



Declines in methane uptake in forest soils

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Forest soils are a sink for atmospheric methane (CH₄) and play an important role in modulating the global CH₄ budget. However, whether CH₄ uptake by forest soils is affected by global environmental change is unknown. We measured soil to atmosphere net CH₄ fluxes in temperate forests at two long-term ecological research sites in the northeastern United States from the late 1990s to the mid-2010s. We found that annual soil CH₄ uptake decreased by 62% and 53% in urban and rural forests in Baltimore, Maryland and by 74% and 89% in calcium-fertilized and reference forests at Hubbard Brook, New Hampshire over this period. This decrease occurred despite marked declines in nitrogen deposition and increases in atmospheric CH₄ concentration and temperature, which should lead to increases in CH₄ uptake. This decrease in soil CH₄ uptake appears to be driven by increases in precipitation and soil hydrological flux. Furthermore, an analysis of CH₄ uptake around the globe showed that CH₄ uptake in forest soils has decreased by an average of 77% from 1988 to 2015, particularly in forests located from 0 to 60 °N latitude where precipitation has been increasing. We conclude that the soil CH₄ sink may be declining and overestimated in several regions across the globe.

soil methane uptake | increased precipitation | hydrological flux | long-term ecological research sites | greenhouse gases

Methane (CH₄) is the second-most significant anthropogenic greenhouse gas after carbon dioxide (CO₂) (1) and its atmospheric concentration has been increasing rapidly over recent decades (2). Forest soils are a major sink for atmospheric CH₄ (3–5) and some studies suggest that this sink has increased in recent decades (6). Methane in soils is consumed by aerobic methanotrophs and produced by anaerobic methanogens (7), so its net flux is controlled by a complex suite of factors that affect diffusion and soil oxygen conditions (8). While measurements of CH₄ flux have been made in forests worldwide (SI Appendix, Fig. S1), most studies have been relatively short term (<2 y) (SI Appendix, Fig. S2), inhibiting our ability to examine how this flux responds to subtle changes in environmental conditions. The study reported here is the longest duration of a total of 498 entries obtained from 317 peer-reviewed journal articles published before December 2017 (SI Appendix, Supplementary Methods and Dataset S1).

Urbanization is a dominant driver of global environmental change (9, 10) and cities produce 70% of anthropogenic emissions of greenhouse gases (11). Urbanization creates unique environmental conditions—with alterations of soil temperature, moisture (12), and nitrogen (N) deposition (13)—and remnant natural ecosystems within cities can be useful study areas for assessment of how environmental change influences biogeochemical processes (14), such as soil CH₄ uptake (15). Studies have found that urban soils have lower CH₄ uptake than rural soils in forests (16), but there has been little analysis of long-term changes in CH₄ uptake in highly dynamic urban versus reference rural ecosystems.

Precipitation and atmospheric chemistry are other important drivers of global environmental change (17). Marked changes in the amounts of rainfall and deposition of acid and nitrogen that have occurred in many parts of the world, including northeastern

North America (18, 19), may affect soil CH₄ uptake (20). There is a clear need for long-term analysis of how this important flux is (or is not) responding to multiple components of global environmental change.

Methods

Monthly in situ chamber measurements of soil–atmosphere CH₄ fluxes were made at two United States National Science Foundation-funded long-term ecological research (LTER) sites in the northeastern United States. Methane fluxes were measured in four urban and four rural forest sites of similar age and composition at the Baltimore, Maryland LTER site (SI Appendix, Table S1) from November 1998 to December 2016 (21) and at four calcium (Ca)-fertilized and four reference sites along an elevation gradient in two forested watersheds at the Hubbard Brook, New Hampshire LTER site (SI Appendix, Table S2), from 2002 to 2015. Measurements in Baltimore were approximately monthly and those at Hubbard Brook were limited to summer months. To our knowledge, these are the longest duration of soil–atmosphere CH₄ flux measurements in forests (SI Appendix, Fig. S2). We also continuously monitored soil moisture (12), N leaching (with zero tension lysimeters) (22), and microbial activity (23) at various soil depths at these sites (SI Appendix, Supplementary Methods).

To provide global context for our detailed site measurements, we retrieved data on annual ($n = 756$) (Dataset S2) and monthly ($n = 9,789$) (Dataset S3) CH₄ uptake in forest soils around the world from 317 published articles, and assembled precipitation data (Dataset S4) for the period from 1987 to 2016. We used these data to estimate mean annual CH₄ uptake over 30° latitude bands across the globe and to examine relationships between changes in precipitation and CH₄ uptake in a latitudinal context (SI Appendix, Supplementary Methods).

