characteristics along an urban-to-rural gradient in southern China

Characteristic	Site		
	MFS (urban)	DHS (suburban)	HSD (rural)
Site code			
Latitude and longitude	23°18'5.87"N; 113°27'0.57"E	23°8'57.27"N; 112°31'3.07"E	23°27'42.85"N; 111°54'19.78"E
Distance from urban core (km)	36.7	97.8	169.3
Altitude (m)	150	225	395
Mean slop (°)	21.2	24.5	27.3
Annual precipitation (mm)	1742	1625	1690
Average annual temperature (°C)	22.1 (0.5)	22.2 (0.4)	21.0 (0.5)
N deposition in rainfall (kg N ha ⁻¹ yr ⁻¹)	32.4 (1.1) a	40.2 (1.3) a	18.6 (0.7) b
NH ₄ ⁺ :NO ₃ ⁻ ratio	0.47 b	0.61 b	1.23 a
Soil type	Lateritic red earth	Lateritic red earth	Lateritic red earth

Data of temperature and precipitation were collected from nearby meteorological stations (from April 2009 to March 2010). Values of average annual temperature and N deposition in rainfall are presented as means with SE in parentheses ($n = 12$ and 5 , respectively for temperature and N deposition). Different letter within a row presented significantly different (one-way ANOVA, $P < 0.05$). NH₄⁺:NO₃⁻ ratio, the ratio of NH₄⁺:NO₃⁻ in rainfall; MFS, Maofengshan; DHS, Dinghushan; HSD, Heishiding.

