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Abstract

Nitrous oxide (N_2O) and methane (CH_4) emissions were measured along three natural geothermal soil temperature (T_s) gradients in freely drained upland soils in a volcanic area in Iceland. Two of the T_s gradients (underneath a grassland (GN) and a forest site (FN), respectively) were recently formed (in May 2008) and thus subjected to relatively short-term warming. The third T_s gradient, underneath another grassland site (GO), had been subjected to long-term soil warming (over at least 45 years). The N_2O and CH_4 emissions were measured using the static chamber method. In addition, subsurface soil gas concentrations (5-20 cm) were studied. N_2O emissions from GN were slightly higher than those from GO in the temperature elevation range up to +5 $^{\circ}$C, while CH_4 uptake rates were similar. Under moderate soil warming (< +5 $^{\circ}$C) there were no significant increases in gas flux rates within any of the sites, but when soil warming exceeded +20 $^{\circ}$C, both N_2O and CH_4 emissions increased significantly at all sites. While net uptake of CH_4 (up to -0.15 mg CH_4 m^{-2} h^{-1}) and occasional N_2O uptake (up to -12 µg N_2O m^{-2} h^{-1}) were measured in the unwarmed plots at all sites, net emissions were only measured from the warmest plots (up to 2600 µg N_2O m^{-2} h^{-1} and up to 1.3 mg CH_4 m^{-2} h^{-1}). The subsurface soil N_2O concentrations increased with soil warming, indicating enhanced N-turnover. Subsurface soil CH_4 concentrations initially decreased under moderate soil warming (up to +5 $^{\circ}$C), but above that threshold they also increased significantly. A portion of the N_2O and CH_4 emitted from the warmest plots may, however, be geothermally derived, this should be further confirmed with isotope studies. In conclusion, our research suggests that moderate increases in soil temperature (up to +5 $^{\circ}$C) may not significantly increase N_2O and CH_4 emissions at these upland soils, both in the short and longer term. However, warming trends exceeding +5 $^{\circ}$C as predicted for 2100 in pessimistic scenarios may cause increased trace gas emissions and thus significant positive feedbacks to climate change.

1. Introduction

Nitrous oxide (N_2O) and methane (CH_4) are important greenhouse gases (GHGs). With a 100-year time horizon, the global warming potential (GWP) of N_2O is 265 times that of carbon dioxide (CO_2) (IPCC, 2014). N_2O is mainly produced in soils as a consequence of of two microbial activities; aerobic nitrification and anaerobic denitrification (Priemé and Christensen, 2001). The processes forming N_2O as an intermediate product are controlled by several factors such as temperature, moisture, pH and N-availability (Barnard et al., 2005; Brown et al., 2012). This implies that the emissions of N_2O are sensitive to changing environmental conditions. Upland forest soils in the Nordic countries are usually negligible sources of N_2O, whereas N-fertilized agricultural soils and drained peat soils are major sources (Maljanen et al., 2010a).

Methane has 28 times the GWP of CO_2 over a 100-year time horizon (IPCC, 2014) and is formed in soils by anaerobic methanogenesis (Le Mer and Roger 2001; Serrano-Silva et al., 2014). The production of CH_4 is primarily controlled by oxygen content, but is additionally controlled by soil temperature, pH, moisture and salinity (Le Mer and Roger 2001; Serrano-Silva et al., 2014). Methane that is produced in deeper soils layers can be transported to the atmosphere via ebullition through wet soils, via diffusion, or via the aerenchyma of vascular plants (Marushchak et al., 2016; Serrano-Silva et al., 2014). Methane can be oxidized in the soil by methanotrophic microbes, both under aerobic and anaerobic conditions (Knittel and Boetius, 2009). The optimum conditions for this process include neutral soil pH, a soil temperature of ~25 $^{\circ}$C and low salinity (Serrano-Silva et al., 2014). As a consequence, the CH_4 efflux from the soil is the net result of both methane production and methane oxidation.

As both processes are controlled by several environmental factors, any change therein can affect this efflux (Le Mer and Roger, 2001). Upland mineral soils are usually small sinks for atmospheric CH_4, whereas waterlogged wetlands are the major sources of CH_4 at northern latitudes (Maljanen et al., 2010a).

To study the effects of climate change on N_2O and CH_4 fluxes, warming experiments are often employed. However, short-term manipulative warming treatments can be impacted by several confounding factors and can be considered as over-simplistic (De Boeck et al., 2015). Natural temperature gradients (e.g. thermal gradients as a result of geothermal activity) on the other hand offer a number of benefits that makes them suitable "field laboratories" for research on GHG responses to soil warming (Kayler et al., 2015; O'Gorman et al., 2014). Geothermal activity can remain stable for many years, making it possible to investigate long-term warming effects, but major tectonic events can also create new hotspots, exposing previously unwarmed ecosystems to higher temperatures and enabling studies of recent (short-term) temperature responses (O'Gorman et al., 2014).

Both types of natural soil temperature gradients can be found in the Hellisheiði geothermal systems in southwest Iceland. In May 2008, a major earthquake in southern Iceland affected the geothermal systems close to its epicenter (Halldorsson and Sigbjörnsson, 2009). A part of this geothermal system moved from its previous location to a new and previously unwarmed area, creating a natural soil warming experiment. This offers a unique opportunity to study how various ecosystem processes, including N_2O and CH_4 dynamics, are affected by short-term temperature changes. Other geothermal systems in the area were not affected by the earthquake, providing natural soil temperature gradients that can be used to study long-term soil temperature effects on N_2O and CH_4 dynamics.

The "ForHot" research network was established in 2012 to bring European scientists together to study how changes in soil temperature affect various ecosystem processes in both natural grasslands and a planted 45-year old Sitka spruce forest in southern Iceland. The large range in temperature elevations at the ForHot sites offers both conditions similar to the predicted climate change during the next century as well as more extreme temperatures that can yield new insights into stress physiology (Kayler et al., 2015; O'Gorman et al., 2014).

The major aim of this study within the ForHot project was to investigate changes in N_2O and CH_4 flux rates along soil temperature gradients to better predict the impacts of future soil warming on atmospheric impacts of terrestrial ecosystems. Our hypothesis was that a significant increase in soil temperature will accelerate microbial processes and turn these sites from N_2O and CH_4 sinks to sources.