Significance

Atmospheric methane (CH₄) concentration has been increasing rapidly over recent decades. Forest soils are a major sink for atmospheric CH₄, but evidence from long-term in situ observation is limited, so little is known about how the soil CH₄ sink responds to changing environmental conditions. We measured soil to atmosphere net CH₄ fluxes at long-term ecological research sites in Baltimore, Maryland (1998–2016) and Hubbard Brook, New Hampshire (2002–2015) and found significant decreases in CH₄ uptake at both sites. Moreover, a literature review showed that CH₄ uptake in forest soils around the world is also declining, especially forests from 0–60 °N latitude, where precipitation has been increasing. We conclude that the current soil CH₄ sink may be overestimated over large regional areas.

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Results and Discussion

Mean CH_4 uptake rates were $1.3 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$ in the Baltimore rural forest soil (Fig. 1A) and $1.0 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$ in the Hubbard Brook reference forest soil (Fig. 1B) over the full study period.

In Baltimore, CH_4 uptake significantly decreased by 62% (from 1.2 to $0.46 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$) and 53% (from 2.0 to $0.96 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$) in the urban and rural forest soils, respectively, over the 18-y study period (Fig. 1A and *SI Appendix*, Tables S3 and S4). These declines were significant over all individual sites (*SI Appendix*, Figs. S3 and S4 and Table S5). Methane uptake varied seasonally with high values in summer and autumn (*SI Appendix*, Fig. S5) due to higher temperatures and lower soil moisture (*SI Appendix*, Fig. S6).

At Hubbard Brook, CH_4 uptake significantly decreased by 74% (from 1.7 to $0.44 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$) and 89% (from 2.0 to

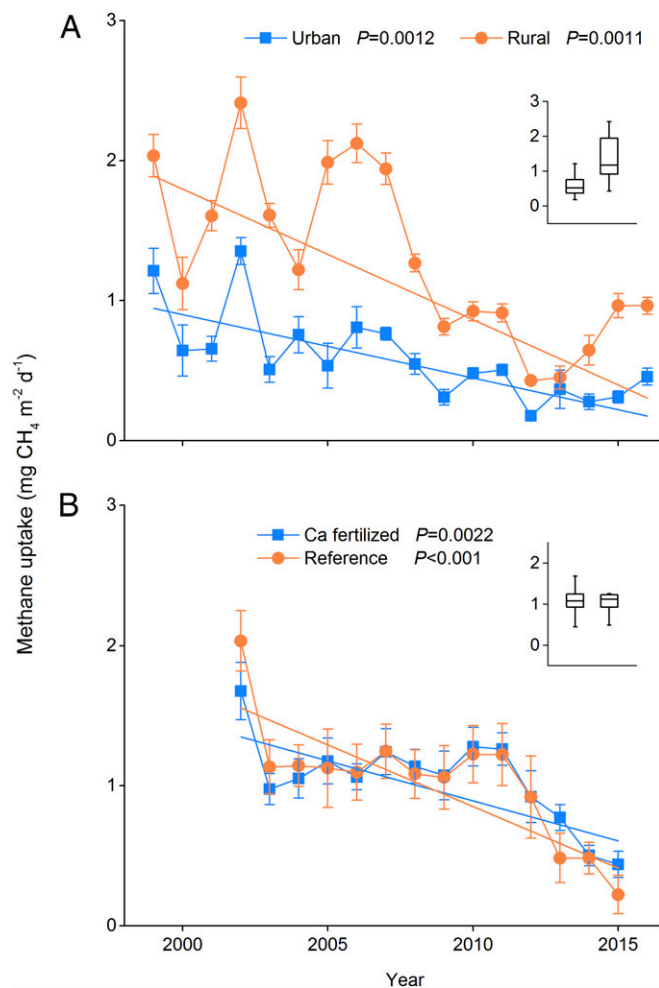


Fig. 1. Annual CH_4 uptake. (A) CH_4 uptake (\pm SE, $n = 8$ –16) in urban and rural forests in Baltimore, Maryland, from 1999 to 2016 (annual means calculated for a “water year” from October to September). (B) CH_4 uptake (\pm SE, $n = 12$) in calcium (Ca) fertilized and reference forests at Hubbard Brook, New Hampshire, from 2002 to 2015. Differences between forest types are significant in Baltimore (all $P < 0.05$ except in 2000 and 2013) but not significant (all $P > 0.05$) at Hubbard Brook for each year. The *Insets* represent average CH_4 uptake during the study period in urban (*Left*) versus rural (*Right*, $n = 18$, $P < 0.001$) or Ca-fertilized (*Left*) versus reference (*Right*, $n = 14$, $P = 0.87$) forests. The boxes show median and 5th and 95th percentiles. All trends with time are statistically significant ($P < 0.01$) (*SI Appendix*, Table S4).