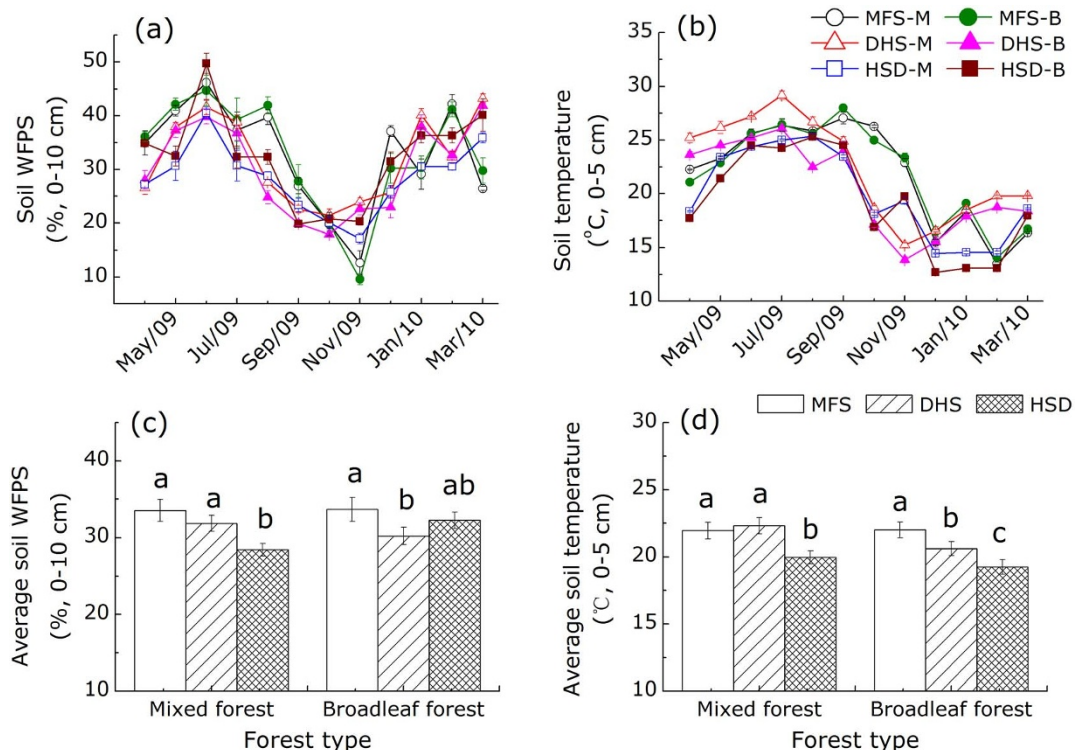


Figure 2 | Soil temperature and WFPS at forests along the urban-to-rural gradient. (a) Seasonal patterns of soil WFPS at 0–10 cm depth; (b) seasonal patterns of soil temperature at 5 cm depth; (c) annual mean soil WFPS; and (d) annual mean soil temperature (Mean value \pm 1 standard error, $n = 5$). In panel (a) and (b), letters M and B following the site abbreviations denote mixed forest and evergreen broadleaf forest, respectively. Different letter “a” and “b” denotes significant difference between forests for the same forest type ($P < 0.05$). MFS, Maofengshan; DHS, Dinghushan; HSD, Heishiding.

changes of land uses¹¹, there is considerable uncertainty about the unmanaged or intact forests within urban areas acting as CH_4 sinks that have been observed^{8,10,12}. The rates of CH_4 uptake in the suburban and rural forests (2.4 to $3.3 \text{ kg CH}_4\text{-C ha}^{-1} \text{ yr}^{-1}$) were comparable with previous studies in tropical and subtropical regions of southern China (2.5 to $4.3 \text{ kg CH}_4\text{-C ha}^{-1} \text{ yr}^{-1}$)^{13–15}. In the suburban (DHS) site, the soil of BF oxidized more CH_4 than that of MF stand, a trend observed in previous studies^{13,16}. The BF is an old-growth forest (more than 400 years), which represents a forest type in an advanced successional stage within the study region¹⁷. Forest composition, species assemblages and soil density (Table S1) are more favorable than MF, leading to increased CH_4 consumption in this BF stand^{4,13,16}.

Consistent with our previous hypothesis, the urban forest soils had the lowest capacity of CH_4 consumption compared to rural and suburban sites. Our results were comparable with previous reports

that CH_4 uptake would be markedly lower in urban than in rural forest soils. For example, Goldman et al. found that CH_4 uptake rates in intact forests in urban center of New York City (NY, USA) were 30% lower than that of rural forest soils⁸. Similar results were found in urban forests in the Baltimore metropolitan area (MD, USA)^{10,11}. These results have suggested that there is an urban atmospheric effect on CH_4 consumption in urban forests¹². In the present study, the reduction of CH_4 uptake in urban forests might be influenced by several factors as follows.

Firstly, the reduction of CH_4 uptake was primarily influenced by the lower diffusion of CH_4 into urban forest soils. Multiple regression analyses indicated that soil WFPS accounted for 52% of the variance in CH_4 uptake for both MF and BF stands (Table 3). Higher soil WFPS and bulk density in the urban forests could increase resistance to atmospheric CH_4 and O_2 transport into the soils, reducing the

Table 2 | Soil properties (0–10 cm depth) of forests along the urban-to-rural gradient

Forest type	MF				Significance	BF			
	MFS	DHS	HSD	Significance		MFS	DHS	HSD	Significance
NH_4^+ (mg kg^{-1})	6.4(0.5) b	8.5(1.6) b	17.2(1.9) a	$P < 0.01$	9.0(1.1) b	8.8(1.0) b	16.7(2.2) a	$P < 0.01$	
NO_3^- (mg kg^{-1})	17.5(1.4) a	21.7(1.5) a	8.1(1.3) b	$P = 0.01$	23.1(2.3) a	22.5(2.3) a	11.7(1.4) b	$P < 0.01$	
Total N (g kg^{-1})	1.7(0.1) b	2.2(0.1) a	2.4(0.2) a	$P = 0.01$	2.2(0.1) b	2.7(0.2) b	3.2(0.1) a	$P < 0.01$	
MBC (mg kg^{-1})	189(25) b	278(30) ab	294(15) a	$P = 0.02$	389(18) b	515(37) a	509(38) a	$P = 0.05$	
MBN (mg kg^{-1})	31.2(4)	30.8(4)	22.7(2)	$P > 0.05$	44.2(5)	45.7(7)	37.4(4)	$P > 0.05$	
pH	3.87(0.01)b	3.97(0.07) ab	4.14(0.04) a	$P < 0.01$	3.86(0.08) b	3.73(0.03) b	4.22(0.03) a	$P = 0.01$	
Al^{3+} (mmol kg^{-1})	85(2) a	82(2) a	59(1) b	$P = 0.04$	101(4) a	102(3) a	69(1) b	$P = 0.03$	
Bulk density (g cm^{-3})	1.2(0.1) a	1.2(0.1) a	1.0(0.2) b	$P = 0.02$	1.2(0.1) a	1.1(0.0) ab	1.0(0.2) b	$P = 0.01$	

