TITLE: Soil moisture as the key factor of atmospheric CH_4 uptake in forest soils under environmental change

RUNNING TITLE: Atmospheric CH_4 uptake by forest soils

AUTHORS: Lei Liu[1, 2], Marc Estiarte[2, 3], Josep Peñuelas[2, 3]

1. Institute of Ecology, Jiangsu Key Laboratory of Agricultural Meteorology, Nanjing University of Information Science & Technology, Nanjing 210044, China.
2. CREAF, E08193, Cerdanyola del Vallès, Catalonia, Spain.
3. CSIC, Global Ecology Unit, CREAF-CSIC-UAB, E08193 Bellaterra, Catalonia, Spain.

Corresponding author: Lei Liu

Email: liulei406@gmail.com

KEY WORDS: CH_4 uptake; forest; Environmental change; Elevated CO_2; N enrichment; Drought; Warming

Abstract

Methane (CH_4) is an important anthropogenic greenhouse gas that can be produced and consumed by microorganisms in soils. We present a meta-analysis of the potential effects of environmental change on CH_4 uptake by forest soils. Such effects have not been reliably estimated even though aerobic methanotrophs in forest soils are the largest biological sink for atmospheric CH_4. Differences in the annual rate of CH_4 uptake between forests are likely caused by differences in vegetation, microbial communities, and the physic-chemistry of soil environments, but we found no clear different patterns at annual scale among tropical, temperate, and boreal forests. The meta-analysis indicated that the rates of CH_4 uptake in forest ecosystems were significantly decreased under elevated CO_2 and N enrichment, but the rates increased under drought. The effects of warming on the rates of CH_4 uptake were inconsistent in forest soils, and the response ratio accordingly suggested that a warmer climate would have no significant effect on the rate of CH_4 uptake. The seasonality of CH_4 uptake in natural forest soils and the clear results of the drought experiments evidence the importance of soil moisture. However, our linear model did not unravel a clear negative effect of climatic water surplus nor mean annual precipitation on soil CH_4 uptake. Therefore, process-based and ecosystem-specific models of CH_4 flux are also warranted for predicting the responses of ecosystemic CH_4 fluxes to climate change.

1. Introduction

Methane (CH_4) is an important greenhouse gas with a warming potential 25 times greater than that of CO_2 and is responsible for about 20% of the realized global warming (IPCC, 2007). Atmospheric CH_4 concentration has been increasing from the pre-industrial value of around 715 ppb to the current value of near 1800 ppb (Heimann, 2011). The global annual rate of the increase of atmospheric CH_4 caused by the imbalance between sources and sinks, decreased from an average of 3.3 ppb y^{-1} in the 1980s to 1.3 ppb y^{-1} in the 2000s, although the rate began to increase again in 2007 (Kirschke et al., 2013). The decadal and inter-annual variation of the rate of increase is not yet fully understood. Two alternative causes have been suggested to explain the reduction over the last three decades (Heimann, 2011): one suggests a decrease in fossil-fuel emissions (Aydin et al., 2011), and the other suggests a decrease in emissions from rice cultivation in Asia due to higher fertilizer application and reduction in water use (Kai et al., 2011).

Most of the atmospheric CH_4 is oxidized in the troposphere by chemical reactions with hydroxyl radicals (OH), which comprises approximately 90% of the global sinks (Schlesinger and Bernhardt, 2013). The second largest global sink of atmospheric CH_4 is the consumption by aerated soils. The amount of CH_4 oxidized by soil methane-oxidizing bacteria (MOB) was estimated to be between 26 and 42 Tg y^{-1} for 2000-2009 (Kirschke et al., 2013). Forest soils represent approximately 50% of this sink and coarse forest soils have the highest rates of CH_4 uptake (Dutaur and Verchot, 2007).

The main factor regulating the CH_4 uptake capacity of soils is the diffusion rate of gases

that regulates the availability of CH_4 to MOB across the soil profile. A number of soil characteristics, such as texture, structure, moisture content, temperature, and mineral-nitrogen (N) content, are important to CH_4 uptake. The diffusion of gases in soils structurally depends on the soil texture and degree of compaction (Castaldi and Fierro, 2005) because they affect either the diffusion of gas through the soil or the activity and size of the soil microbial populations involved in CH_4 metabolism (Castaldi and Fierro, 2005; King, 1997; Lin et al., 2015; Price et al., 2003; Verchot et al., 2000). Although the relationship between CH_4 uptake and soil moisture was identified decades ago (Bowden, 1998), uncertainty remains as to how future climate change will affect the CH_4 uptake in soils of forest ecosystems and their role in the global CH_4 cycle.

Several components of environmental change, such as altered precipitation, warming, rising atmospheric CO_2 concentration and increased atmospheric N deposition, can potentially alter the soil properties or biology that control the uptake of CH_4 in forest soils and determine the size of their atmospheric CH_4 sink (Blankinship et al., 2010). The effect of environmental change on CH_4 uptake by forest soils has not been reliably estimated, although forests have been responsible for most of the terrestrial uptake of atmospheric CH_4. We reviewed the literature with three main aims: (1) to synthesize the information available on the rates of CH_4 uptake in forest soils in different biomes, (2) to evaluate the response of CH_4 uptake to components of environmental change and to summarize the current state of our understanding of the mechanisms underlying the responses, and (3) to test for across-sites controls by climate and water balance in CH_4 uptake in forest soils. We also identify topics requiring further study.

2. Materials and methods

We gathered data on CH_4 uptake rates in the forests and under different treatments to thereafter conduct a meta-analysis and accomplish the first two aims. For the third aim we also gathered data on climate and soil moisture from world databases and used them to model their relations with the CH_4 uptake by forest soils.

