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Supplementary Information for

Declines in methane uptake in forest soils

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Supplementary Methods

Study area

This study was conducted at long-term study sites of the Baltimore Ecosystem Study (BES) and the Hubbard Brook Ecosystem Study (HBR), which are components of the U.S. National Science Foundation funded Long-Term Ecological Research (LTER) network. BES research is centered on the Gwynns Falls watershed in Baltimore, Maryland, USA (39°15' N, 76°30' W; www.beslter.org). The mean annual air temperatures are 14.5 °C and 12.8 °C, and the mean annual precipitation are 1125 mm and 1153 mm for the urban and rural forests, respectively (12) (Fig. S7). Vegetation at these sites is dominated by tulip poplar (*Liriodendron tulipifera* L.) and oaks, primarily chestnut (*Quercus prinus* L.), scarlet (*Quercus coccinea* Münchh.) and white (*Quercus alba* L.). Plant community composition, soil characterization, nitrogen (N) mineralization and nitrification, soil solution chemistry and soil-atmosphere trace gas fluxes are described by Groffman et al. (21) and are summarized in Table S1.

In 1998, four urban and four rural forest sites (four chambers per site) were established at three remnant forests with at least 80% forest canopy to evaluate the influence of urban environment on forest ecosystems (Table S1; more details were described in ref. 21). The urban forest sites are located in Hillsdale Park (HD) and Leakin Park (LEA, two sites each), which are public parks close to the urban core of Baltimore City (5-7 km). Sampling at plot HD2 was discontinued in June 2005 due to continuing vandalism. The rural forest sites are located in Oregon Ridge Park (two sites at an upper slope location (ORU) and two sites at a middle slope location (ORM)), which is 20-25 km from the urban core of Baltimore City. Sampling was stopped at the two ORM sites in June 2010. All sites are more than 100 m from roads or houses, except for the Hillsdale sites, which are located in a smaller tract less than 100 m from an urban neighborhood (15).

HBR is located in the White Mountain National Forest in New Hampshire, USA (43° 56' N, 71° 45' W; www.hubbardbrook.org). Vegetation is dominated by American beech (*Fagus grandiflora*), sugar maple (*Acer saccharum*) and yellow birch (*Betula alleghanieusis*). The site was selectively cut around 1900 and was damaged by a hurricane in 1938. Soils are shallow (75-100 cm), acidic (pH 3.9), Typic Haplorthods developed from unsorted basal tills. Detailed site information can be found in Groffman et al. (23) and is summarized in Table S2.

This study was conducted in two forested watersheds at HBR: watershed 1 and Bear Brook, which is just west of the long-term biogeochemical reference watershed (watershed 6) at the site. In October 1999, watershed 1 was fertilized with wollastonite ($CaSiO_3$) at a rate of 850 kg Ca/ha to replace Ca that had been depleted by decades of acid deposition (31). Watershed 1 has been continuously monitored for streamflow since 1956 and stream chemistry since 1963 (17). Both the Ca fertilized and reference forests are located on south facing slopes (20-30% slope) along an elevation gradient with similar vegetation. Four sites (three chambers per site) were established in hardwood forests at low (520-560 m), middle (600-650 m) and high elevations (725-750 m) and in a spruce-fir-birch forest (770-850 m) in each watershed (23).

CH_4 flux data collection and analysis

Methane flux at the soil-atmosphere interface was measured using an *in situ* chamber method (32). Four replicate permanently installed polyvinyl chloride (PVC) base rings, at least 5 m apart, were installed at each site in Baltimore forests in 1998 and three replicates were installed in Hubbard Brook forests in 2002. For sampling, PVC cylinders (287 mm in diameter and 40 mm high, fitted with an airtight septum on the chamber tops as gas sampling ports) were placed on the base rings and 9 ml gas samples were collected using polypropylene syringes at 0, 10, 20 and 30 minutes following placement of the PVC cylinders (15, 33). All samplings were carried out between 10:00-14:00 hours. Gas samples were transferred to evacuated glass vials (vacuum for 2 minutes) and stored at room temperature prior to analysis by gas chromatography (GC, Shimadzu, Kyoto, Japan) with a flame ionization detector at the Cary Institute of Ecosystem Studies laboratory in Millbrook, NY, USA. The smallest CH_4 concentration that the GC was able to detect was 0.0156 ppm. Standard CH_4 gases were processed along with each batch of flux samples. These standards were generated by filling sample vials from a standard tank in the field laboratory and then processing these samples along with samples taken from flux chambers in the field. Percent recoveries of the trip standards were used to correct flux values for leakage or gas absorption that may occur between sampling and analysis. These measurements began in Baltimore in November 1998 and were made at approximately monthly (4-6 week) intervals. Measurements at Hubbard Brook began in August 2002 and were made 1-5 times per year during the growing season.

To rule out the possibility that the long-term decline in soil CH_4 uptake was due to repeated use and possible soil compaction in the *in situ* chambers, we installed two new chambers in one of the urban (LEA1) and one of the rural forest sites (ORU2) in Baltimore in June 2015. Methane fluxes were measured in both new and old chambers through February 2016. Seven months of sampling found no significant (both $P>0.05$; Fig. S15) differences between old and new chambers, suggesting that the observed declines in CH_4 uptake in these forest soils were not an artifact of long-term sampling.

As described in Groffman et al. (34), CH_4 fluxes were calculated from the linear rate of change in gas concentration, the chamber internal volume and soil surface area. Flux rate calculations were not corrected for actual *in situ* temperature and pressure. Single points were removed from regressions if they were more than 6 times higher or lower than the other three values or if they contradicted a clear trend in the other three points. This procedure prevents inclusion of high flux rates based on non-significant regressions. Non-significant regressions were used in flux calculations. This is preferable to setting these regressions to zero, which biases the statistical distribution of rates.

Methane uptake rate (U_{CH4}, mg CH_4 m^{-2} d^{-1}) was calculated as follows:

$$U_{CH4} = -\rho \times \frac{V}{A} \times \frac{\Delta c}{\Delta t} \times 0.001$$

where ρ is the density of CH_4 (0.716×10^6 mg m^{-3}) under standard conditions (temperature of 0 ℃ and pressure of 101.325 kPa); V (m^3) and A (m^2) are the volume and area of the chamber, respectively; $\Delta c/\Delta t$ (10^{-6} m^3 m^{-3} h^{-1}) is the rate of change in CH_4 concentration inside the chamber during the measurement period (0, 10, 20 and 30 minutes).

Soil moisture at various soil depths in Baltimore

In the Baltimore forests, time domain reflectometry waveguide probes (Soilmoisture Equipment Corporation, Santa Barbara, CA, USA) were installed horizontally at 10, 20, 30 and 50 cm depths and vertically over 0-20 cm depth to measure volumetric soil moisture. Soil moisture data were recorded by SoilMoisture Trace System I (Model 6050 × 1, version 2000) on each monthly sampling date. Given that soil moisture at all soil depths showed similar patterns, we only present data from the 10 cm soil depth.

Soil water flux in Baltimore

In the Baltimore forests, zero tension lysimeters (two or three replicates in two or three plots, total 6 replicates at each site) were installed in soils at 50 cm depth in fall 1998 to collect leachate exported from upper soil depths (22). At the same time, porous cup tension lysimeters (four replicates per plot) were installed at 10 and 50 cm depths. To collect leachate samples from tension lysimeters, lysimeters were manually suctioned by pump (inner pressure >0.6 MPa) 24 hours before sampling. Leachate samples were collected with a hand pump at approximately monthly intervals at approximately the same time that measurements of CH_4 flux and soil moisture were made. The volume of leachate collected from lysimeters was measured immediately, and approximately 100 ml samples were stored at 4 ℃ prior to analysis. Concentrations of nitrate (NO_3^-) and ammonium (NH_4^+) were analyzed colorometrically using a flow injection analyzer. Monthly data were compiled to produce annual values using a “water year” from October to September of the following year. All data are available in the BES LTER (www.beslter.org) data repository.

Soil sampling at Hubbard Brook

In the Hubbard Brook forests, soil samples (Oi/Oe, Oa/A and 0-10 cm mineral soils) at the four elevation sites (5 plots per site) in both watersheds were collected once (July) or three (May, July and October) times per year beginning in 2002. Four soil core samples were taken at each plot and were hand-sorted and mixed and stored at 4 ℃ prior to analysis (less than 1 week).