2. Methods

2.1 Study site

The ForHot study sites are located in southwest Iceland, in the surroundings of the village Hveragerdi (64.008°N, 21.178°W), on land owned by the Agricultural University of Iceland (Fig. 1). In 2004-2014, the area had a mean annual air temperature of 5.2 °C and a mean annual precipitation of 1431 mm (Icelandic Met Office, IMO). The growing season normally starts in May and ends in late August. The soil type at the study sites is Brown Andosol (Arnalds, 2015), with relatively high pH (5.5-7.0) and large soil water retention capacity (O'Gorman et al., 2014).

On the 29th of May, 2008, a major earthquake (magnitude 6.3 on the Richter scale) occurred in southwest Iceland (Halldorsson and Sigbjörnsson, 2009), where ca. 70-100 years pass between such large earthquake episodes in this region. The 2008 earthquake caused large structural damages to infrastructures and affected geothermal systems close to its epicenter. One such geothermal system moved from its previous location to a new and previously unwarmed area (Þorbjörnsson et al., 2009), and the new belowground geothermal channels within the bedrock resulted in soil temperature increases in the soil above. The soil temperature elevation measured at 10 cm soil depth reaches >50 °C where the channels are closest to the surface (O'Gorman et al., 2014). The recently warmed area is covered by two different ecosystem types: a) a planted 45 year old Sitka spruce (*Picea sitchensis*) forest (Forest New, FN) and b) a natural, unmanaged treeless grassland (Grassland New, GN) dominated by *Festuca* sp., *Agrostis* sp. and moss (Fig. 1).

The third study site (Grassland Old, GO) is located 2.5 km NW from GN and FN on an older temperature gradient in Grændalur (the 'green valley') allowing to study the long-term effects of warming. It is covered by the same grassland type as GN. At the GO site the earliest survey of geothermal hot-spots was made in 1963-1965 (45 years prior to the 2008 earthquake) (Kristján Sæmundsson, pers. comm.). In autumn 2008, the locations of the new and old geothermal hot-spots in the area were mapped and published (Þorbjörnsson et al., 2009). This survey was used to select the GO and GN sites for the ForHot study. Further, the existence of some of the hot-spots prior to the 2008 earthquake at GO is also supported by regular field measurements of soil temperature since 2005 in a previous study in Grændalur (Daebeler, 2014). The geothermal activity has most likely been persistent in Grændalur for centuries, however, as according to local knowledge its name is derived from the subarctic grasslands staying green during most of the winter on the warmest hot-spots. The oldest

historical document that mentions this place name was published in 1708 (Magnússon and Vídalín, 1918-1921). Additional evidence for persistent geothermal warming at the GO site are geothermal clay layers found at various depths in the soil profile (Leblans, pers. communication). This indicates that over longer time periods, the warming may have fluctuated somewhat, as was observed at other nearby, older hot-spots following the 2008 earthquake (Daebeler, 2014).

2.2 Chamber measurements

The gas fluxes (N_2O and CH_4) were measured using the static chamber method (e.g. Maljanen et al., 2010b). The flux measurements were made along temperature gradients in FN, GN and GO during years 2012-2014 in several campaigns during the growing season (May-August) and in GN additionally during one winter campaign in February 2015. There were five to six gas flux sampling points on each gradient. The sampling points were covering unwarmed soil temperature (T_s) and elevated T_s, up to ~ +40, +20 and +11 $^{\circ}$C in FN, GN and GO respectively. The warming levels measured in 2012 are indicated in Figs. 3-6 with site code (FN, GN or GO) and temperature elevation as +x $^{\circ}$C. Because the temperatures along the gradient are slightly changing all the time the actual measured temperatures during gas sampling are shown in the box plots in Figs. 3-6. There were two replicate flux chambers in growing season 2012 and in winter 2015 and three in growing seasons 2013-2014 at each sampling point. The metal flux chambers (ø = 26 cm, h = 30 cm) had a hole in the top for a sampling line. Prior to sampling the sharp edge at the bottom of the chamber was twisted 3-5 cm into soil and the top was sealed with a rubber septum. A total of four gas samples (30 ml) were collected 5, 10, 20, and 30 min after installing the chamber with a 60 ml syringe (Terumo). Within 12 h, the samples were injected into 12 ml Labco pre-evacuated vials (Labco Exetainer®) for gas analysis with a gas chromatograph at University of Eastern

Finland (UEF). Soil temperatures were recorded next to the chambers at each sampling location at depths of 3, 5, 10, 15 and 20 cm.

2.3 Soil gas sampling

Soil N_2O and CH_4 concentrations were measured at the sampling points simultaneously with the gas flux measurements in June 2014. Gas samples of 20 ml were taken with a stainless steel sampling probe (ø = 3 mm, l = 30 cm) at three soil depths; 5, 10 and 20 cm. Samples of N_2O and CH_4 were treated and analysed as described above.

2.4 Gas concentration analysis

Concentrations of N_2O and CH_4 were determined with a gas chromatograph (Agilent 6890N, Agilent Technologies, USA), equipped with an autosampler (Gilson, USA) an electron capture (ECD), and flame ionization detectors (FID). Compressed air, containing 0.836 µl l^{-1} of N_2O, and 2.02 µl l^{-1} of CH_4, was used for daily calibration. The gas flux rates were calculated from the linear increase or decrease in the gas concentration with time in the headspace of the chamber. If there were any indications of failure in the gas sampling or gas analysis, the results were excluded from the calculations.

2.5 Soil analysis

Soil samples (sampling depths 0-5 cm and 5-10 cm) were collected from all sites in June 2014 and transported to UEF for analysis. Soil pH and electric conductivity (EC) were measured from soil/water slurry (15 ml soil: 50 ml milliQ-H_2O) from homogenized and pooled samples. Soil nitrate (NO_3^-), nitrite (NO_2^-), chloride (Cl^-), sulfate (SO_4^{2-}), ammonium (NH_4^+) and dissolved organic carbon (DOC) concentrations were measured from soil extracts. For anion analysis, 15 ml soil and 50 ml H_2O were shaken at 175 rpm for one hour,