2.1. Data source.

We systematically searched all peer-reviewed journal articles and those that investigated CH_4 uptake in forest ecosystems. The literature search was done through the Web of Science seeking for the keywords "methane or CH_4" and "forest". We reviewed all the found articles and selected those that met the following two criterias: (1) the study was conducted in situ field measurements for several months; (2) the CH_4 uptake could be extracted directly from the texts, tables, and figures. When several publications include data from the same locations we obtained the data as annual average. When one publication includes several experiments under different abiotic conditions, such as different locations, tree species, or stand ages, we considered them different observations. In total we found 134 datasets for forest soils at 134 sites from 110 papers in 26 countries (Table S1). Most of the studies used static chambers, but some used stable isotopes. The mean annual CH_4 uptake rates published in the primary or secondary literature were used when provided and when annual averages were not provided they were calculated based on figures. We used Plot Digitizer version online to digitally extract data from figures when the results were graphically reported. CH_4

uptake rates were standardized to kg CH_4 ha^{-1} y^{-1}. The study sites comprise tropical forests, coniferous-, deciduous-, or mixed-temperate forests and boreal forests (Fig. 1). For each site identified by the latitude and longitude coordinates, we extracted monthly average temperature and precipitation from the WorldClim database (http://www.worldclim.org/) with a spatial resolution of around 1 km at the equator, and aggregated them to mean annual temperature (MAT) and mean annual precipitation (MAP). Sites were climatically characterized by MAT, MAP, temperature and precipitation during the summer month (Tsum and Psum) and potential evapotranspiration (PET). We defined summer month as July in the Northern hemisphere and January in the Southern hemisphere. PET was estimated on a monthly basis from monthly average temperature and latitude with the function "thornthwaite" from the R package:*spei*.

In addition to CH_4 uptake, we estimated soil water-holding capacity (SWHC) from soil moisture data obtained from the Soil Moisture and Ocean Salinity (SMOS) database (https://smos-ds-02.eo.esa.int/oads/access/). For every site and from June 2010 to June 2017, we gathered the products that comprise soil moisture measurements geo-located in an equal-area grid system ISEA 4H9 as percent of water-filled soil volume in "SMOS level 1 and 2 Science data" collection and with a spatial resolution in the range of 30-50 km. We assumed that during the period 2010-2017 maximum and minimum soil moisture levels were reached and we proceeded to obtain maximum and minimum soil water content after removing the 5% extremely low or high values. For each site, we calculated SWHC as maximum minus minimum soil water content and extended it as

the soil was 50 cm deep. In total, SWHC was available for 124 sites. Afterwards, soil moisture data from satellite was standardized to the site amplitude in the historical record and expressed as a percent of the estimated site SWHC which we coined as standardized soil moisture (SSM). The mean annual SSM (MASSM), summer SSM (SSMsum) and winter SSM (SSMwin) were also included in the database. We defined summer month as July or January and winter month as January or July respectively for the Northern or the Southern hemisphere.

For each site, we used Stephenson's bucket (1990) approach to adjust a climatic water balance using average monthly values of PET, precipitation and site SWHC to estimate monthly values of actual evapotranspiration (AET), water deficit (WD), water surplus (WS) and mean annual soil water content (SWC) (details for the estimation of water balance variables are in the Appendix.). Water balance variables indicate how much energy and water are available at the same time (AET), how much evaporative demand is not met by available water (WD) and how much water is unusable surplus (WS) (Stephenson, 1990).

2.2 Meta-analysis.

To examine the effects of environmental-change components on the CH_4 uptake rate in forest ecosystems, among the 134 datasets gathered previously, we selected the datasets including experimental treatments simulating the following components of the environmental change: elevated atmospheric CO_2, warming, drought or water addition, and N enrichment. To increase the number of datasets, we also included studies with a reduced number of field measurements as well as data from laboratory incubations with

149 warming treatment or with N addition. Details of the studies are summarized in
150 supplementary Table S2. We calculated the response ratios from each study as described
151 by Hedges *et al.* (1999). Briefly, the natural-log response ratio (ln*RR*) was calculated
152 as:

153 $\ln(Xi/Xn) = \ln Xi - \ln Xn$

154 where *Xi* and *Xn* are the values of each observation in the treatment and corresponding
155 control plots, respectively. The sampling variance for each ln*RR* was calculated as:

156 $$\ln[(1/n_i) \times (Si/Xi)^2 + (1/n_n) \times (S_n/X_n)^2]$$

157 where n_i and n_n, S_i and S_n, and X_i, and X_n are the treatment and control sample sizes,
158 standard deviations, and mean responses, respectively. The natural-log response ratios
159 (from here onwards simplified as response ratios) were determined by specifying
160 studies as random factors using the *rma* model in the R metafor package. The effects
161 on CH_4 uptake rates and the differences between the treatment and control plots were
162 considered significant if the 95% confidence interval (CI) of ln*RR* did not overlap zero.
163 All statistical analyses were performed in RStudio 3.1.2 (R Core Team 2015) using the
164 R package metafor 1.9–2.

165 *2.3 Linear modelling of CH_4 uptake*

166 We built linear models to explain forest annual CH_4 uptake at site level from climatic
167 variables, standardized soil moisture variables and derived water balance variables.
168 Water balance variables are a lineal combination of MAP and PET (e.g PET =
169 AET+WD; MAP=AET+WS), so MAP and PET were excluded when water balance
170 variables were allowed to enter the models.

171 Model selection was performed using procedures based on AIC implemented in the
172 *Mumin* package in R environment Partial residual plots of the models were obtained
173 using the *visreg* package to evaluate the effect of each variable on the model.

3. Results

3.1 CH_4 uptake in forest soils

The mean uptake rate for all studies was 4 kg CH_4 ha^{-1} y^{-1}, with bootstrapped 95% CIs between 1.77 and 5.85. Ninety percent of a total of 134 observations were between 0.119 and 15.82 kg CH_4 ha^{-1} y^{-1}. The highest reported uptake rate was 40.52 kg CH4 ha^{-1} y^{-1} in a tropical forest in India. The lowest reported rate was -120 kg CH_4 ha^{-1} y^{-1} in a boreal forest, but this was an unusually low value. Only five of the observations were net CH_4 sources, representing 3.7% of all observations, whereas the remaining 129 observations were net sinks, with 13.4% of the values between 0 and 1 kg CH_4 ha^{-1} y^{-1} and 81.3% above 1 kg CH_4 ha^{-1} y^{-1}. No clear differences among forest types were detected. Mean and bootstrapped 95% CIs were 4.08 (2.42, 6.89), 5.1 (3.96, 6.34), and -2.74 (-21.5, 8.16) for tropical, temperate, and boreal forests, respectively, with the low rates of the boreal forests driven by a single site with an extremely low rate, without which the mean increased to 5.64 (2.61, 9.22). Median rates were 2.82, 3.32, and 2.62 kg CH_4 ha^{-1} y^{-1} for tropical, temperate, and boreal forests respectively (Fig. 1).