Soil moisture was determined gravimetrically by drying soil samples at 60 ℃ for 48 h. Amounts of soil NO_3^- and NH_4^+ were determined by extraction with 2 mol/l KCl followed by colorimetric analysis with a flow injection analyzer. Microbial biomass C and N contents were measured using the chloroform fumigation-incubation method and potential net N mineralization and nitrification were measured in 10 day incubations of field moist soil and laboratory temperatures. In this study, we only used 0-10 cm mineral soil data collected in summer from 2002 to 2015. All these data are available in the HBR LTER (www.hubbardbrook.org) data repository.

Atmospheric and climate data

Atmospheric CH_4 concentration data were retrieved from the U.S. National Oceanic and Atmospheric Administration (NOAA) Global Greenhouse Gas Reference Network (2) (https://www.esrl.noaa.gov/gmd/ccgg/trends_ch4; Dataset S5). Atmospheric N deposition data were retrieved from the U.S. Environmental Protection Agency at the Beltsville (35) (BEL116; https://www3.epa.gov/castnet/site_pages/BEL116.html) and Woodstock sites (36) (WST109; https://www3.epa.gov/castnet/site_pages/WST109.html) to represent N deposition levels in Baltimore and Hubbard Brook respectively (Dataset S6).

Monthly temperature and precipitation data in Baltimore were retrieved from the U.S. NOAA National Centers for Environmental Information at the Baltimore Washington International Airport site (37) (https://gis.ncdc.noaa.gov/maps/ncei/summaries/monthly; Dataset S7) and were compiled into mean annual values by “water year”. For Hubbard Brook, temperature data were taken from weather stations 1 and 6 and precipitation data were taken from rain gages 1, 2 and 3 in watershed 1 and rain gages 9, 10 and 11 in watershed 6.

Data analysis

Annual values for Baltimore were based on a “water year” (from October to September of the following year), while Hubbard Brook annual means were based on a calendar year. Seasonal dynamics were analyzed by combining data from different years for each month. We first tested for the effects of forest type (urban versus rural in Baltimore or Ca fertilized versus reference at Hubbard Brook) over time using a repeated measures analysis of variance (ANOVA). We also used one-way ANOVA to examine changes in annual CH_4 uptake over time for each forest type and general linear models for each individual site. Differences between forest types in Baltimore and at Hubbard Brook were examined by paired t-test or Wilcoxon signed-rank test if sample sizes were unequal using MATLAB R2012a (MathWorks Inc., Natick, MA, USA).

A total of 28 factors were monitored in Baltimore forests but some data were missing on certain sampling dates (sample sizes were unequal), so we used a partial least squares regression in SIMCA 14.0 (Umetrics, Umeå, Sweden) to distinguish the importance of the different factors on soil CH_4 uptake. We also used a structural equation model in AMOS 22.0 (IBM SPSS, Chicago, IL, USA) to examine the effects of soil leachate volume (and/or soil moisture), concentrations of soil NO_3^- and NH_4^+, microbial biomass N and microbial respiration to examine the relative importance of soil hydrological flux, N cycling and microbial activity on soil CH_4 uptake. In this analysis, each entry must have the data for all of the variables (leachate volume, volumetric soil moisture, NO_3^- concentration, NH_4^+ concentration and microbial respiration in Baltimore; gravimetric soil moisture, NO_3^- concentration, NH_4^+ concentration, microbial biomass nitrogen and microbial respiration at Hubbard Brook) and the entries were removed if data for one or more variables were missing (n=308 in Baltimore and n=155 at Hubbard Brook). The data were resampled (499 times) using a Monte Carlo method. Path coefficients were estimated using a Maximum likelihood method and confidence intervals were 95%.

Global *in situ* soil-atmosphere CH_4 measurements in forests

We compiled studies that measured soil-atmospheric CH_4 fluxes in forests worldwide (Dataset S1) using Web of Science (http://apps.webofknowledge.com). The search terms were "CH_4", "methane" or "trace gas" ("greenhouse gas", "GHG"). The disciplinary categories included environmental science, geoscience, soil science, meteorology atmospheric science, plant science, ecology, agronomy, microbiology, forestry and associated multidisciplinary sciences. Only peer-reviewed journal articles published before December 2017 with clearly defined study periods were used, and proceeding papers, reviews and meeting abstracts were excluded. Only *in situ* measurements with closed-chamber methods in forests and only the data collected from plots without any treatment were used. Years with less than three data points (e.g., 1981 and 1983) were not included in temporal trend analyses. To assess whether the declines in soil CH_4 uptake in the two studied temperate forests are general in other forests, the data collected in Baltimore (*15*) and Hubbard Brook (ref. 3 in *SI Reference*) were excluded from this literature analysis. A total of 317 articles (ref. 8-324 in *SI Reference*) that reported *in situ* CH_4 fluxes at the soil-atmosphere interface in forests were used. The average field monitoring duration was 1.4 years and 80% of the studies continually measured soil-atmosphere CH_4 fluxes for less than 2 years (Fig. S2).

A total of 756 entries of annual CH_4 uptake data (Dataset S2) were directly retrieved from the 317 published articles. Monthly CH_4 uptake data (9789 entries; Dataset S3) were also compiled if the annual data were not available. These monthly values were compiled to estimate annual uptake to expand the scope of our regional-scale analysis (3). All of these uptake estimates (10545 entries, including both annual and monthly data) were grouped into 30° latitude bands (60-90 °N, 30-60 °N, 0-30 °N, 0-30 °S and 30-60 °S). Annual precipitation data (from 1987 to 2016) were retrieved from the meteorological stations closest to these *in situ* CH_4 flux measurement sites using the NOAA website (https://gis.ncdc.noaa.gov/maps/ncei/cdo/annual; Dataset S4). We then calculated means for annual CH_4 uptake and mean annual precipitation for each 30 ° latitude band. In addition, continually monitored volumetric soil moisture data (a total of 3704 entries) were retrieved from the articles and used to examine the relationship between CH_4 uptake and soil moisture. When the data were presented as figures, we used Engauge Digitizer 4.1 (Free Software Foundation Inc., Boston, MA, USA) to extract values.

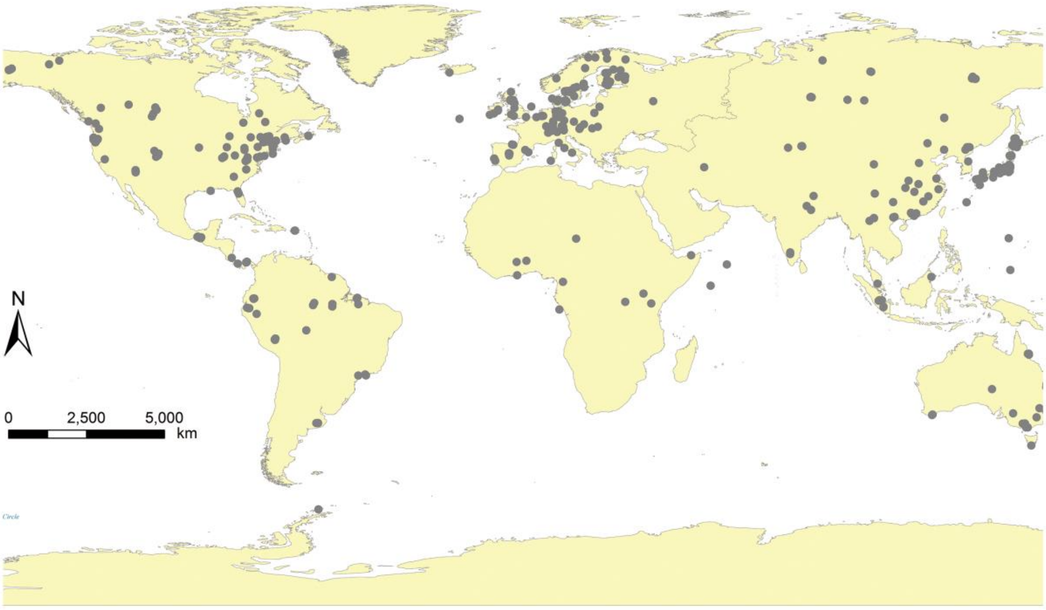


Fig. S1. Global distribution of *in situ* CH_4 flux measurements. A total of 501 entries were obtained from 317 peer-reviewed journal articles that were published before December 2017.