196 filtered, and analyzed with an ion chromatograph (DX 120, Dionex Corporation, USA). DOC
197 concentrations were measured from H_2O-extracts with a TOC analyzer (TOC/TNb Analyzer
198 Multi N/C 2100S, Analytik Jena AG). To extract NH_4^+, 15 ml soil and 50 ml 1M KCl were
199 used. NH_4^+ was analyzed with a spectrophotometer (Ultrospec 3000 Pro, Biochrom, UK)
200 from the filtered KCl extracts using the method of Fawcett and Scott (1960). Gravimetric soil
201 moisture was determined by drying the soil for 24 h at 105 °C. Organic matter (OM)
202 concentration was determined by loss on ignition at 550 °C. Soil C and N contents were
203 determined with an EA-analyzer (Flash EA 1112 Series, Thermo Electron Corporation,
204 USA).
205
206 **2.6 Statistical analyses**
207 Because the flux data was not normally distributed even after log-transformations the
208 correlations between the gas flux rates and T_s were tested with non-parametric Spearman
209 rank correlation tests. For the correlations between other soil variables, Pearson correlation
210 tests were used. The differences in the gas flux rates were tested with Mann-Whitney U-test
211 (IBM SPSS statistics 19).
212
213 **3. Results**
214
215 **3.1 Soil temperature**
216 Across the summer and autumn measurement campaigns, the mean T_s at 10 cm depth in the
217 unwarmed plots was 8.3, 8.7 and 8.4 °C in FN, GN and GO. T_s gradually increased along the
218 sampling transect up to 50.4, 28.4 and 18.0 °C, in FN, GN and GO respectively (Fig. 2; Figs.
219 3-5.). During the winter campaign at the GN site in February 2015 (Fig. 6), T_s were lower
220 (ranging from -0.5 to 12.4 °C) than during the growing season (Fig. 4).

221

222 **3.2 Soil properties**

223 The measured soil properties are presented in Table S1. Soil pH increased with increasing
224 temperature ($r = 0.805$, $p = 0.001$; $r = 0.811$, $p = 0.002$) in FN and GN, respectively (Fig. 2).
225 The pH of unwarmed soils varied from 5.2 in FN to 5.7 in GO, and up to 7.3 in GN (Fig. 2).
226 Soil OM concentrations at depths of 5-10 cm decreased with T_s see Fig 2. ($r = -0.693$, $p =$
227 0.038; $r = -0.731$, $p = 0.039$; $r = -0.841$, $p = 0.009$ for FN, GN and GO respectively). Soil C
228 and N concentrations decreased with increasing T_s, but this decrease was only significant in
229 FN ($r = -0.819$, $p = 0.007$ and $r = -0.750$, $p = 0.020$) and GN ($r = -0.749$, $p = 0.320$ and $r = -$
230 0.737, $p = 0.370$). However, soil gravimetric moisture, EC, NO_3^-, NH_4^+, DOC, SO_4^{2-} or Cl^-
231 content did not correlate with T_s at any of the sites (Table S1). Soil nitrite (NO_2^-)
232 concentrations were lower than the detection limit of 0.01 µg g^{-1} soil in all samples.

233

234 **3.3 Gas flux rates**

235 With a moderate T_s increase (up to +5 °C), no statistically significant increase of N_2O or CH_4
236 flux rates were observed at any of the sites. However, beyond the +5 °C warming threshold,
237 both N_2O and CH_4 emissions increased (Figs. 3-5). Nitrous oxide emissions from the short-
238 term warmed grassland (GN) were slightly higher than those from the long-term warmed
239 grassland (GO), associated with temperature increase of ≤+5 °C but CH_4 uptake rates were
240 similar. While a net uptake of CH_4 (up to -0.15 mg CH_4 m^{-2} h^{-1}) and occasionally also N_2O
241 uptake (up to -12 µg N_2O m^{-2} h^{-1}) were measured in the unwarmed plots, net emissions were
242 measured at all the warmest plots. Because T_s in the sampling plots varied with time (due to
243 variation in ambient air temperature and minor fluctuations in the geothermal warming) the
244 actual temperatures during sampling are shown in Figs. 3-6. The measured N_2O emissions
245 from plots with the most extreme T_s increase (Site FN ~ +40 °C and GN ~ +20 °C) were two

246 magnitudes higher (maximum up to 2600 µg N_2O m^{-2} h^{-1}) compared to the unwarmed control
247 plots (less than 20 µg N_2O m^{-2} h^{-1}) (Table 1, Figs. 3, 4). At the GO site, the highest N_2O
248 emissions were also measured at the warmest plot, which was on average ~ +11 °C warmer
249 than the unwarmed soils (Fig. 5). However, at all other sampling points at the GO site, N_2O
250 emissions were similar along the gradient. The CH_4 fluxes only correlated significantly with
251 increasing T_s at the GO site (Table 1).
252
253 The winter N_2O emission rates at the GN plots did not differ significantly from those during
254 the growing season. However, the winter CH_4 fluxes were less often negative (uptake) than
255 during the growing season ($p < 0.001$; Mann-Whitney U-test). The winter N_2O emissions
256 increased with increasing T_s ($r = 0.488$, $p < 0.001$, Fig. 6), while the winter CH_4 fluxes did
257 not correlate with T_s.
258
259 **3.4 Gas concentrations in soil**
260 The soil N_2O and CH_4 concentrations, sampled with a metal probe from 5, 10 and 20 cm soil
261 depth varied significantly between plots and depths (Fig. 7). Soil N_2O concentrations
262 increased with depth, up to 100 µl l^{-1}, at GN and FN sites. At GO site the N_2O concentrations
263 were similar, less than 1 µl l^{-1}, in all plots and all depths except slightly higher in the warmest
264 (+11 °C) plot (Fig. 7). The CH_4 concentrations also increased with depth in the warmest
265 plots, but the opposite was true for the unwarmed control plots, where soil CH_4 concentration
266 was the lowest at 20 cm depth (Fig. 7).
267

4. Discussion

In this study soil temperature increases of less than 5 °C did not significantly affect soil emissions of N_2O and CH_4. The N_2O emissions from the unwarmed forest soil were relatively low (mean 2.5 µg m^{-2} h^{-1}), and were in the range of previous reports for boreal upland forest soils in the Nordic countries (e.g. Maljanen et al., 2010a; 2010b). The N_2O emissions from the unwarmed grassland soil were also low (mean emissions from GN were 3.9 µg m^{-2} h^{-1} and from GO 2.2 µg m^{-2} h^{-1}). Only in the plots with extreme soil temperature elevations (up to +53 ^{o}C), N_2O emissions were significantly enhanced (up to 2600 µg m^{-2} h^{-1}). The magnitude was higher than earlier reported from drained peat soils, which are considered to be the hot spots for N_2O emissions in the Nordic countries (Maljanen et al., 2010a). The increase in N_2O emissions at the warmest plots may be related to higher nitrogen mineralization and nitrification rates, which are frequently found in soils subjected to warming treatment in the North (e.g. Biasi et al., 2008). However, N_2O may be at least partly geothermally derived as reported by Klusman et al. (2000) from geothermal fields in the USA and discussed further below.