3.2 Soil CH_4 uptake across sites

After removing two sites with the lowest and the highest CH_4 uptake, the database used for the modelling included 132 sites and included climatic data for all of them. The best linear model fitted to the soil CH_4 uptake in the 132 sites and including only climate as explanatory variables was MAT*MAP (adj R^2= 0.10). There was almost no variance explained (adj R^2 = 0.02) if the interaction was not included.

We used the subset of 107 sites having SMOS data for all the twelve months to include the SMOS derived soil moisture. (Table S3). The addition of SMOS derived soil moisture variables (MASSM, SSMsum SSMwin) did not change the model selection and the MAP*MAT model behaved similarly with the subset including 107 sites (adj R^2= 0.09). When water balance variables substituted MAP and PET the best model was MAT*WS (adj. R^2 = 0.12). Including Tsum yielded a model with lower AICc (and adj R^2 = 0.14) but Tsum was discarded because it was non-significant after model averaging. Partial residual plots (Fig. 2) show the interaction effect of MAT and WS on soil CH_4 uptake. Higher WS occurs only at the warmer sites of the dataset and low and mild WS occurred at forests within the three temperature intervals. Soil CH_4 uptake did not respond to WS across the warmer sites, which presented uptake values in the low range. In the milder and colder sites CH_4 uptake tended to increase from low to mild WS sites. A slight effect of MAT was present only at the wetter sites.

3.3 Alteration of atmospheric CH_4 uptake in forest soils by environmental-change components

The meta-analysis of 10 experiments indicated that the CH_4 uptake by soils in forest ecosystems significantly increased an average of 150% under drought conditions. In contrast, the meta-analysis of 7 experiments showed that elevated CO_2 significantly decreased an average of 33% the CH_4 uptake in temperate forests, the only forest type where data on CH_4 uptake on CO_2-enrichment experiments have been reported. Similarly, the meta-analysis of 29 experiments showed that N-enrichment significantly

218 decreased CH_4 uptake rates an average of 36% (Fig. 3). Our meta-analysis of 6
219 experiments did not identify a clear effect of ecosystem warming on CH_4 uptake by
220 forest soils (Fig. 3).
221

4. Discussion

Previous work with a dataset including all terrestrial ecosystems found that MAP and MAT explained about 3% and 2% of the global CH_4 uptake variation (Dutaur and Verchot 2007). Our modelling for forest ecosystems explained a bit more of the variance but neither showed a strong climatic control across forests in spite of differences in vegetation. Different tree species likely produce litter of different quality and soils differing in organic matter content and in chemical, physical and biological characteristics that are important for CH_4 uptake (Barrena et al., 2013; Borken and Beese, 2006). Lower rates of CH_4 uptake were found in coniferous than in deciduous forest soils (Borken et al., 2003), suggesting the occurrence of differences among broad forest types. Differences in CH_4 uptake are also related to differences in the structure and function of the soil microbial community (Aronson et al., 2013; Nazaries et al., 2011) and communities may differ among forests. For instance, lower diversity and abundance of methane-oxidizing bacteria in spruce than in beech forest soils (Degelmann et al. 2010) suggests that tree species may influence the activity of soil methane-oxidizers. Nevertheless, the lack of clear differences in CH_4 uptake among forest types indicates low climatic control.

Beyond climate, at the biome scale soil-textural class is an important determinant of soil CH_4 fluxes (Verchot et al., 2000), with coarse and medium-textured soils consuming more CH_4 than fine-textured soils (Dutaur and Verchot, 2007). Soil texture is a local characteristic and the proportion of mineral particles is not going to be altered by environmental change. Soil aeration depends on the soil water content that fills the

networks of small pores and impedes the transport of gases (Hartmann et al., 2011; Hiltbrunner et al., 2012) and is most likely going to be altered by climate change. Similarly soil temperature and nutrient content will also be altered. Actually, moisture and temperature are temporally variable under natural conditions and are underlying the seasonality in CH_4 uptake by forest soils. For example, some tropical forests shift between sources in wet seasons to sinks in dry seasons or increase the sink strength from wet to dry seasons (Teh et al., 2014). Temperate and boreal forests may also release CH_4 under wet or water-saturated conditions (Gundersen et al., 2012). Similarly, differences in CH_4 uptake among years in the same forest are also dependent on differences in precipitation (Matson et al., 2009).

The meta-analysis of drought experiments confirmed short term effects of moisture on soil CH_4 uptake. Increases in CH_4 uptake under experimental *drought* have been described in tropical, temperate and boreal forests (Billings et al., 2000; Borken et al., 2006; Davidson et al., 2008, 2004). Most effects of drought are likely due to improved diffusion of atmospheric gases into the soil, although very low moisture can have direct effects on microbial activity. A lower diffusion alters the CH_4 source-sink balance of soils because reduces the supply of atmospheric CH_4 into the soil leading to substrate limitation of the activity of methanotrophic organisms (Blankinship et al., 2010), and reduces the supply of O_2, which may increase methanogenesis (Borken et al., 2006). When soil moisture is too low, gases diffuse without restriction but microbes are physiologically stressed and its activity, including methanotrophs, is reduced, which reduces the CH_4 uptake (Price et al., 2004). The stimulation of methanotrophic activity

266 after rains in deserts and semiarid regions provides evidence of the stress imposed on
267 methanotrophs in very dry soils (McLain and Martens, 2005). Therefore, the sensitivity
268 of soil CH_4 uptake to soil moisture can be described by a parabolic curve reflecting that
269 CH_4 uptake is limited at very low moisture by biological activity and is limited by CH_4
270 diffusion at high moisture (Fest et al., 2017).
271 The seasonality of CH_4 uptake in forest soils and the clear results of the drought
272 experiments evidence the importance of soil moisture. Recent studies show that a
273 decline in CH_4 uptake at a global scale coincides with increases in precipitation in forest
274 soils (Ni and Groffman, 2018; Yu et al., 2017). Accordingly, we expected wetter soils,
275 less aeration and lower CH_4 uptake at high MAP and, specially, at high climatic WS
276 (note that MAP and WS are highly correlated). However, satellite-derived soil moisture
277 and climatic water balance variables did not increase much the explanation of the
278 variance in CH_4 uptake. The modelling did not unravel a clear negative effect of
279 climatic WS nor MAP on CH_4 uptake. Certainly, the uncertainties introduced by the
280 type of data used could partly account for the poor fit. The spatial resolution of the
281 satellite data in the range of 30-50 km can introduce large errors because soil
282 characteristics can vary at a much smaller scale. Moreover, precipitation may also vary
283 at relatively small scales.
284 The possible effects of the other environmental change components addressed by our
285 meta-analysis on soil CH_4 uptake were less evident than for drought. Meta-analysis did
286 not reveal *warming* effects on soil CH_4 uptake, although the low number of reviewed
287 experiments prevents considering it a definitive result. We must keep in mind that