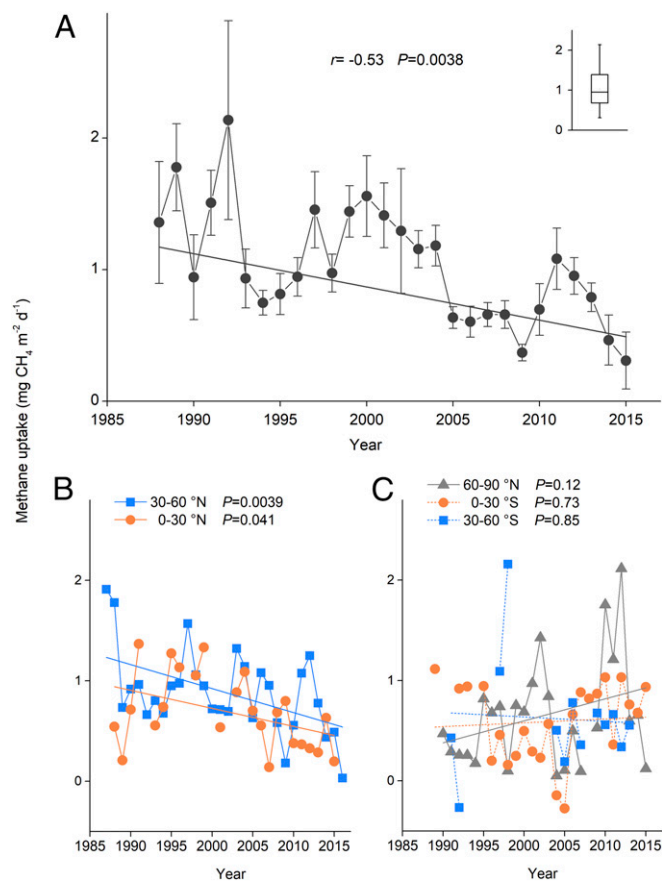


Fig. 2. Methane uptake in forest soils retrieved from published studies. (A) Global annual CH_4 uptake in forest soils from 1988 to 2015. The *Inset* represents average CH_4 uptake for the full period from 1988 to 2015. The boxes show median and 5th and 95th percentiles. Error bars denote SEs. (B) Annual CH_4 uptake in forest soils from 0 to 30°N and 30 – 60°N latitude. (C) Annual CH_4 uptake in forest soils from 60 to 90°N , 0 – 30°S and 30 – 60°S latitude. All trends with time are statistically significant in A and B ($P < 0.05$) (*SI Appendix*, Table S4) but are not significant in C. The data in A were actually measured annual CH_4 uptake and those in B and C include both actually measured and estimated annual CH_4 uptake (*SI Appendix*, *Supplementary Methods*). The data collected in Baltimore and Hubbard Brook were excluded from this literature analysis.

$0.22 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$) in the Ca-fertilized and reference forest soils, respectively, over the 14-y study period (Fig. 1B and *SI Appendix*, Table S4). If data from 2002 were removed, the decreases were 55% (from 0.98 to $0.44 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$) and 80% (from 1.1 to $0.22 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$) in the Ca-fertilized and reference forest soils, respectively, over the 13-y study period.

Global mean CH_4 uptake in forest soils (derived from our literature analysis) over the period from 1988 to 2015 was $1.03 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$ (Fig. 2A), similar to the global mean uptake level ($1.15 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$) reported in a recent meta-analysis (4). Our analysis also shows a 77% decrease in CH_4 uptake over the past three decades, from $1.4 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$ in 1988 to $0.31 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$ in 2015. These declines were only observed in forests located in the 0 – 30°N and 30 – 60°N latitude bands (Fig. 2B); they were not present in the Arctic or in the Southern Hemisphere (Fig. 2C and *SI Appendix*, Table S4).