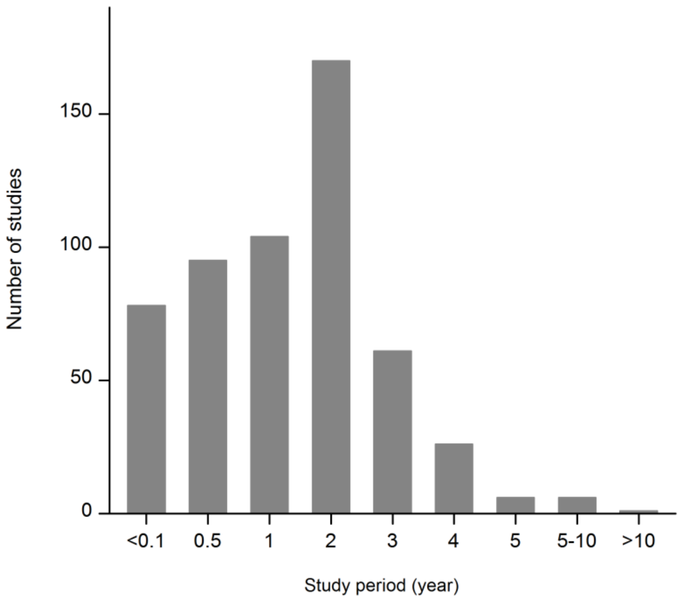


Fig. S2. Length of study periods of in situ CH_4 flux measurements obtained from 317 peer-reviewed journal articles that were published before December 2017. 80% of these studies were less than two years.

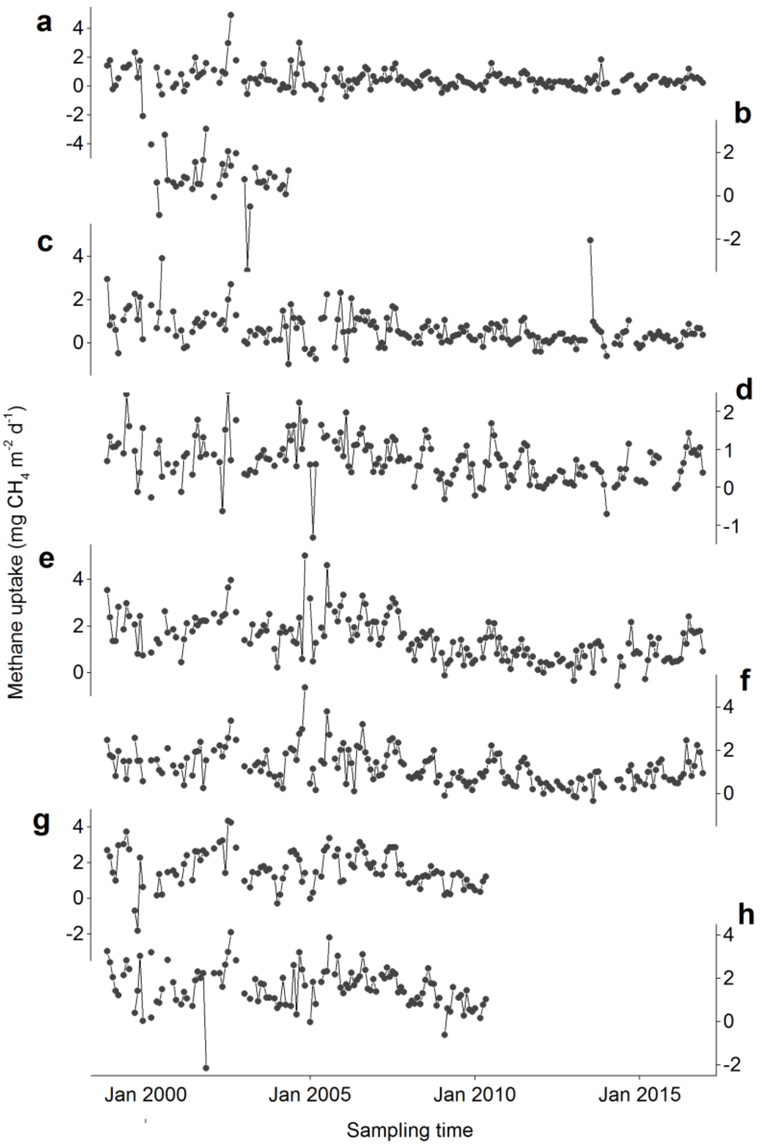


Fig. S3. Monthly CH_4 uptake in Baltimore forests from November 1998 to December 2016. (*a*) Hillsdale Park 1. (*b*) Hillsdale Park 2. (*c*) Leakin Park 1. (*d*) Leakin Park 2. (*e*) Oregon Ridge upper slope 1, (*f*) Oregon Ridge upper slope 2. (*g*) Oregon Ridge middle slope 1. (*h*) Oregon Ridge middle slope 2. Sampling was discontinued at the Hillsdale Park 2 site in June 2004 and at the two Oregon Ridge middle slope sites in June 2010. Sampling was carried out approximately monthly (4-6 week). Values are means of 4 replicates without error bars shown.