288 warming in the field often leads not only to warmer but also to drier soils (Luo et al.,
289 2013), which may increase gas diffusion and CH_4 uptake rates (Price et al., 2003).
290 Warming may also accelerate soil N mineralization, leading to higher NH_4^+
291 concentrations that might suppress CH_4 uptake (Karbin et al., 2015). The opposite
292 effects of improved gas diffusion and of increased soil NH_4^+ (Lüke and Frenzel, 2011)
293 may explain that warming in field experiments has shown scarce effects on CH_4 uptake
294 (Karbin et al., 2015; Price et al., 2004).
295 The meta-analysis provided more clear evidence of the effects of *elevated atmospheric*
296 *CO_2* despite the number of experiments was as low as for warming. Dijkstra et al. (2012)
297 reported that elevated CO_2 tended to increase CH_4 emissions in wetlands, peat lands,
298 and rice paddy fields, but the effects were highly variable in upland soils. We found an
299 average of 33% decrease (Fig. 3) in the CH_4 uptake by soils of temperate forests, the
300 only forest type where CO_2-enrichment experiments have been reported. The decrease
301 was very clear at some sites (Dubbs and Whalen, 2010; Phillips et al., 2001) and was
302 associated to increases in soil moisture. The lower CH_4 uptake could be due to a reduced
303 CH_4 diffusion into moister soils, although increases in CH_4 production by methanogens
304 were also likely (Dubbs and Whalen, 2010; Phillips et al., 2001). The reduction in CH_4
305 uptake under elevated CO_2 is relevant because 20% of the world's forests are temperate
306 (Pan et al., 2013). However, tropical forests represent 51% and boreal forests 29% of
307 the world's forests so research on the response of their soil CH_4 uptake to atmospheric
308 CO_2 increase is warranted.
309 *Anthropogenic N enrichment* has a suite of detrimental effects on ecosystem services

and the meta-analysis confirmed the decrease in CH_4 uptake rates. Some experiments reported a very clear inhibitory effect of the fertilization with N on CH_4 uptake (Adamsen and King, 1993; Fender et al., 2012; Steinkamp et al., 2001; Wang and Ineson, 2003) although a synthesis of studies in non-wetland ecosystems reported dose-dependent effects, with smaller N enrichments tending to stimulate soil CH_4 uptake and larger ones tending to inhibit it (Aronson and Helliker, 2010). These opposite N dose-dependent effects suggest that the historical N status of soils is the most important predictor of the response of CH_4 uptake to future N inputs (Aronson and Helliker, 2010). Increase in soil NH_4^+ may reduce soil CH_4 uptake because NH_4^+ competes with CH_4 at the reaction site of the enzyme methane monooxygenase, the first step of the CH_4 oxidation pathway (Bodelier and Laanbroek, 2004). The non-competitive inhibition of CH_4 uptake by NO_3^- have been attributed to the increase in NH_4^+ concentrations caused by NO_3^- (Fender et al., 2012). Furthermore, nitrate (NO_3^-) has also been reported to have direct inhibitory effects on CH_4 uptake (Fender et al., 2012; Mochizuki et al., 2012; Wang and Ineson, 2003).

We reviewed individual components of global environmental change, but it is still unknown whether the effects of multiple components are additive, cancel each other, or synergistically increase the individual effects. Ambus and Robertson (1999) reported that elevated CO_2 reduced CH_4 uptake only when no N was deposited. Another study found no interaction between altered precipitation and warming in four ecosystems along a 50-km climatic gradient from warm and dry to cold and wet, although the authors suggested that a wet and warm climate would cause the largest reduction in

terrestrial CH_4 uptake (Blankinship et al., 2010). The lack of interactive effects from these experiments may be due to inadequate statistical power or because the time required by the interactive effects to appear was longer than the duration of the experiments (Norby and Luo, 2004). Longer field experiments are therefore needed to unravel the interactive effects of climate change on soil CH_4 fluxes.

Moreover, modeling complements field experiments, thus overcoming the difficulties associated with long-term studies and identifying important interactive effects among multiple factors of climate change on ecosystemic processes (Luo et al., 2008). For example, the Dynamic Land Ecosystem Model, a process-based model, unraveled that the interaction between environmental-change components (including climatic variability, N deposition, elevated levels of atmospheric CO_2 and application of N fertilizer) led to a decrease in CH_4 uptake over the last three decades in North America (Xu et al., 2010).

The drought experiments show the key position of soil moisture in the short-term control of CH_4 uptake in forest soils. The weak control of climatic (i.e. long term) precipitation on soil CH_4 uptake across sites contrasts notoriously with the clear short-term effects of experimental reduction in precipitation. It suggests that despite the immediate effects of weather, the CH_4 consumption is determined in the long term, after texture is considered, by characteristics derived from vegetation, soil nutrients and microbial communities that are weakly controlled by climate but strongly by local conditions. The importance of the structural properties of ecosystems on soil CH_4 uptake is evidenced when changing the tree-species or after transformation of forests

354 into grasslands (Hiltbrunner et al., 2012; Smith et al., 2000). Despite the absence of
355 evidences of warming effects, we cannot discard alterations in the CH_4 uptake if
356 warming causes structural changes in the long term. The possible long term effects
357 mediated by soil and vegetation changes challenge the modelling and the prediction of
358 CH_4 consumption and an effort is needed to disentangle which are the vegetation-
359 dependant soil traits governing CH_4 uptake rates.

360 **5. Conclusions and outlook**

361 Our in-depth review of the effects of particular environmental-change components on
362 CH_4 uptake in forest soils reveals that the rates were significantly reduced by elevated
363 CO_2 and N enrichment, that were increased by drought and that were not consistently
364 altered by warming. Very few studies, however, have analyzed the interactive effects of
365 multiple environmental-change components due to the challenge of the complexity of
366 forests. More field experiments are, therefore, required to expand our knowledge of the
367 impacts of these multiple factors on CH_4 uptake and CH_4-cycling microbial
368 communities. The poor relation between climatic variables and CH_4 uptake unveiled by
369 linear modelling may be influenced by uncertainties of the data sources, but it also
370 indicates that process-based and ecosystem-specific models of CH_4 flux are necessary
371 for predicting the response of ecosystemic CH_4 fluxes to climate change.