Unwarmed forest and grassland soils were small sinks for CH_4 (mean -0.4 mg m^{-2} h^{-1}), with only the warmest plots producing net CH_4 emissions (up to 1.33 mg m^{-2} h^{-1}). Elevated temperatures (in the range of +5 to +25 $^{\circ}$C) have been shown to decrease CH_4 uptake in soils (Dijkstra et al., 2011). In our study, moderate warming (up to +5 $^{\circ}$C) led to a slight increase in CH_4 uptake, but when temperature increased by more than 5 $^{\circ}$C, the soils turned into net CH_4 sources. Our data suggest a seasonal trend in CH_4 fluxes. In summer, the cooler plots (below +5 $^{\circ}$C) were primarily CH_4 sinks, while the warmer plots were CH_4 sources. In winter, on the contrary, all CH_4 fluxes at GN site were close to zero as a consequence of the low soil

temperatures. Methane can also be produced in both biological and geothermal processes (Tassi et al., 2012). Especially the very high CH_4 emissions from the warmest plots are difficult to explain from biological sources only. Upland mineral soils with low organic matter and water content are usually sinks for atmospheric CH_4 as a result of methane oxidation in the oxic top soil and such high emissions are uncommon (Le Mer and Roger 2001; Megonigal and Guenther 2008).

There is some evidence that supports the assumption that the source of the high N_2O and CH_4 emissions are non-biological, from a geothermal source. Rey (2015) reported that in geothermal areas the geological CO_2 can play an important role in the soil CO_2 emissions, which has been observed also in nearby geothermal areas in Iceland (Fridriksson et al., 2006). This could also be true for N_2O (Klusman et al., 2000). The concentration of mineral N (NH_4^+ or NO_3^-) did not increase with soil temperature (Supplementary Table 1) or correlate with N_2O emissions. However, further studies are needed to confirm the hypothesis that non-biological sources contribute to the overall N_2O and CH_4 release in geothermal areas, particularly isotope studies of both GHGs in soil profile and surface emissions. The isotope signal of N_2O from geothermal sources has, to the best of our knowledge, not been characterized so far, but it is likely that it differs significantly from biological or atmospheric sources (as found for N_2 from hydrothermal fluids; Caliro et al., 2015). Similarly, CH_4 from geothermal reservoirs exhibits a different $\delta^{13}C$ values than biologically produced CH_4 (it is more enriched in ^{13}C; Klusman et al., 2000) which would aid the source identification of CH_4. As mentioned already above, we hypothesize that geothermal CH_4, reported by e.g. Etiope and Klusman (2002) and Klusman et al. (2000), may also be present in the ForHot research area since such high CH_4 emissions at the warmest plots are difficult to explain from upland mineral soils. The very high concentrations of CH_4 in the deeper soil profiles indicates

318 there must be a significant source for CH_4. High CH_4 concentrations, not associated with any
319 geothermal origin, have not been found in upland mineral soils in contrast to wetland soils
320 (LeMer and Roger 2001).
321
322 These geothermally active sites can act as local hot spots for N_2O or CH_4 emissions
323 (Klusman et al., 2000). While N_2O emissions from such sites are not well documented, the
324 global geothermal emissions of CH_4 are estimated in the range of 3-7 Mt $year^{-1}$ (Etiope and
325 Klusman, 2002; Etiope et al., 2008; Lacroix, 1993). Etiope (2009) reported that even one
326 third of Europe's methane emissions can be geological origin. Data on geothermal emissions
327 of N_2O are lacking, but would be important for better estimating the global N_2O budget.
328
329 However, there can be also biological factors explaining partly the change in GHG flux rates
330 with increasing temperature. Soil organic matter content as well as C content decreased with
331 increasing soil temperature in the FN and GN site, indicating that organic matter is already
332 more decomposed in the warmer plots during the first six years after the formation of these
333 gradients. The availability of organic matter can positively affect the CH_4 production as well
334 as N_2O production in the soil (Harrison-Kirk et al., 2013; Le Mer and Roger, 2001; Maljanen
335 et al., 2009). On the other hand, in the GO site, the increase in soil temperature was smaller
336 than in the other sites and we did not see any dramatic changes either in the flux rates or in
337 the soil gas concentrations. However, there was a clear decrease in soil OM, C and N content
338 with increasing soil temperature, which could indicate that most of the C and N had already
339 been lost from the warmest plots, possibly limiting the microbiological production of CH_4
340 and N_2O.
341

The clear increase in soil pH with increasing soil temperature could favor denitrification. Nitrification and denitrification activities are favored by neutral soil pH (e.g. Brenzinger et al., 2015), but at the same time N_2O reductase is favored by high pH. Therefore, N_2O should be more efficiently reduced to N_2 at higher pH. The activity of methanogens (CH_4 production) and methanotrophs (CH_4 oxidation) is favored also by neutral soil pH (Le Mer and Roger, 2001). In this study, the net CH_4 production was increasing more than CH_4 oxidation, as it increased with pH.

We did not observe any significant changes in the N_2O or CH_4 flux rates associated with moderate soil temperature increases (< 5 $^{\circ}$C). The expected increase in soil temperature as a result of global warming for the subarctic sites ranges from 2.6 to 8.5 $^{\circ}$C by the end of this century (IPCC, 2014). If the most pessimistic scenario would come true, more N_2O and CH_4 emissions from northern soils can be expected, if the sources of these two trace gases investigated here are from biological origin. When soil temperature increased by more than 10 $^{\circ}$C, the flux rates increased significantly at the short-term warmed FN and GN sites, but not at the long-term warmed GO site, which could be the result of less C and N available at the long term warmed sites. In any case, we can conclude that a moderate increase (< 5 $^{\circ}$C) in soil temperature with climate change would not significantly increase N_2O or CH_4 emissions from the studied soils. In this study, wecannot separate the role of microbial activity from geothermal causes in explaining the dramatic increase in N_2O or CH_4 flux rates from the warmest soils, but we speculate that geothermal sources play a role. By employing stable isotope analysis of CH_4 and N_2O, we could further examine the mechanisms that underlie changes in flux dynamics. By doing so, the role of geothermal and biological processes can be investigated in more detail which is needed in order to conclusively interpret the results from studies on thermal gradients in geothermal areas, in predicting climate change impacts.

Better estimates on geothermal sources of N_2O and CH_4 would be needed in order to better constrain the global budget of these trace gases.

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Tables

Table 1. Spearman rank correlation coefficients between soil temperature at 5 and 10 cm depth and N_2O and CH_4 flux rates during the growing seasons from the temperature gradients at the forest (FN), the new grassland (GN) and the old grassland (GO) site. ** = $p < 0.001$, * = $p < 0.05$.