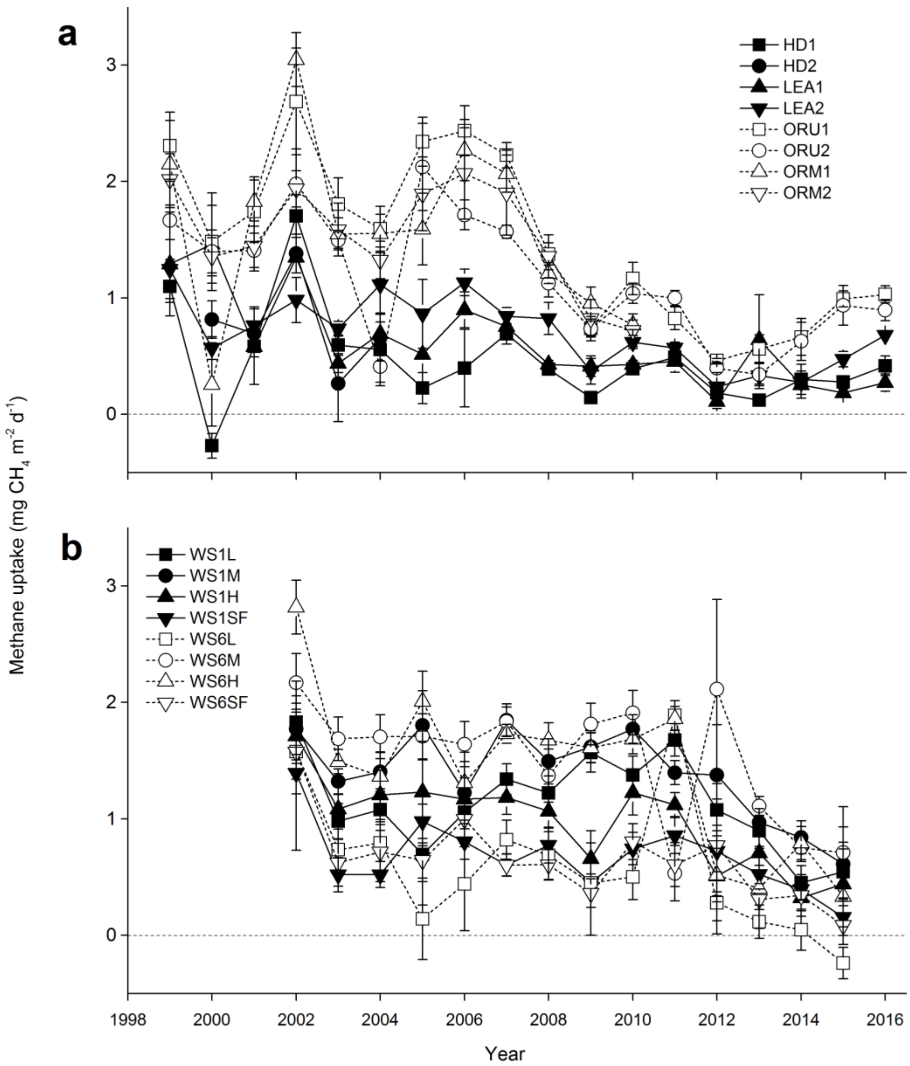


Fig. S4. Annual CH_4 uptake in each site. (*a*) Baltimore forests. HD1: Hillsdale Park 1, HD2: Hillsdale Park 2, LEA1: Leakin Park 1, LEA2: Leakin Park 2, ORU1: Oregon Ridge upper slope 1, ORU2: Oregon Ridge upper slope 2, ORM1: Oregon Ridge middle slope 1, ORM2: Oregon Ridge middle slope 2. The first four sites are urban forests and the latter four sites are rural forests. Values (±SE, n=4) are means of monthly data for each "water year" (from October to September of the following year). (*b*) Hubbard Brook forests. The sites are located along an elevation gradient with hardwood forests at low (L), middle (M) and high (H) elevations and a spruce-fir-birch (SF) forest in both watershed 1 (WS1, Ca fertilized) and watershed 6 (WS6, reference). Values are means ±SE (n=3). Methane uptake in all sites (except the spruce-fir-birch forest in watershed 1 at Hubbard Brook) significantly decreased over time (P<0.05; Table S5).

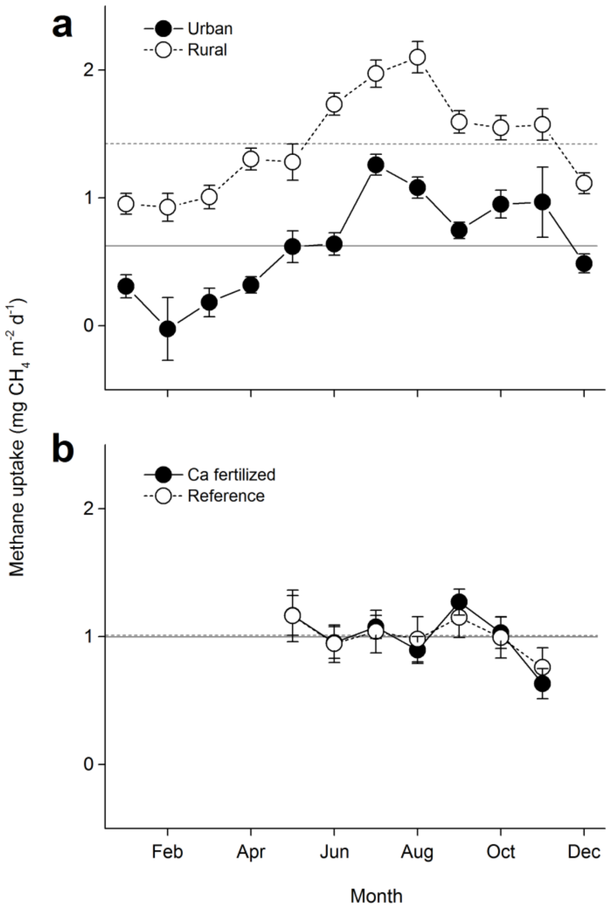


Fig. S5. Seasonal CH_4 uptake. (*a*) Baltimore forests. (*b*) Hubbard Brook forests. Values (±SE, *n*=16 in Baltimore and *n*=12 at Hubbard Brook) are means of all years for each month. Differences between urban and rural forests in Baltimore were significant ($P<0.05$) for all months except November. There were no significant differences between Ca fertilized and reference forests at Hubbard Brook. The horizontal lines represent the average values for urban (or Ca fertilized, *solid*) and rural (or reference, *dot*) forests separately.

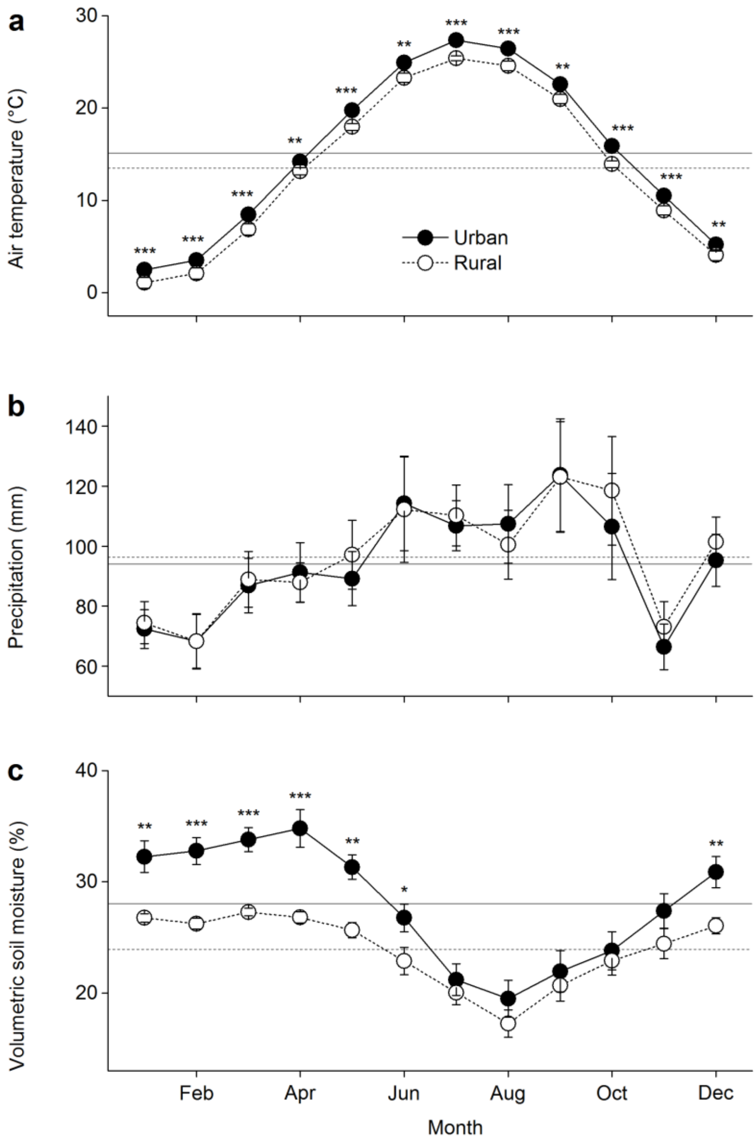


Fig. S6. Monthly temperature, precipitation and soil moisture in Baltimore forests. (*a*) Air temperature. (*b*) Precipitation. (*c*) Volumetric soil moisture at 10 cm soil depth. Values (±SE, n=18) are means of all years (from November 1998 to November 2016) for each month. The horizontal lines represent the average values for urban (*solid*) and rural (*dot*) forests separately. Asterisks show significant differences between urban and rural forests at $^{*}P<0.05$, $^{**}P<0.01$ and $^{***}P<0.001$.

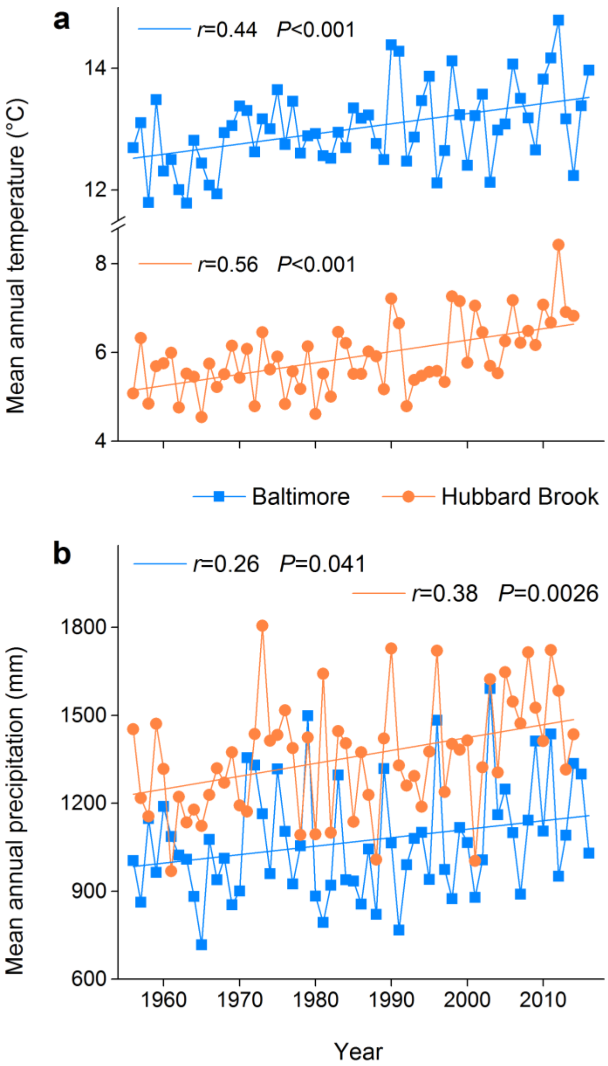


Fig. S7. Temperature and precipitation. (*a*) Annual air temperatures in Baltimore and Hubbard Brook. (*b*) Mean annual precipitation in Baltimore and Hubbard Brook. Pearson's *r* and *P* values of linear regression are shown in each panel.