372 The studies on CH_4 uptake in natural soils have ignored the environmental importance
373 of phosphorus, which is becoming unbalanced due to increased C and N availabilities
374 (Peñuelas et al., 2013). It is urgent to incorporate P on the agenda to provide data on its
375 possible significance for the CH_4 uptake and for the abundance, activity and structure

376 of methanotrophic communities. Attention should also be paid to the indirect effect of
377 climate change on plant communities and the decomposition of litterfall in forest
378 ecosystems because potential changes can directly affect factors important for CH_4
379 uptake in forest ecosystems such as moisture content, temperature and gas diffusion in
380 soils.

Acknowledgements

This research was supported by the international postdoctoral exchange fellowship program by the office of China Postdoctoral Council (No. 20140051) and by the European Research Council Synergy grant ERC-2013-SyG610028-IMBALANCE-P, the Spanish Government project CGL2016-79835-P, and the Catalan Government grant SGR 2017-1005

388 **References**

389 Adamsen, A.P.S., King, G.M., 1993. Methane consumption in temperate and subarctic
390 forest soils: Rates, vertical zonation, and responses to water and nitrogen. Applied
391 and Environmental Microbiology 59, 485–490.

392 Ambus, P., Robertson, G.P., 1999. Fluxes of CH_4 and N_2O in aspen stands grown under
393 amnient and twice-ambient CO_2. Plant and soil 209, 1–8.
394 https://doi.org/10.1016/j.agee.2011.12.016

395 Aronson, A.E.L., Helliker, B.R., 2010. Methane flux in non-wetland soils in response
396 to nitrogen addition : a meta-analysis Linked references are available on JSTOR
397 for this article : Methane flux in non-wetland soils in response to nitrogen addition :
398 a meta-analysis. Ecology 91, 3242–3251. https://doi.org/10.2307/20788157

399 Aronson, E.L., Allison, S.D., Helliker, B.R., 2013. Environmental impacts on the
400 diversity of methane-cycling microbes and their resultant function. Frontiers in
401 Microbiology 4, 1–15. https://doi.org/10.3389/fmicb.2013.00225

402 Aydin, M., Verhulst, K.R., Saltzman, E.S., Battle, M.O., Montzka, S.A., Blake, D.R.,
403 Tang, Q., Prather, M.J., 2011. Recent decreases in fossil-fuel emissions of ethane
404 and methane derived from firn air. Nature 476, 198–201.
405 https://doi.org/10.1038/nature10352

406 Barrena, I., Menéndez, S., Duñabeitia, M., Merino, P., Florian Stange, C., Spott, O.,
407 González-Murua, C., Estavillo, J.M., 2013. Greenhouse gas fluxes (CO_2, N_2O and
408 CH_4) from forest soils in the Basque Country: Comparison of different tree species
409 and growth stages. Forest Ecology and Management 310, 600–611.

https://doi.org/10.1016/j.foreco.2013.08.065

Billings, S.A., Richter, D.D., Yarie, J., 2000. Sensitivity of soil methane fluxes to reduced precipitation in boreal forest soils. Soil Biology & Biochemistry 32, 1431–1441.

Blankinship, J.C., Brown, J.R., Dijkstra, P., Allwright, M.C., Hungate, B.A., 2010. Response of Terrestrial CH_4 Uptake to Interactive Changes in Precipitation and Temperature Along a Climatic Gradient. Ecosystems 13, 1157–1170. https://doi.org/10.1007/s10021-010-9391-9

Bodelier, P.L.E., Laanbroek, H.J., 2004. Nitrogen as a regulatory factor of methane oxidation in soils and sediments. FEMS Microbiology Ecology 47, 265–277. https://doi.org/10.1016/S0168-6496(03)00304-0

Borken, W., Beese, F., 2006. Methane and nitrous oxide fluxes of soils in pure and mixed stands of European beech and Norway spruce. European Journal of Soil Science 57, 617–625. https://doi.org/10.1111/j.1365-2389.2006.00752.x

Borken, W., Davidson, E.A., Savage, K., Sundquist, E.T., Steudler, P., 2006. Effect of summer throughfall exclusion, summer drought, and winter snow cover on methane fluxes in a temperate forest soil. Soil Biology and Biochemistry 38, 1388–1395. https://doi.org/10.1016/j.soilbio.2005.10.011

Borken, W., Xu, Y.J., Beese, F., 2003. Conversion of hardwood forests to spruce and pine plantations strongly reduced soil methane sink in Germany. Global Change Biology 9, 956–966. https://doi.org/10.1046/j.1365-2486.2003.00631.x

Bowden, K.M.N. and G.M.R., 1998. Carbon dioxide and methane fluxes by a forest

soil under laboratory-controlled moisture and temperature conditions. Soil Biology & Biochemistry 30, 1591–1597.

Castaldi, S., Fierro, A., 2005. Soil-atmosphere methane exchange in undisturbed and burned Mediterranean shrubland of southern Italy. Ecosystems 8, 182–190. https://doi.org/10.1007/s10021-004-0093-z

Davidson, E.A., Ishida, F.Y., Nepstad, D.C., 2004. Effects of an experimental drought on soil emissions of carbon dioxide, methane, nitrous oxide, and nitric oxide in a moist tropical forest. Global Change Biology 10, 718–730. https://doi.org/10.1111/j.1529-8817.2003.00762.x

Davidson, E.A., Nepstad, D.C., Ishida, F.Y., Brando, P.M., 2008. Effects of an experimental drought and recovery on soil emissions of carbon dioxide, methane, nitrous oxide, and nitric oxide in a moist tropical forest. Global Change Biology 14, 2582–2590. https://doi.org/10.1111/j.1365-2486.2008.01694.x

Dijkstra, F.A., Prior, S.A., Runion, G.B., Torbert, H.A., Tian, H., Lu, C., Venterea, R.T., 2012. Effects of elevated carbon dioxide and increased temperature on methane and nitrous oxide fluxes: Evidence from field experiments. Frontiers in Ecology and the Environment 10, 520–527. https://doi.org/10.1890/120059