Site FN	T_5	T_{10}	N_2O
T_{10}	0.996**		
N_2O	0.422**	0.411**	
CH_4	0.411**	0.410**	0.408**

Site GN	T_5	T_{10}	N_2O
T_{10}	0.993**		
N_2O	0.468**	0.495**	
CH_4	0.432**	0.439**	0.393**

Site GO	T_5	T_{10}	N_2O
T_{10}	0.996**		
N_2O	0.001	0.023	
CH_4	0.595**	0.598**	0.011

Figures

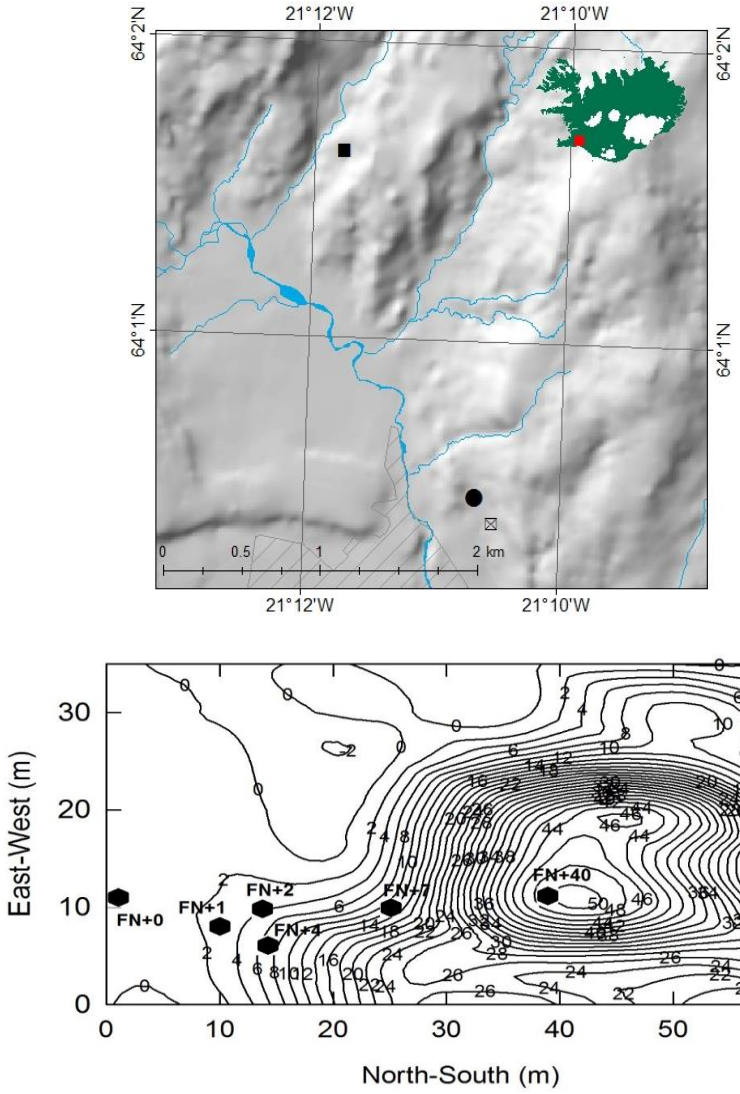
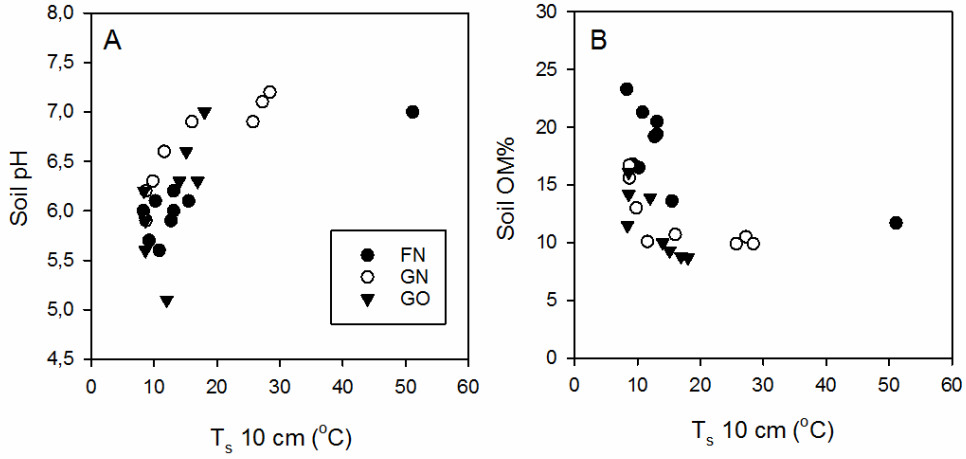
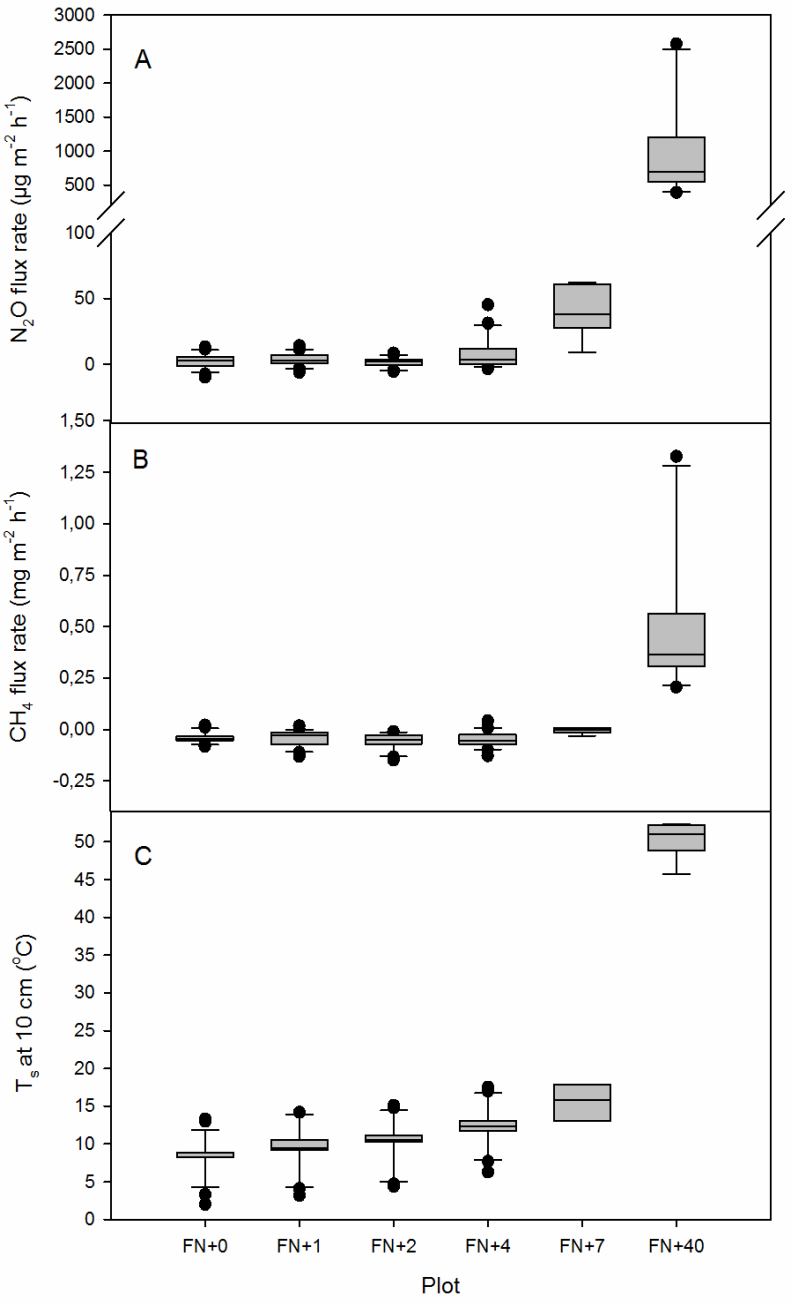