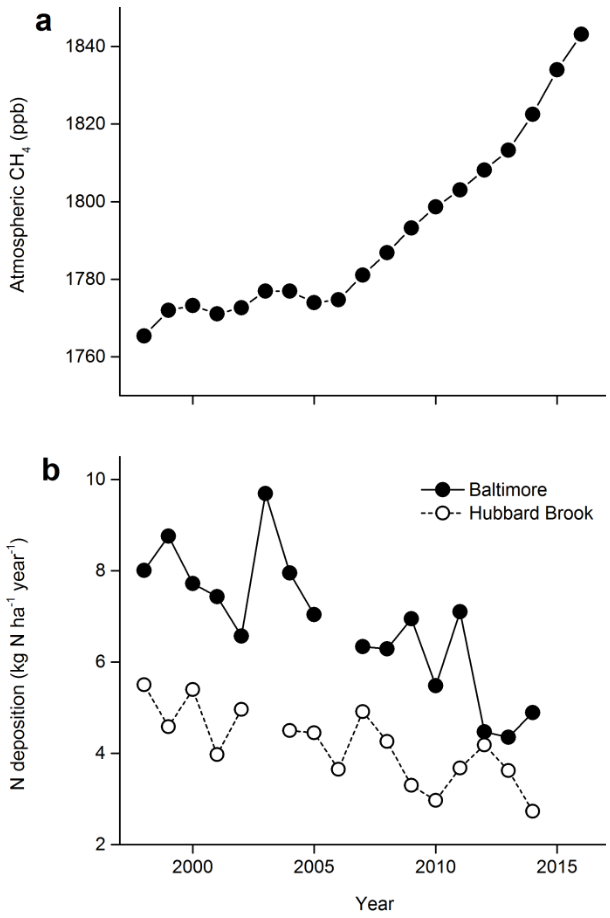


Fig. S8. Atmospheric CH_4, and nitrogen (N) deposition. (*a*) Global mean atmospheric CH_4 concentration. (*b*) Total atmospheric N deposition in Baltimore and Hubbard Brook.

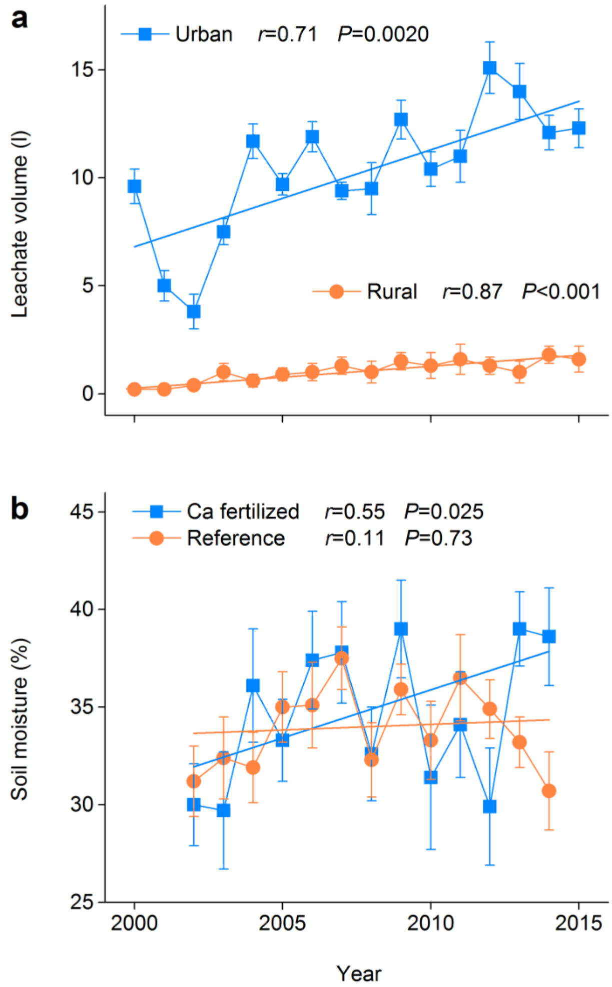


Fig. S9. Soil hydrology. (*a*) Leachate volume (±SE, *n*=24) collected from zero tension lysimeters at 50 cm soil depth in urban and rural forests in Baltimore. Values were compiled into "water year" (from October to September of the following year). (*b*) Gravimetric soil moisture (±SE, *n*=20) of 0-10 cm mineral soils in calcium (Ca) fertilized and reference forests at Hubbard Brook.

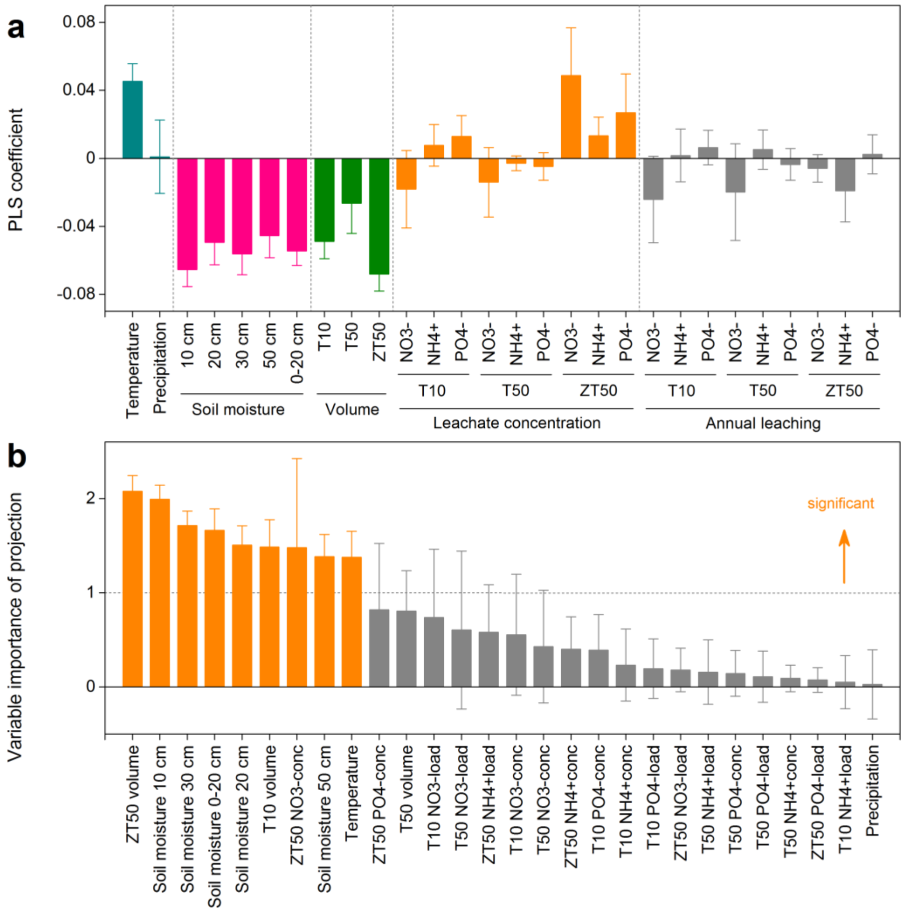


Fig. S10. Partial least squares (PLS) regression results. (*a*) PLS coefficients for 28 factors measured in Baltimore. Values show effect directions of each factor on soil CH_4 uptake. (*b*) Variable importance of each factor. Values greater than 1 (colored) are significant. T10: tension lysimeters at 10 cm soil depth, T50: tension lysimeters at 50 cm soil depth, ZT50: zero tension lysimeters at 50 cm soil depth, conc: concentration, load: annual leaching. Error bars are standard errors (*n*=1030).