Methane uptake has been found to be strongly affected by several factors (8, 24, 25), several of which have varied in surprising ways over the course of our long-term studies (18, 26) (*SI Appendix*, Fig. S7). The declines in CH_4 uptake that we observed at two LTER sites coincided with increases in atmospheric CH_4

concentration (*SI Appendix, Fig. S8A*), marked declines in atmospheric N deposition (13) (*SI Appendix, Fig. S8B*), and increases in air temperature (*SI Appendix, Fig. S7A*), all of which should have stimulated soil CH₄ uptake (27, 28). Previous research at the Baltimore sites found that strong differences in CH₄ uptake between urban and rural forest soils (15) were driven by higher rates of N cycling, especially nitrification, in the urban forest soils (29). The results here confirm the large differences in N cycling and availability between urban and rural forest soils (*SI Appendix, Table S3*), but there was no significant long-term temporal change in either the concentration or flux of inorganic N in soil leachate over the 18-y time period (both $P > 0.05$). At Hubbard Brook, soil N cycling decreased during the study period (30). We therefore cannot attribute the declines in CH₄ uptake in these temperate forest soils to changes in either N deposition or cycling.

There is a long-term trend of increased precipitation in the northeastern United States (26) (*SI Appendix, Fig. S7B*). In Baltimore there was also a marked increase in leachate volume (the water collected in zero tension lysimeters) (*SI Appendix, Fig. S9A*) and significant temporal variation in soil moisture (*SI Appendix, Table S3*) over the 18-y time period. A multivariate analysis of 28 variables found that leachate volume and soil moisture were the dominant factors influencing CH₄ uptake in Baltimore forest soils (*SI Appendix, Fig. S10*). At Hubbard Brook we have observed significant increases in soil moisture over the past 14 y (*SI Appendix, Fig. S9B*). The trend in soil moisture at Hubbard Brook was only significant in the Ca-fertilized watershed, but trends in this variable are difficult to see in infrequently sampled plots (one to five times per year). More comprehensive analyses, based on continuous data, show general increases in soil moisture at Hubbard Brook (26). These results suggest that increases in soil water flux underlie the long-term observed decline in CH₄ uptake at both sites (*SI Appendix, Fig. S11*), either by inhibiting the flow of substrate to consumers or by increasing production in anaerobic microsites (7). The fact that CH₄ uptake decreased in both urban and rural forest soils in Baltimore, and in both Ca-fertilized and reference forest soils at Hubbard Brook with very different temperature (*SI Appendix, Fig. S7A*) and N availability regimes (22, 29), supports the idea that CH₄ uptake in forest soils is more sensitive to soil moisture than to temperature or N cycling (3).

At a much larger scale, the decline in CH₄ uptake in forest soils in the latitudes of 0–60 °N (*Fig. 2B* and *SI Appendix, Fig. S12*) was coincident with increases in precipitation in these areas

(*SI Appendix, Fig. S13*). This result is in accordance with another global analysis (6). In addition, our analysis around the world also found that CH₄ uptake was consistently lower at high soil moisture levels (*SI Appendix, Fig. S14*), further supporting the idea that changes in precipitation and hydrological flux are the key drivers of regional-scale changes in CH₄ uptake in forest soils.

Declines in CH₄ uptake were not observed in arctic areas or in the Southern Hemisphere (*Fig. 2C*). One possible reason for this is that there have been few in situ measurements in these areas (*SI Appendix, Fig. S12*), hindering our ability to assess the long-term trends in CH₄ uptake in these forest soils. Moreover, some studies have found that soil CH₄ uptake in subtropical and temperate forests is lower in the Northern Hemisphere than in the Southern Hemisphere (3, 6). Thus, the effects of changes in hydrological flux (i.e., increase in precipitation) on CH₄ uptake in Southern Hemisphere forest soils may be important and worthy of further study.

Global budgets show that upland soils are a major sink for atmospheric CH₄ (3–5), and some analyses suggest that this sink has increased in recent decades (6). Here we find that soil CH₄ uptake in two temperate forests in the northeastern United States has declined to rates of 0.18–0.22 mg CH₄ m⁻² d⁻¹, and our regional-scale analysis shows a marked decline in annual mean CH₄ uptake over the past three decades to 0.31 mg CH₄ m⁻² d⁻¹ in 2015. These uptake rates are much lower than current estimates for temperate forests (3, 4, 6) (0.65–1.6 mg CH₄ m⁻² d⁻¹). Our results therefore suggest that the current soil CH₄ sink may be overestimated in areas where precipitation and soil hydrological flux are increasing, particularly for the tropical/subtropical and temperate forests located between 0 and 60 °N latitude. Moreover, our results suggest that soil CH₄ uptake is much more sensitive to subtle shifts in soil hydrology than to marked changes in N cycling, increases in atmospheric CH₄ concentrations, and increases in temperature. These results need to be considered in assessments of the global CH₄ budget and of how this budget is likely to change in the future along with climate and atmospheric deposition.

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