Fig. 1. Location of the three study sites in southern Iceland (upper panel). The circle marks the recently warmed forest site (FN), the crosshatched square the recently warmed grassland (GN) and the filled square the grassland with long-term soil warming (GO). The hatched area represents the village of Hveragerdi. Soil warming isotherms (°C) within the FN site in spring 2012 (lower panel), where the natural background soil temperature (Ts) varied by -2 to +2 °C in differently shaded parts of the stand and the actual geothermal warming was ca. 2 °C lower than the isolines indicate. The location of the study plots is also shown, where the measured geothermal warming at 10 cm depth were determined as 0, +1, +2, +4, +7 and ca. +40 °C.



529

530 **Fig. 2.** Soil pH (A) and organic matter (B) concentration (%) at a depth of 5-10 cm, plotted
531 against soil temperature (T_s) at a depth of 10 cm, measured in June 2014. Black circles
532 represent the forest site (FN), open circles represent the new grassland site (GN) and triangles
533 represent the old grassland (GO).



534

535 **Fig. 3.** Flux rates of N_2O (A) and CH_4 (B) and the average soil temperature (Ts) at a depth of
536 10 cm during sampling at forest (FN) site, measured during the growing seasons of 2012 -
537 2014. The median, 25th and 75th percentiles are shown in the box with whiskers indicating
538 variability outside the 25th and 75th percentiles. Plot codes at the x-axis show the average
539 soil temperature increase (°C). Plots FN+0 to FN+4 were measured eight times, while plots
540 FN+7 and FN+40 were measured four times. Each time, two or three replicate chambers were
541 used (see Methods).