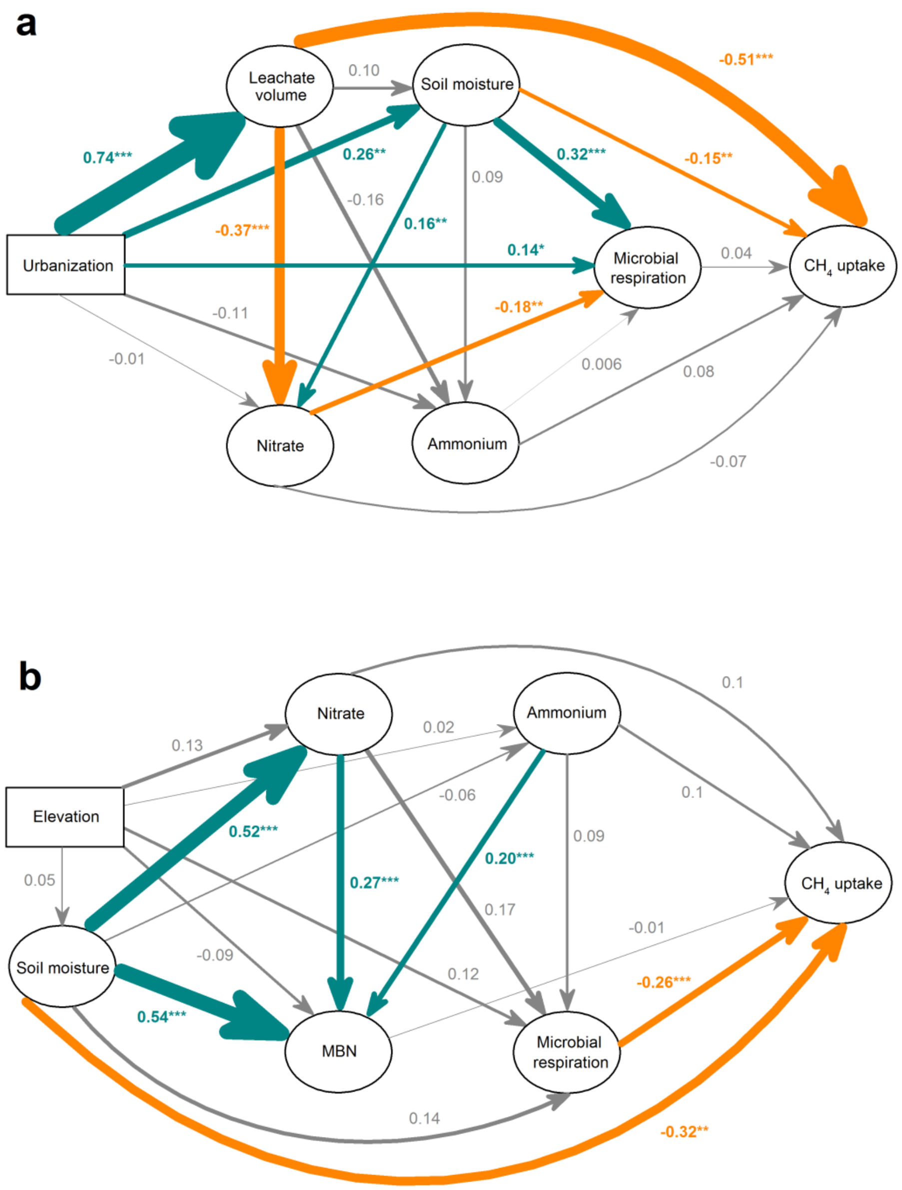


Fig. S11. Structural equation model results. (*a*) Baltimore forests. (*b*) Hubbard Brook forests. Green and orange arrows denote significant positive and negative effects, and gray arrows denote non-significant effects. Width of arrows and associated numbers (standard regression weights) represent the strength of path coefficients, and the bold coefficients with asterisks are significant ($^{*}P$<0.05, $^{**}P$<0.01 and $^{***}P$<0.001). *n*=308 in Baltimore and *n*=155 at Hubbard Brook. Soil moisture is volumetric at 10 cm soil depth in Baltimore and gravimetric at 0-10 cm in the mineral soil at Hubbard Brook. Leachate volume, nitrate and ammonium concentrations were collected from zero tension lysimeters at 50 cm depth in Baltimore. All data from Hubbard Brook were collected from 0-10 cm in the mineral soil. MBN: microbial biomass nitrogen.

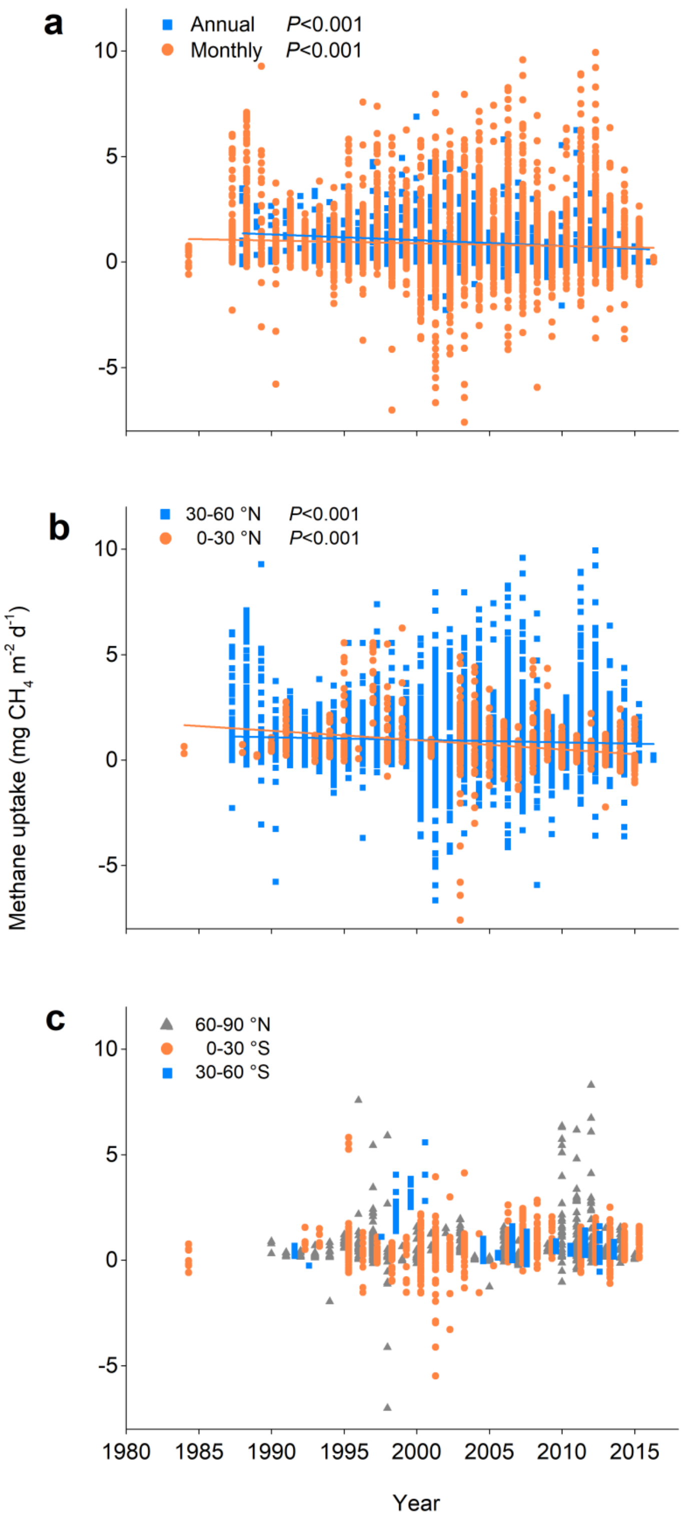
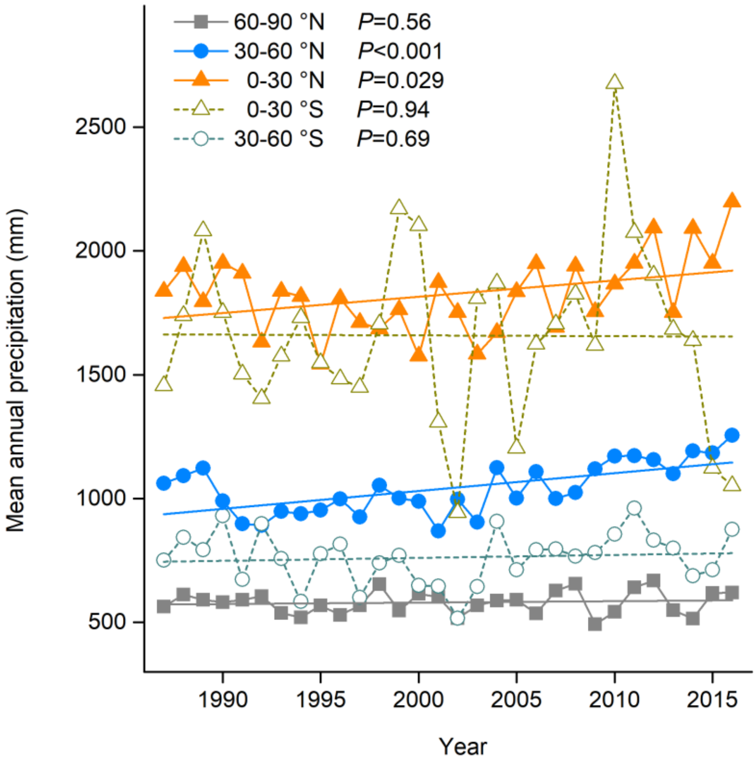