Soil samples were collected in July 2009. Values are presented as mean with SE in parentheses ($n = 5$). Different letters within the same row from the same forest type denote significant difference between the three sites [one-way ANOVA with Tukey’s HSD, $P < 0.05$]. MBC, microbial biomass C; MBN, microbial biomass N; MF, mixed forest; BF, broadleaf forest; MFS, Maofengshan; DHS, Dinghushan; HSD, Heishiding.

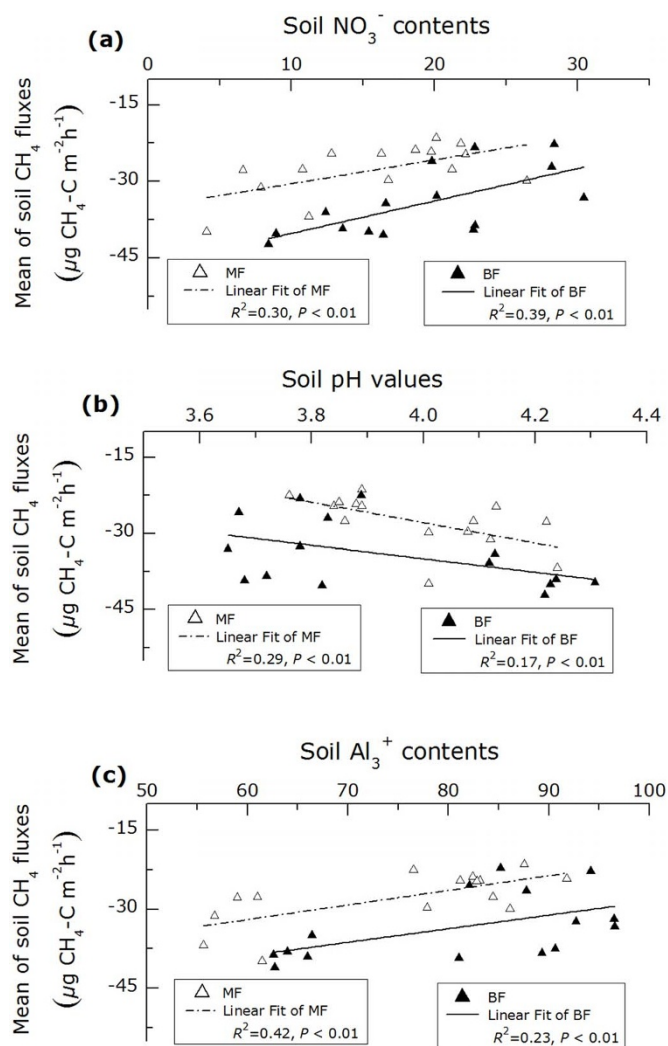


Figure 3 | Linear regressions between annual CH₄ uptake rates and soil properties. (a) soil NO₃⁻ contents and CH₄ fluxes, (b) soil pH values and CH₄ fluxes, and (c) soil Al³⁺ contents and CH₄ fluxes. CH₄ fluxes used in this figure were measured in field at the same day of soil sampling (July 2009). MF, mixed forest; BF, evergreen broadleaf forest.

activity of methanotrophic bacteria^{4,18}. The seasonal changes in CH₄ uptake were also best explained by gas diffusion, and were most related with lower soil WFPS during the fall. There was no significant relationship between CH₄ uptake and soil temperature, which was consistent with previous studies in this region^{13,15,19}. These results suggest that soil temperature is not the key factor for controlling CH₄ consumption in subtropical forests of southern China.