Dubbs, L.L., Whalen, S.C., 2010. Reduced net atmospheric CH_4 consumption is a sustained response to elevated CO_2 in a temperate forest. Biology and Fertility of Soils 46, 597–606. https://doi.org/10.1007/s00374-010-0467-7

Dutaur, L., Verchot, L. V., 2007. A global inventory of the soil CH_4 sink. Global Biogeochemical Cycles 21, 1–9. https://doi.org/10.1029/2006GB002734

Fender, A.C., Pfeiffer, B., Gansert, D., Leuschner, C., Daniel, R., Jungkunst, H.F., 2012. The inhibiting effect of nitrate fertilisation on methane uptake of a temperate forest soil is influenced by labile carbon. Biology and Fertility of Soils 48, 621–631. https://doi.org/10.1007/s00374-011-0660-3

Fest, B., Hinko-Najera, N., von Fischer, J.C., Livesley, S.J., Arndt, S.K., 2017. Soil Methane Uptake Increases under Continuous Throughfall Reduction in a Temperate Evergreen, Broadleaved Eucalypt Forest. Ecosystems 20, 368–379. https://doi.org/10.1007/s10021-016-0030-y

Gundersen, P., Christiansen, J.R., Alberti, G., Brüggemann, N., Castaldi, S., Gasche, R., Kitzler, B., Klemedtsson, L., Lobo-Do-Vale, R., Moldan, F., Rütting, T., Schleppi, P., Weslien, P., Zechmeister-Boltenstern, S., 2012. The response of methane and nitrous oxide fluxes to forest change in Europe. Biogeosciences 9, 3999–4012. https://doi.org/10.5194/bg-9-3999-2012

Hartmann, A.A., Buchmann, N., Niklaus, P.A., 2011. A study of soil methane sink regulation in two grasslands exposed to drought and N fertilization. Plant and Soil 342, 265–275. https://doi.org/10.1007/s11104-010-0690-x

Heimann, M., 2011. Enigma of the recent methane budget Bespoke cells for the human brain. Nature 476, 157–158.

Hiltbrunner, D., Zimmermann, S., Karbin, S., Hagedorn, F., Niklaus, P.A., 2012. Increasing soil methane sink along a 120-year afforestation chronosequence is driven by soil moisture. Global Change Biology 18, 3664–3671. https://doi.org/10.1111/j.1365-2486.2012.02798.x

476 Intergovernmental Panel on Climate Change (IPCC), 2007. Climate change 2007: the
477 physical science basis, Contribution of Working Group I to the Fourth Assessment
478 Report of the Intergovernmental Panel on Climate Change. Published for the
479 Intergovernmental Panel on Climate Change.
480 https://doi.org/10.1260/095830507781076194
481 Kai, F.M., Tyler, S.C., Randerson, J.T., Blake, D.R., 2011. Reduced methane growth
482 rate explained by decreased Northern Hemisphere microbial sources. Nature 476,
483 194–197. https://doi.org/10.1038/nature10259
484 Karbin, S., Hagedorn, F., Dawes, M.A., Niklaus, P.A., 2015. Treeline soil warming does
485 not affect soil methane fluxes and the spatial micro-distribution of methanotrophic
486 bacteria. Soil Biology and Biochemistry 86, 164–171.
487 https://doi.org/10.1016/j.soilbio.2015.03.022
488 King, G.M., 1997. Responses of atmospheric methane consumption by soils to global
489 climate change. Global Change Biology 3, 351–362.
490 https://doi.org/doi:10.1046/j.1365-2486.1997.00090.x
491 Kirschke, S., Bousquet, P., Ciais, P., Saunois, M., Canadell, J.G., Dlugokencky, E.J.,
492 Bergamaschi, P., Bergmann, D., Blake, D.R., Bruhwiler, L., Cameron-Smith, P.,
493 Castaldi, S., Chevallier, F., Feng, L., Fraser, A., Heimann, M., Hodson, E.L.,
494 Houweling, S., Josse, B., Fraser, P.J., Krummel, P.B., Lamarque, J.F., Langenfelds,
495 R.L., Le Quéré, C., Naik, V., O'doherty, S., Palmer, P.I., Pison, I., Plummer, D.,
496 Poulter, B., Prinn, R.G., Rigby, M., Ringeval, B., Santini, M., Schmidt, M.,
497 Shindell, D.T., Simpson, I.J., Spahni, R., Steele, L.P., Strode, S.A., Sudo, K.,

498 Szopa, S., Van Der Werf, G.R., Voulgarakis, A., Van Weele, M., Weiss, R.F.,
499 Williams, J.E., Zeng, G., 2013. Three decades of global methane sources and sinks.
500 Nature Geoscience 6, 813–823. https://doi.org/10.1038/ngeo1955
501 Lin, X., Wang, S., Hu, Y., Luo, C., Zhang, Z., Niu, H., Xie, Z., 2015. Experimental
502 Warming Increases Seasonal Methane Uptake in an Alpine Meadow on the Tibetan
503 Plateau. Ecosystems 18, 274–286. https://doi.org/10.1007/s10021-014-9828-7
504 Lüke, C., Frenzel, P., 2011. Potential of pmoA amplicon pyrosequencing for
505 methanotroph diversity studies. Applied and Environmental Microbiology 77,
506 6305–6309. https://doi.org/10.1128/AEM.05355-11
507 Luo, G.J., Kiese, R., Wolf, B., Butterbach-Bahl, K., 2013. Effects of soil temperature
508 and moisture on methane uptake and nitrous oxide emissions across three different
509 ecosystem types. Biogeosciences 10, 3205–3219. https://doi.org/10.5194/bg-10-
510 3205-2013
511 Luo, Y., Gerten, D., Le Maire, G., Parton, W.J., WENG, E., ZHOU, X., KEOUGH, C.,
512 BEIER, C., CIAIS, P., CRAMER, W., DUKES, J.S., EMMETT, B., HANSON,
513 P.J., KNAPP, A., LINDER, S., NEPSTAD, D., RUSTAD, L., 2008. Modeled
514 interactive effects of precipitation, temperature, and CO2 on ecosystem carbon and
515 water dynamics in different climatic zones. Global Change Biology 14, 1986–
516 1999. https://doi.org/10.1111/j.1365-2486.2008.01629.x
517 Matson, A., Pennock, D., Bedard-Haughn, A., 2009. Methane and nitrous oxide
518 emissions from mature forest stands in the boreal forest, Saskatchewan, Canada.
519 Forest Ecology and Management 258, 1073–1083.

https://doi.org/10.1016/j.foreco.2009.05.034

McLain, J.E.T., Martens, D. a, 2005. Studies of methane fluxes reveal that desert soils can mitigate global climate change. Connecting mountain islands and desert seas: biodiversity and management of the Madrean Archipelago II 496–499.