Fig. S12. Methane uptake in forest soils retrieved from published studies. (*a*) Global actually measured (1988 to 2015, *n*=756) and estimated (by using monthly uptake data, 1984 to 2016, *n*=9789) annual CH_4 uptake in forest soils. (*b*) Annual CH_4 uptake in forest soils from 0-30 °N (*n*=1558) and 30-60 °N (*n*=6887) latitude. (*b*) Annual CH_4 uptake in forest soils from 60-90 °N (*n*=759), 0-30 °S (*n*=968) and 30-60 °S (*n*=373) latitude. The data in panels (*b*) and (*c*) include both actually measured and estimated annual CH_4 uptake. The data collected at the Baltimore and Hubbard Brook sites were excluded from these analyses.



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276 **Fig. S13.** Annual precipitation over 30 °latitude bands from 1987 to 2016. Data were
277 compiled from the NOAA website (https://gis.ncdc.noaa.gov/maps/ncei/cdo/annual) and were
278 collected at or near in situ soil-atmosphere CH_4 measurement sites identified in our literature
279 review (Dataset S4). *P* values of linear regression are shown for each latitude band.

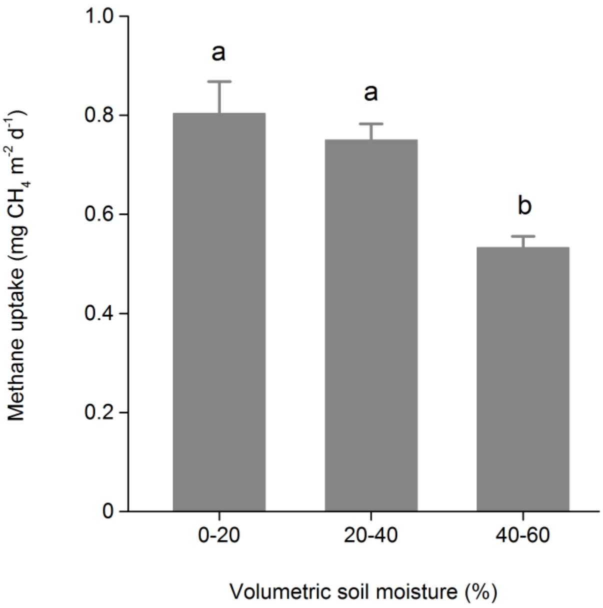

Fig. S14. Methane uptake (±SE) at different soil moisture levels. Values were compiled from the literature with *n*=1240, 1765 and 699 for 0-20%, 20-40% and 40-60% soil moisture, respectively. Different lowercase letters denote significant (P<0.05) differences among soil moisture levels.

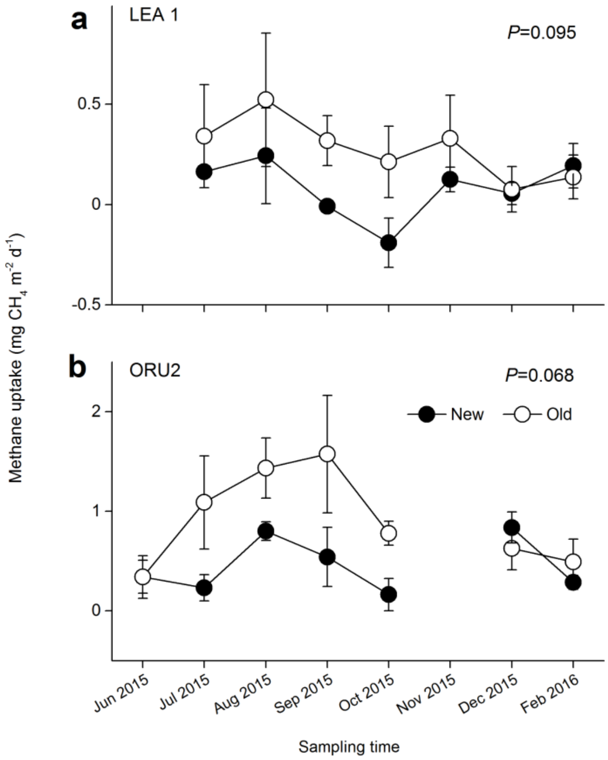

Fig. S15. Methane uptake in new and old chambers. (*a*) Leakin Park 1 site (LEA 1). (*b*) Oregon Ridge upper slope 2 site (ORU2). Values are mean ±SE. *n*=2 for new chambers and *n*=4 for old chambers. There were no significant differences between new and old chambers at both sites.

Table S1. Long-term measurement sites in urban and rural forests in Baltimore, MD, USA.

Site[1]	Land use	Latitude and Longitude	Area (m^2)	Soil classification	pH	Bulk density (g cm^{-3})	Sand (%)	Silt (%)	Clay (%)	SOM[2] (%)	NO_3^- (mg kg^{-1})	NH_4^+ (mg kg^{-1})
HD1	Urban	39°19’28” N, 76°42’16” W	900	Jackland (fine, smecitic, mesic Typic Hapludalf)	3.48	1.0	9.8	76.0	14.2	5.9	0.054	2.1
HD2	Urban	39°19’31” N, 76°42’29” W	900	Jackland (fine, smecitic, mesic Typic Hapludalf)	3.95	*n.d.*[3]	9.8	76.0	14.2	4.8	0.84	0.6
LEA1	Urban	39°18’01” N, 76°41’37” W	1600	Legore (fine-loamy, mixed, mesic, Ultic Hapludalf)	4.14	1.2	35.3	49.9	14.8	4.1	0.24	0.7
LEA2	Urban	39°18’05” N, 76°41’34” W	1600	Occaquon (loamy-skeletal, mixed, subactive Typic Dystrudept)	3.52	1.3	53.3	36.6	10.1	3.5	0.012	4.7
ORU1	Rural	39°28’51” N, 76°41’23” W	1600	Glenelg (fine-loamy, paramicaceous, mesic Typic Hapludult)	3.87	0.8	46.3	35.9	17.8	7.1	0.009	5.5
ORU2	Rural	39°29’13” N, 76°41’23” W	1600	Glenelg (fine-loamy, paramicaceous, mesic Typic Hapludult)	3.93	1.1	39.7	42.0	18.3	5.3	0.043	2.7
ORM1	Rural	39°28’51” N, 76°41’18” W	1600	Glenelg (fine-loamy, paramicaceous, mesic Typic Hapludult)	3.87	1.0	43.3	34.5	22.2	6.2	0.036	1.3
ORM2	Rural	39°29’13” N, 76°41’19” W	1600	Manor (coarse-loamy, paramicaceous, semiactive, mesic Typic Dystrochrept)	4.02	1.2	38.3	41.6	20.1	5.9	0.035	1.7

[1]HD: Hillsdale Park, LEA: Leakin Park, ORU: Oregon Ridge upper slope, ORM: Oregon Ridge middle slope.
[2]Soil organic matter.
[3]Not determined.
Data from ref. 15 and 20.