Secondly, higher N cycling rates and soil NO₃⁻ contents, which are caused by high rates of atmospheric N deposition, significantly decreased soil CH₄ uptake in the urban forests. Soil NO₃⁻ contents accounted for 30% of the variance in CH₄ uptake (Table 3). Previous studies have shown that NO₃⁻ and/or NO₂⁻ produced from the NO₃⁻ reduction were possibly toxic to CH₄-oxidizing microbes^{20,21}, which might be a potential role for the reduction of CH₄ consumption in our urban forests. Although we did not measure net N mineralization and nitrification rates, the fact that these results were consistent with patterns of N deposition and soil N cycling already published for this urban-to-rural gradient²², strongly suggest that higher rates of N cycling underlie the reductions of CH₄ consumption in the urban forests^{10,12}. We found that soil NH₄⁺ and TN contents tended to be higher in the rural than that in suburban and urban forest sites, which was in conflict with the generally accepted idea that soil N contents tend to be higher in urban than rural forests²³. However, our result was comparable with previous studies within the same region^{22,24}. The cause may be a higher rate of N losses in the urban site²².

Thirdly, CH₄ uptake might also be reduced by the changes in soil pH value and Al³⁺ content of the urban forests. A decrease of soil pH in urban forests might lead to decreased CH₄ uptake²⁵, which was noted in the Broadbalk Experiment in southeastern England²⁶. Soil acidification might cause a release of heavy metals, such as Al³⁺, inhibiting soil CH₄ uptake²⁶. Al³⁺ toxicity could inhibit CH₄ uptake²⁷, which potentially explains the results from our study.

Finally, the suppressive effect of urbanization-induced environmental changes on CH₄-consuming bacteria might be another mechanism for the CH₄ reduction. Urban environments might decrease CH₄ uptake indirectly through changes in the habitat of the methanotrophic bacteria²⁸. Long-term N deposition could cause a decrease in methanotrophic populations via niche competition with nitrifiers²⁹, leading to a reduction in CH₄ consumption³⁰. In the present study, a significant decrease in soil MBC at the urban site compared to the rural and suburban forest sites, might negatively affect the size, composition and activity of the CH₄ oxidizing community^{10,31}. The urbanization-induced environmental changes might have profound implications for CH₄ consuming communities and further research should be conducted.

In summary, the largest consumption of CH₄ was found in the rural, followed by the suburban and urban forest sites. The data presented here strongly indicated that the reduction of CH₄ uptake in urban forests was due to higher soil WFPS, and adverse effects of urbanization-induced environmental changes, such as atmospheric N deposition, higher soil NO₃⁻ content and Al³⁺ toxicity. This is the first *in situ* study to attempt to clarify the effects of urbanization-induced environmental changes on soil CH₄ uptake in tropical and subtropical forests. Our results suggest that the projected environmental changes associated with urbanization would decrease CH₄ consumption in urban forest soils of subtropical regions, and this reduction of CH₄ consumption needs to be considered in global CH₄ budgets.

Table 3 | Responses of CH₄ uptake rates to soil variables: multiple regression analysis and partitioning the variance in R²

Forest type		MF			BF			MF + BF		
Y variable	X variable	R ²	P	% of R ²	R ²	P	% of R ²	R ²	P	% of R ²
CH ₄ uptake	All variables	0.74	0.02		0.76	0.01		0.50	0.01	
	WFPS	0.30		41	0.41		54	0.26		52
	NO ₃ ⁻	0.32		43	0.16		21	0.15		30
	Al ³⁺	0.10		13	0.14		18	0.05		10
	pH	0.02		3	0.05		7	0.04		8

All sampling times were combined on July 2009. The relative importance of each "X" variable in the regression is indicated by the % of R² for which it accounts. WFPS are the soil water content when the soil cores were harvested. MF, mixed forest; BF, broadleaf forest. MF + BF, combined the data of the six forests.



Methods

Study area. The study sites were located along a 150 km gradient that extends from MFS (an urban site near Guangzhou City), through DHS (a suburban site), to HSD (a rural site) within the Pearl River Delta (PRD) region of Guangdong Province, South China (Fig. S2). Some characteristics of the study sites were presented in Table 1. The PRD region is one of the regions experiencing rapid urbanization with its population increasing nearly two fold from 1982 (54 million) to 2010 (104 million)³². The study region has a subtropical monsoon climate. There was a gradient for environmental variables from Guangzhou City to its surroundings (Table 1).