Mochizuki, Y., Koba, K., Yoh, M., 2012. Strong inhibitory effect of nitrate on atmospheric methane oxidation in forest soils. Soil Biology and Biochemistry 50, 164–166. https://doi.org/10.1016/j.soilbio.2012.03.013

Nazaries, L., Tate, K.R., Ross, D.J., Singh, J., Dando, J., Saggar, S., Baggs, E.M., Millard, P., Murrell, J.C., Singh, B.K., 2011. Response of methanotrophic communities to afforestation and reforestation in New Zealand. ISME Journal 5, 1832–1836. https://doi.org/10.1038/ismej.2011.62

Ni, X., Groffman, P.M., 2018. Declines in methane uptake in forest soils. Proceedings of the National Academy of Sciences 115, 201807377. https://doi.org/10.1073/pnas.1807377115

Norby, R.J., Luo, Y., 2004. Evaluating ecosystem responses to rising atmospheric CO2 and global warming in a multi-factor world. New Phytologist 162, 281–293. https://doi.org/10.1111/j.1469-8137.2004.01047.x

Pan, Y., Birdsey, R.A., Phillips, O.L., Jackson, R.B., 2013. The Structure, Distribution, and Biomass of the World's Forests. Annual Review of Ecology, Evolution, and Systematics 44, 593–622. https://doi.org/10.1146/annurev-ecolsys-110512-135914

Peñuelas, J., Poulter, B., Sardans, J., Ciais, P., Van Der Velde, M., Bopp, L., Boucher,

O., Godderis, Y., Hinsinger, P., Llusia, J., Nardin, E., Vicca, S., Obersteiner, M., Janssens, I.A., 2013. Human-induced nitrogen-phosphorus imbalances alter natural and managed ecosystems across the globe. Nature Communications 4. https://doi.org/10.1038/ncomms3934

Phillips, R.L., Whalen, S.C., Schlesinger, W.H., 2001. Influence of atmospheric CO2 enrichment on methane consumption in a temperate forest ecosystem. Global Change Biology 15, 741–752.

Price, S.J., Kelliher, F.M., Sherlock, R.R., Tate, K.R., Condron, L.M., 2004. Environmental and chemical factors regulating methane oxidation in a New Zealand forest soil. Australian Journal of Soil Research. https://doi.org/10.1071/SR04026

Price, S.J., Sherlock, R.R., Kelliher, F.M., McSeveny, T.M., Tate, K.R., Condron, L.M., 2003. Pristine New Zealand forest soil is a strong methane sink. Global Change Biology 10, 16–26. https://doi.org/10.1046/j.1529-8817.2003.00710x

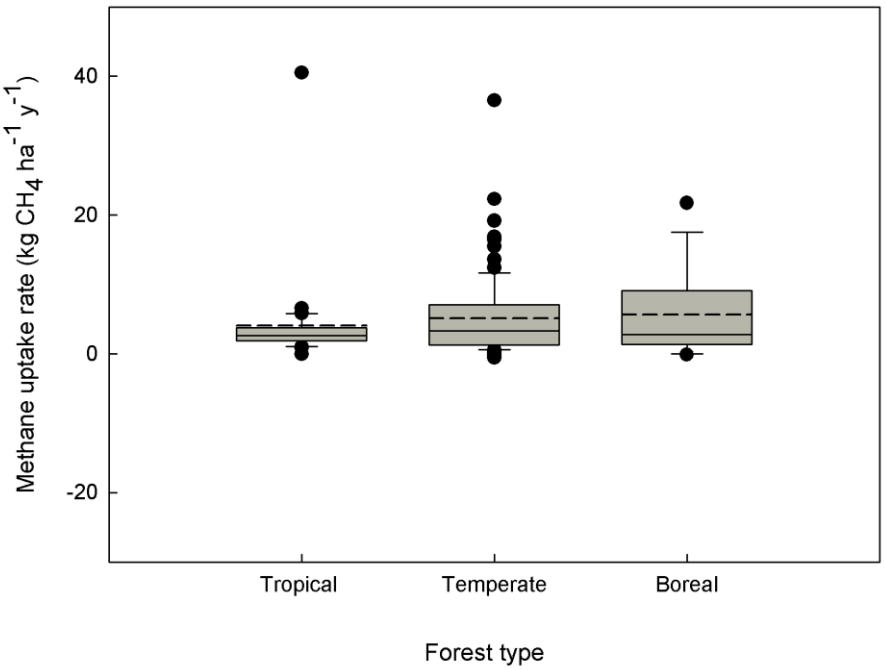
Schlesinger, W.H., Bernhardt, E.S., 2013. Biogeochemistry: An Analysis of Global Change, Third Edition, Biogeochemistry: An Analysis of Global Change, Third Edition. https://doi.org/10.1016/C2010-0-66291-2

Smith, K.A., Dobbie, K.E., Ball, B.C., Bakken, L.R., Sitaula, B.K., Hansen, S., Brumme, R., Borken, W., Christensen, S., Prieme, A., Fowler, D., MacDonald, J.A., Skiba, U., Klemedtsson, L., Kasimir-Klemedtsson, A., Degorska, A., Orlanski, P., 2000. Oxidation of atmosheric methane in Northen European soils, comparison with other ecosystems, and uncertainties in the global terrestral sink.