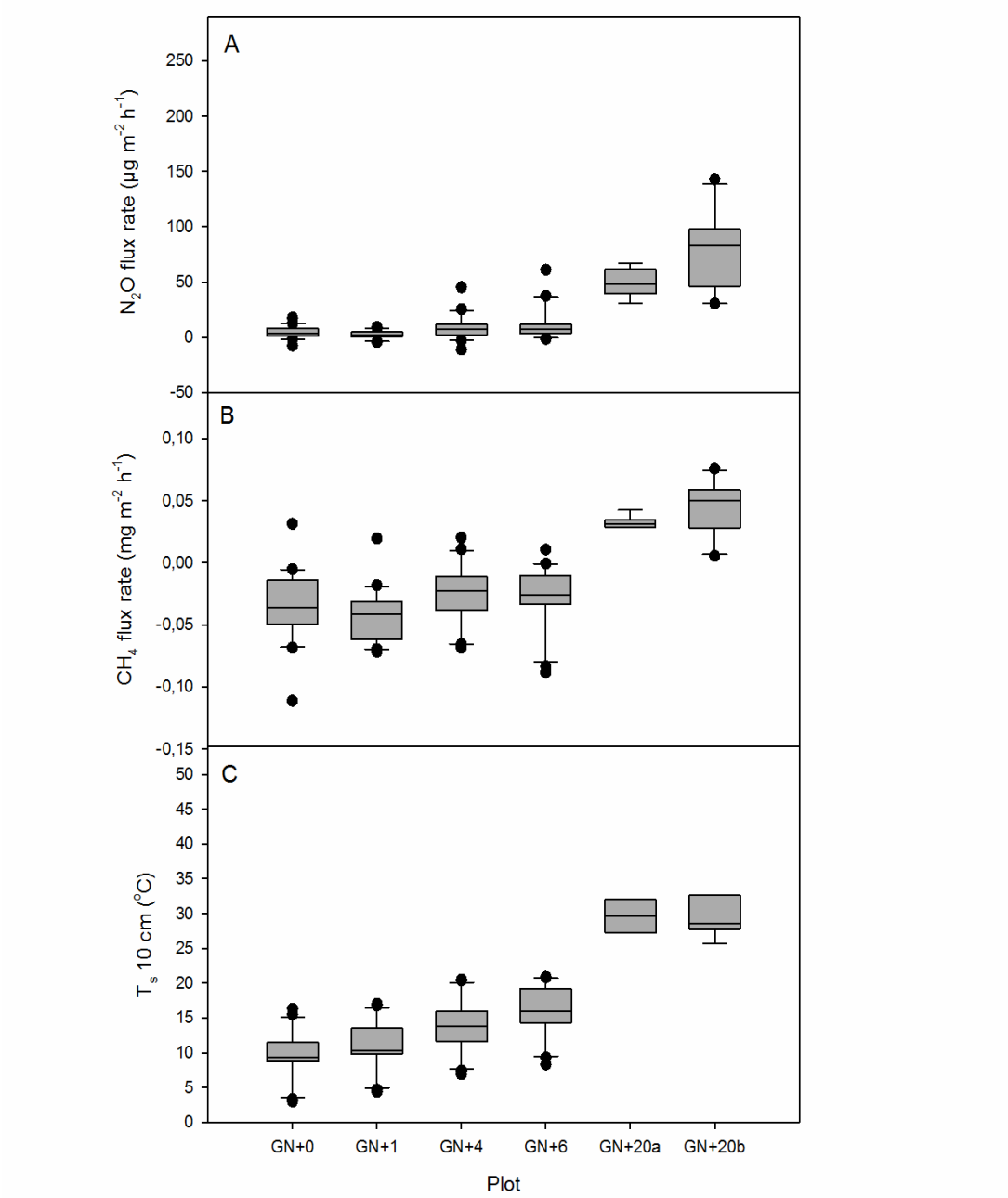


Fig. 4. Flux rates of N_2O (A) and CH_4 (B) and the average soil temperature (T_s) at a depth of 10 cm during sampling at grassland (GN) site, measured during the growing seasons of 2012 - 2014. The median, 25th and 75th percentiles are shown in the box with whiskers indicating variability outside the 25th and 75th percentiles. Plot codes at the x-axis show the average soil temperature increase (^oC). Plots GN+0 to FN+6 were measured eight times, while plots GN+20a and GN+20b were measured four times. Each time, two or three replicate chambers were used.

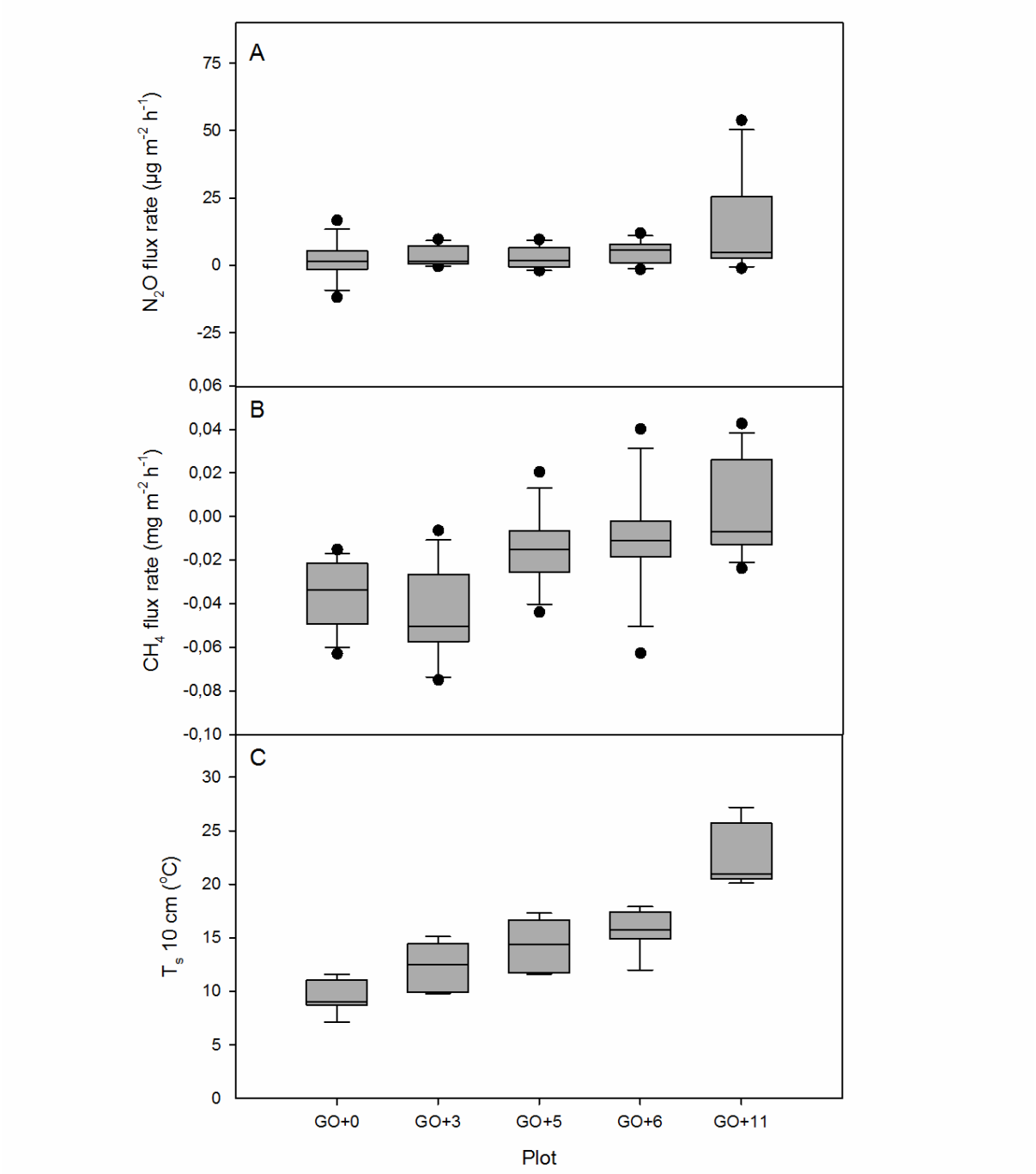
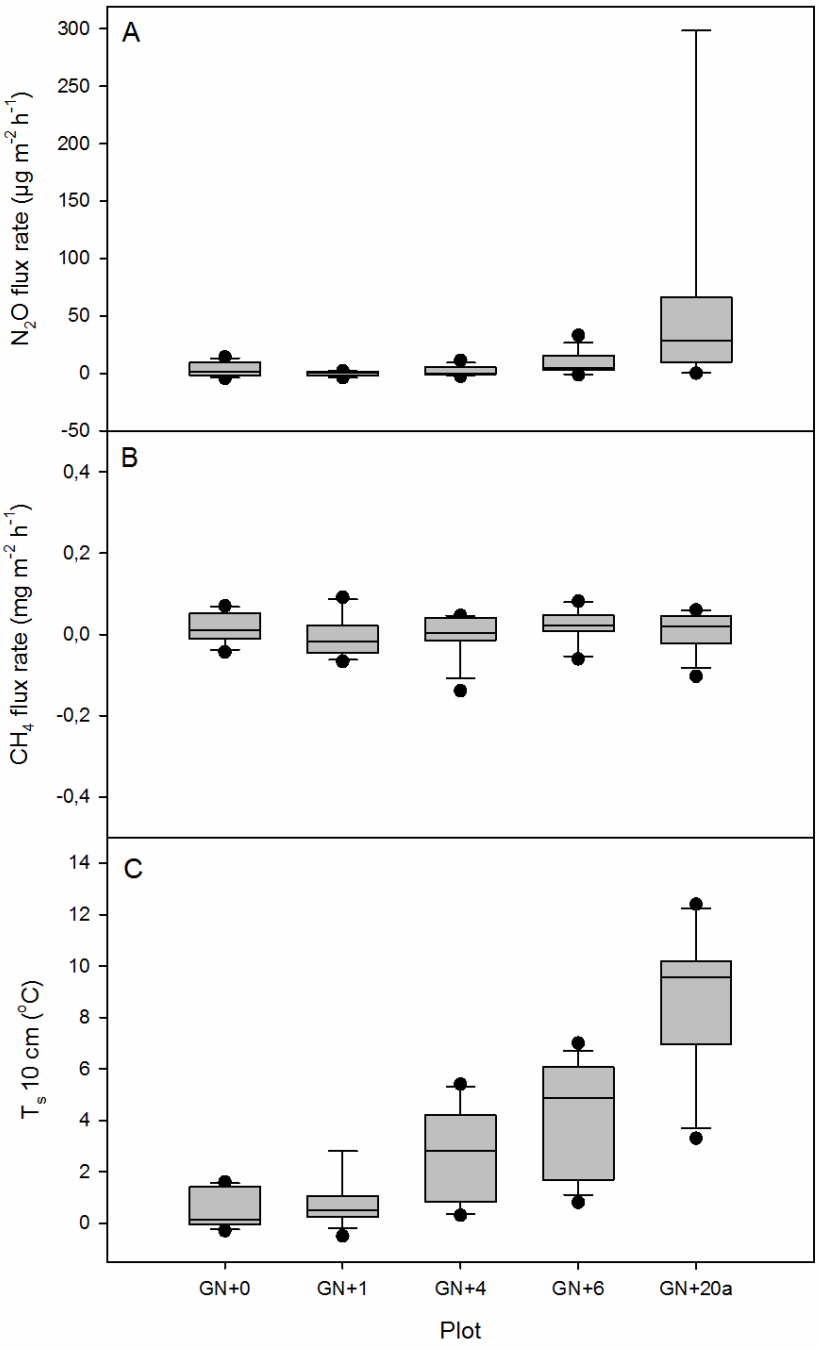
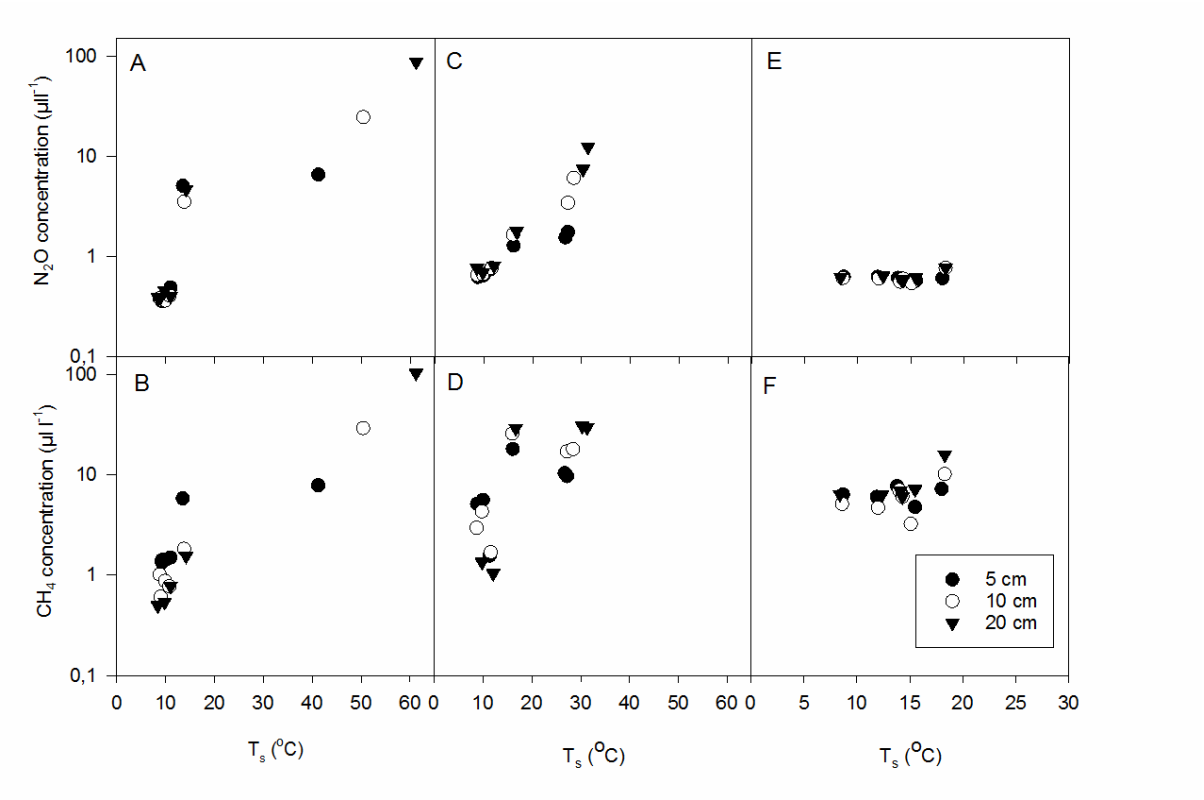