At each site, a MF and a BF were selected for experimental plots. Several criteria were used in study site selection to ensure comparability among the forests: (1) no disturbance after planting (such as fire, insect infestations, logging, and fertilization); (2) forest age between 50 and 70 year (excluding the BF in DHS); (3) soil of lateritic red earth (Table S1). The dominant species and plant characteristics are described in Table S1. Within each forest, five 10 m × 10 m plots were randomly established at least 100 m from the edge to avoid “edge effects”.

Measurement of CH₄ fluxes. Soil CH₄ fluxes were measured from April 2009 to March 2010 using the static chamber method. Gas samples were collected biweekly during the growing season (April to September) and monthly at other times. The chamber design and the measurement procedure were adopted from Zhang et al.¹³. Gas samples were collected with 60 ml plastic syringes at 0, 15 and 30 min after the cover chamber closure, and immediately injected into special gas bags (Yile CO₂ LTD, Shanghai, China). The samples were transferred to a lab and analyzed within 2–3 days by a gas chromatography with flame ionization detection (FID). Atmospheric pressure, air temperature (inside chamber), soil temperature (5 cm depth) and moisture (0–10 cm depth) were measured during each sampling event. Soil moisture was converted to WFPS (%). Details of the measurement were referenced from Zhang et al.¹³.

Soil sampling and analysis. Soil samples (0–10 cm depth) were collected on July 2009. Three soil cores (3.5 cm internal diameter, ID) were collected randomly from each plot and combined to one composite sample. The samples were passed through a 2 mm sieve and divided to two parts. One subsample was used for the analysis of NH₄⁺, NO₃⁻, MBC, and MBN. The other was air dried at 25°C for the estimation of other chemical parameters. Gravimetric water content was determined through oven drying at 105°C for 48 h.

Soil NH₄⁺ and NO₃⁻ contents were determined by extraction with 2 M KCl solution followed by colorimetric analysis on a flow-injection autoanalyzer (Lachat Instruments, Milwaukee, USA). TN content was determined by micro-Kjeldahl digestion³³, followed by detection of ammonium with a UV-8000 Spectrophotometer (Metash Instruments Corp., Shanghai, China). Soil MBC and MBN contents were estimated by chloroform fumigation-extraction method³⁴. For each sample, two fresh soil subsamples (10 g dry soil equivalent) were prepared. One subsample was fumigated with chloroform for 24 h at 25°C. The other was treated as control. Soil MBC and MBN contents were calculated as the difference in extractable C, N between fumigated and non-fumigated subsamples, using the conversion factors of 0.33 and 0.45 for MBC and MBN, respectively^{35,36}.

Atmospheric N deposition. We used ion-exchange resin (IER) columns to monitor N deposition in precipitation at the three sites during the study period. The IER columns and the measurement procedure were adopted from Fang et al.²². The resin columns were collected with interval for three months. The concentrations of NH₄⁺ and NO₃⁻ were determined by 2 M KCl extraction as described above.

Statistical analysis. Repeated Measures Analysis of Variance (ANOVA) was used to examine the differences of soil CH₄ fluxes among the three sites. One-way ANOVA was performed to compare the differences in soil properties, MBC, and MBN within the same forest type. Linear regression analysis was performed to quantify the relationships between CH₄ fluxes and individual soil variables. Multivariate linear regression analysis was performed using CH₄ uptake rate as dependent variable, and soil WFPS, NO₃⁻, Al³⁺ contents, and pH values as independent variables. R² was partitioned to quantify the importance of each independent variable. All statistical analyses were conducted using SPSS 16.0 for windows (SPSS Inc., Chicago, IL, USA). Statistically significant difference was set at P ≤ 0.05 unless otherwise stated. Mean values ± 1 standard error were reported.

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Author contributions

Original ideas for the research came from W.Z., J.M.M. and Y.T.F.; K.Y.W., T.Z., X.M.Z. and H.C. undertook all sampling and analysis; All authors contributed to the interpretation of the work and reviewed the manuscript.

Additional information

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