564 Global Change Biology 6, 791–803.
565 Steinkamp, R., Butterbach-Bahl, K., Papen, H., 2001. Methane oxidation by soils of a
566 N-limited and N-fertilized spruce forest in the Black Forest, Germany. Soil
567 Biology & Biochemistry 33, 145–153.
568 Stephenson, N., 1990. climatic control of vegetation distribution: the role of the water
569 balance. American Naturalist.
570 Teh, Y.A., Diem, T., Jones, S., Huaraca Quispe, L.P., Baggs, E., Morley, N., Richards,
571 M., Smith, P., Meir, P., 2014. Methane and nitrous oxide fluxes across an elevation
572 gradient in the tropical Peruvian Andes. Biogeosciences 11, 2325–2339.
573 https://doi.org/10.5194/bg-11-2325-2014
574 Verchot, L. V., Davidson, E.A., Cattânio, J.H., Ackerman, I.L., 2000. Land-use change
575 and biogeochemical controls of methane fluxes in soils of eastern Amazonia.
576 Ecosystems 3, 41–56. https://doi.org/10.1007/s100210000009
577 Wang, Z.P., Ineson, P., 2003. Methane oxidation in a temperate coniferous forest soil:
578 Effects of inorganic N. Soil Biology and Biochemistry 35, 427–433.
579 https://doi.org/10.1016/S0038-0717(02)00294-8
580 Xu, X.F., Tian, H.Q., Zhang, C., Liu, M.L., Ren, W., Chen, G.S., Lu, C.Q., Bruhwiler,
581 L., 2010. Attribution of spatial and temporal variations in terrestrial methane flux
582 over North America. Biogeosciences 7, 3637–3655. https://doi.org/10.5194/bg-7-
583 3637-2010
584 Yu, L., Huang, Y., Zhang, W., Li, T., Sun, W., 2017. Methane uptake in global forest
585 and grassland soils from 1981 to 2010. Science of the Total Environment 607–608,

586 1163–1172. https://doi.org/10.1016/j.scitotenv.2017.07.082
587

588 **Figure 1.** Boxplots of the rate of methane uptake in soils of tropical (n=30), temperate
589 (n=89), and boreal (n=15) forests. Short dashes represent means, solid lines represent
590 medians, and error bars indicate the 90th and 10th percentiles. An extreme outlier at -
591 120 kg CH_4 ha^{-1} y^{-1} in boreal forests has been omitted.
592



593
594

595 **Figure 2.** Partial residual plot. Partial residual plot of the variability of methane

596 uptake explained by MAT (°C), WS (water surplus (mm)) and the interaction between

597 them. 2a) the relationship between methane uptake and MAT under low, medium and

598 high WS (78, 405, 1010 mm); 2b) the relationship between methane uptake and WS

599 under low, medium and high MAT (4.5, 9.1, 21.8 °C) (*visreg* R package).

600

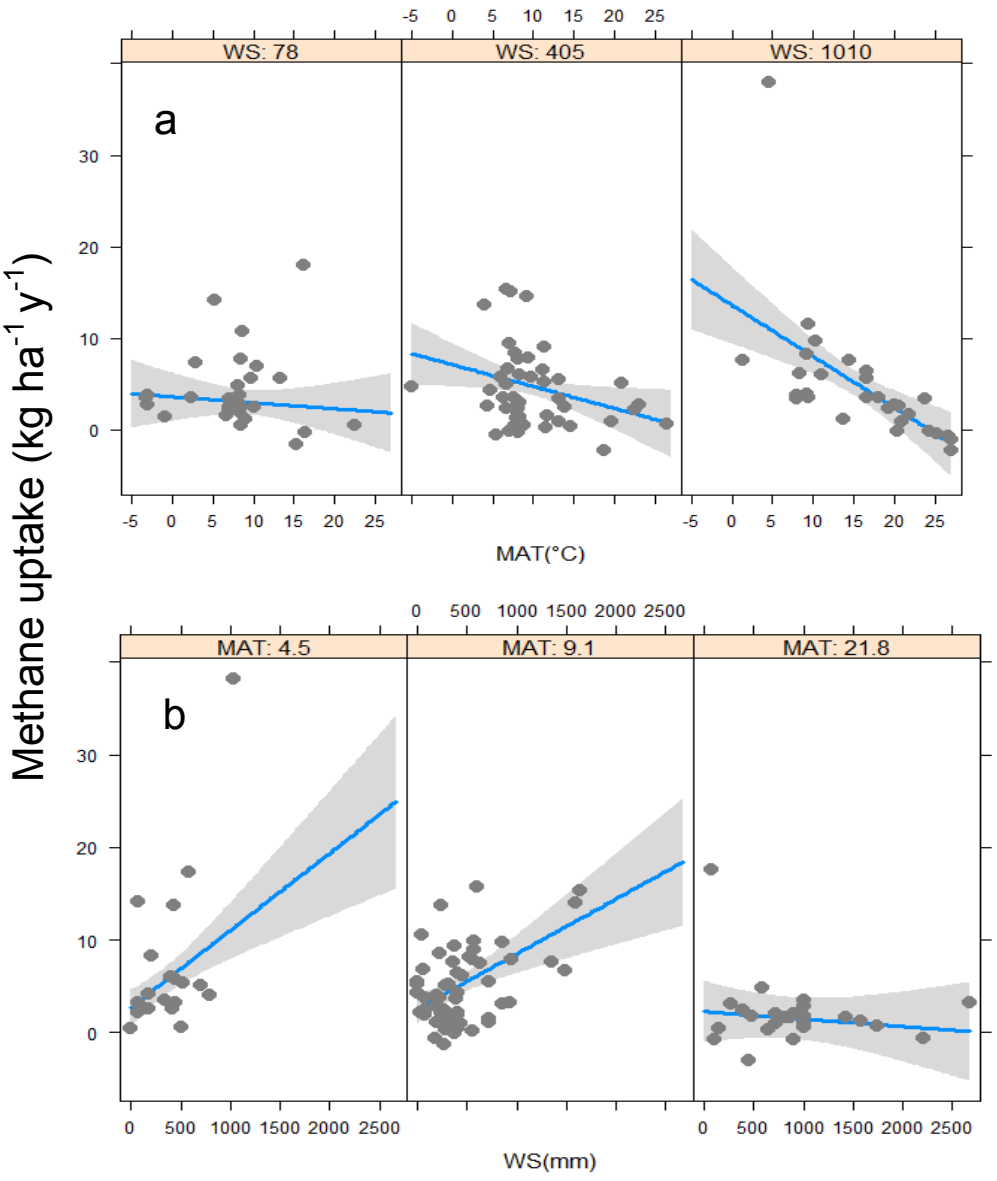


Figure 3. Effect of the environmental-change components drought, warming, elevated CO_2, and N enrichment on the rates of methane uptake represented as percent of change of the treatment versus the control values (horizontal bars, lower x-axis) and the natural-log response ratios associated with each treatment (filled circles). * $p < 0.05$, *** $p < 0.001$, ns not significant. Numbers in brackets after the environmental-change components indicate the number of experiments reviewed for each component.