Fig. 5. Flux rates of N_2O (A) and CH_4 (B) and the average soil temperature (T_s, °C) at a depth of 10 cm during the sampling from the old grassland temperature gradient (GO, measured during the growing seasons of 2013 and 2014. The median, 25th and 75th percentiles are shown in the box with whiskers indicating variability outside the 25th and 75th percentiles. Plot codes at the x-axis show the average soil temperature increase (°C). Each plot was measured five times with two or three replicate chambers.



557

558 **Fig. 6.** Flux rates of N_2O (A) and CH_4 (B) and the average soil temperature (T_s, °C) at a
559 depth of 10 cm during the sampling from the grassland (GN) temperature gradient, measured
560 in February 2015. The median, 25th and 75th percentiles are shown in the box with whiskers
561 indicating variability outside the 25th and 75th percentiles. The plot codes are similar as in
562 Fig. 4 and indicate soil temperature increases during growing the growing season. Each plot
563 was measured seven times with two replicate chambers.



564

565 **Fig. 7.** N_2O and CH_4 concentrations (µl l^{-1}) measured at 5, 10 and 20 cm soil depth at the FN

566 site (A, B), the GN site (C, D) and the GO site (E, F), plotted against soil temperature at

567 sampling depth in June 2014. Note the logarithmic scale on the y-axis.