Table S2. Long-term measurement sites along an elevation gradient in Ca fertilized and reference forests at Hubbard Brook, NH, USA.

Watershed	Ca treatment	Site	Elevation (m)	Soil pH[1]	Potential N mineralization (mg N kg^{-1} soil d^{-1})	Potential N nitrification (mg N kg^{-1} soil d^{-1})	NO_3^- (mg kg^{-1})	NH_4^+ (mg kg^{-1})
Watershed 1	Ca fertilization	Low	520-560	4.9±0.037 a	0.51±0.24 b	0.77±0.21 b	2.14±0.83 b	5.28±1.06 ab
Watershed 1	Ca fertilization	Middle	600-650	4.5±0.055 b	0.86±0.26 b	1.16±0.25 b	2.54±0.70 ab	5.73±0.56 ab
Watershed 1	Ca fertilization	High	725-750	4.6±0.031 b	1.73±0.23 b	1.55±0.08 b	4.37±0.55 ab	3.67±0.37 ab
Watershed 1	Ca fertilization	Spruce-Fir	770-850	4.1±0.039 c	1.81±0.31 b	1.02±0.28 b	1.60±0.82 b	6.94±0.98 a
Watershed 6	Reference	Low	520-560	5.1±0.077 a	0.90±0.20 b	0.59±0.24 b	0.55±0.31 b	4.83±1.42 ab
Watershed 6	Reference	Middle	600-650	*n.d.*[2]	4.10±1.59 ab	3.85±1.35 ab	3.57±0.53 ab	3.88±0.96 ab
Watershed 6	Reference	High	725-750	4.6±0.046 b	6.25±1.68 a	5.57±1.49 a	5.84±1.42 a	2.52±0.89 b
Watershed 6	Reference	Spruce-Fir	770-850	4.6±0.024 b	1.22±0.42 b	0.82±0.29 b	1.63±0.81 b	3.68±0.69 ab

[1]Soil pH was measured in 1:2 soil:H_2O.

[2]Not determined.

Data were collected from 0-10 cm mineral soils in 2002. Different lowercase letters in the same column denote significant (P<0.05) difference among the eight sites.

Table S3. Results of repeated measures ANOVA testing for the effects of forest type over time.

Variables	Time		Forest		Time × Forest	
	F value	*P* value	*F* value	*P* value	*F* value	*P* value
Baltimore						
Annual CH_4 uptake	37.6	**<0.001**	327.3	**<0.001**	11.0	**<0.001**
Monthly CH_4 uptake	20.1	**<0.001**	265.1	**<0.001**	1.1	0.35
Precipitation	17.7	**<0.001**	2.0	0.16	1.4	0.18
Volumetric soil moisture	2.6	**0.0017**	0.47	0.50	0.48	0.95
Leachate volume	3.4	**<0.001**	677.3	**<0.001**	3.2	**<0.001**
Leachate NO_3^-	1.2	0.29	21.3	**<0.001**	1.2	0.31
Leachate NH_4^+	0.98	0.47	11.2	**0.0014**	0.9	0.53
Hubbard Brook						
Annual CH_4 uptake	9.2	**<0.001**	0.02	0.89	0.41	0.97
Monthly CH_4 uptake	2.8	**0.014**	0	0.99	0.16	0.99
Precipitation	79.4	**<0.001**	21.8	**<0.001**	0.59	0.84
Gravimetric soil moisture	2.1	**0.016**	0.62	0.43	1.4	0.18
NO_3^- in 0-10 mineral soil	4.9	**<0.001**	6.0	**0.017**	0.18	0.99
NH_4^+ in 0-10 mineral soil	8.1	**<0.001**	0.48	0.49	0.47	0.93
Microbial biomass N	2.6	**0.0065**	0.44	0.51	1.2	0.31
Microbial respiration	10.6	**<0.001**	5.9	**0.018**	1.9	**0.042**

Bold values are significant (P<0.05).

Table S4. Linear relationships between annual CH_4 uptake and time.

Experimental site	Equation[1]	*n*	R^2	*F* value	*P* value
Baltimore					
Urban forest	y = 91.4 - 0.045 x	18	0.46	15.4	**0.0012**
Rural forest	y = 188.3 - 0.093 x	18	0.47	15.9	**0.0011**
Hubbard Brook (from 2002 to 2015)					
Ca fertilized forest	y = 109.9 - 0.054 x	14	0.46	12.1	**0.0022**
Reference forest	y = 168.7 - 0.083 x	14	0.61	21.0	**<0.001**
Hubbard Brook (from 2003 to 2015)					
Ca fertilized forest	y = 88.0 - 0.043 x	13	0.33	6.8	**0.025**
Reference forest	y = 133.0 - 0.066 x	13	0.52	14.0	**0.0033**
Literature analysis[2]					
Global	y = 51.4 - 0.025 x	28	0.25	10.1	**0.0038**
60-90 °N	y = -43.3 + 0.022 x	25	0.063	2.6	0.12
30-60 °N	y = 48.9 - 0.023 x	30	0.23	9.9	**0.0039**
0-30 °N	y = 36.6 - 0.018 x	24	0.14	4.7	**0.041**
0-30 °S	y = -7.0 + 0.004 x	24	0	0.12	0.73
30-60 °S	y = 9.4 - 0.004 x	13	0	0.04	0.85

[1]y is annual CH_4 uptake and x is time (year).
[2]The data collected in Baltimore and Hubbard Brook were excluded in the literature analysis. The data used for analysis are presented in Fig. 1 and Fig. 2. Bold *P* values are significant (*P*<0.05).

Table S5. Results of one-way ANOVA testing for the effects of time on annual CH_4 uptake at each individual site in Baltimore and at Hubbard Brook.

Baltimore	*df*	*F* value	*P* value	Hubbard Brook	*df*	*F* value	*P* value
Urban forest				**Ca fertilized forest**			
Hillsdale Park 1	17	9.1	**<0.001**	Low elevation	13	3.0	**0.0068**
Hillsdale Park 2	4	3.9	**0.023**	Middle elevation	13	2.5	**0.019**
Leakin Park 1	17	3.6	**<0.001**	Higher elevation	13	3.1	**0.0065**
Leakin Park 2	17	4.0	**<0.001**	Spruce-Fir	13	1.4	0.23
Rural forest				**Reference forest**			
Oregon Ridge upper slope 1	17	11.1	**<0.001**	Low elevation	13	2.4	**0.028**
Oregon Ridge upper slope 2	17	9.6	**<0.001**	Middle elevation	13	3.1	**0.0058**
Oregon Ridge middle slope 1	11	10.7	**<0.001**	Higher elevation	13	7.9	**<0.001**
Oregon Ridge middle slope 2	11	3.5	**0.0019**	Spruce-Fir	13	2.6	**0.018**

n=4 in Baltimore and n=3 at Hubbard Brook.
Bold P values are significant (P<0.05).

Datasets S1 to S7 (separate files)

Dataset S1. Study periods of current global *in situ* measurements of CH_4 uptake in forest soils.
Dataset S2. Global *in situ* measurements of annual CH_4 uptake in forest soils.
Dataset S3. Global *in situ* measurements of monthly CH_4 uptake in forest soils.
Dataset S4. Mean annual precipitation at these sites from 1987 to 2016.
Dataset S5. Atmospheric CH_4 concentration.
Dataset S6. Nitrogen deposition in Baltimore and Hubbard Brook.
Dataset S7. Monthly temperature and precipitation in Baltimore.

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