UC Irvine

UC Irvine Previously Published Works

Title

Consumption of atmospheric methane by tundra soils

Permalink

https://escholarship.org/uc/item/8vs232b0

Journal

Nature, 346(6280)

ISSN

0028-0836

Authors

Whalen, SC Reeburgh, WS

Publication Date

1990

DOI

10.1038/346160a0

Copyright Information

This work is made available under the terms of a Creative Commons Attribution License, available at https://creativecommons.org/licenses/by/4.0/

Peer reviewed

The abrupt change of temperature and vertical velocity component at the depth of 1.08 m observed on the profiles obtained at 01:25 GMT (Fig. 3) can be ascribed to vertical intersection of a frontal interface (see the sketch on Fig. 1 showing eddies and frontal interfaces in the upper ocean boundary layer). The temperature change at this interface was $\Delta\theta \approx -0.05$ °C, the appropriate change of the vertical velocity being $\Delta W \approx$ 2.5 cm s⁻¹. The sign of the vertical velocity change at 1.08 m depth indicated that the cold water was moving along the interface away from the ocean surface. This coincides with water circulation induced by the large-scale eddies in the upper ocean boundary layer (Fig. 1).

Similar profiles obtained at 00:55 GMT (Fig. 3), but having the opposite sign of the vertical velocity change at 0.95 m depth and the same sign of the temperature change as the profiles discussed above, may be interpreted as a result of intersection of the region where the cold water was moving along the frontal interface toward the ocean surface (see Fig. 1).

The profiles obtained at 01:06 GMT have no substantial changes either in temperature or in vertical component of velocity. These profiles can be interpreted as referring to a space between the large-scale eddies. The other four pairs of profiles

shown in Fig. 3 apparently are concerned with cases when the profiler passes through less pronounced regions of the organized vortex structure. Vertical profiles obtained on 23 May had similar temperature and velocity features.

Received 2 April: accepted 2 May 1990

- Thorpe, S. A. Nature 318, 519-522 (1985).
- Thorpe, S. A. Sci. progr., Oxf. 72, 189-206 (1988)
- Csanady, G. T. *J. phys. Oceanogr.* **14**, 402–411 (1984). Soloviev, A. V., Vershinsky, N. V. & Bezverchnii, V. A. *Deep Sea Res.* **35**, 1859–1874 (1988).
- Antonia, R. A., Chambers, A. J., Frishe, C. A. & Van Atta, C. W. J. atmos. Sci. 36, 99-108 (1979). Phong-Anant, D., Antonia, R. V., Chambers, A. J. & Rajagopalans, S. J. J. geophys. Res. 85, 424-432
- Brown, G. L. & Thomas, A. S. *Physics Fluids* **20**, S243–S252 (1977). Thorpe, S. A. & Hall, A. J. *Nature* **328**, 48–51 (1987).
- Volkov, Yu. A. et al. Izv. Akad. nauk SSSR, 25, 695-701 (1989)
- Van Dyke, M. An Album of Fluid Motion (Parabolic, Stanford, 1982).
 Soloviev, A. V. & Vershinsky, N. V. Deep Sea Res. 29, 1437–1449 (1982).
- Shay, T. J. & Gregg, M. C. J. phys. Oceanogr. 16, 1777-1798 (1986).
- Brubaker, J. M. Nature 330, 742-745 (1987).
 Thorpe, S. A. & Hall, A. J. J. Fluid Mech. 101, 687-703 (1980)
- Armorocho, J. & De Vries, J. J. J. geophys. Res. 85, 432-442 (1980).
- 16. Oceanography Tables (Hydrometeoizdat, Leningrad, 1975) (in Russian). 17. Soloviev, A. V. Izv. Akad. nauk SSSR 18, 751–759 (1982).

ACKNOWLEDGEMENTS, I thank V. A. Bezverkhnii, V. V. Turenko, F. M. A. Frmolaev, V. I. Usachev and A. A. Grachev for technical help, and, N. V. Vershinsky and G. S. Golitsyn for their support.

Consumption of atmospheric methane by tundra soils

S. C. Whalen & W. S. Reeburgh

Institute of Marine Science, University of Alaska, Fairbanks, Alaska 99775-1080, USA

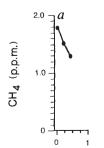
EMISSION of methane from tundra soil contributes about 10% of the global atmospheric methane budget¹. Moreover, tundra soils contain 15% of global soil carbon², so the response of this large carbon reservoir to projected global warming^{3,4} could be important. Coupled biological models³⁻⁶ predict that a warmer climate will ⁶ predict that a warmer climate will methane emission through increased rates methanogenesis. Microbial oxidation of methane is, however, a possible control on emissions that has previously been overlooked. Here we report the results of field and laboratory experiments on methane consumption by tundra soils. For methane concentrations ranging from below to well above ambient, moist soils were found to consume methane rapidly; in non-waterlogged soils, equilibration with atmospheric methane was fast relative to microbial oxidation. We conclude that lowering of the water table in tundra as a result of a warmer, drier climate will decrease methane fluxes and could cause these areas to provide a negative feedback for atmospheric methane.

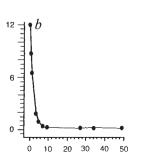
Methane is one of several radiatively active trace gases undergoing an atmospheric concentration increase of 1% yr⁻¹ or more^{7,8}, and has the potential to cause climate change through atmospheric warming⁹⁻¹¹. Global methane budgets balance net

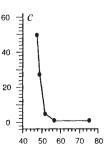
CH₄ sources to the atmosphere against the atmospheric photochemical sink, using ¹⁴C and ¹³C contents, and isotope fractionation factors as constraints^{1,12} on the source terms. Other methane sinks (consumption by soils and marine sediments) are incorporated in the net source terms and are not explicitly identified in these budgets. Some climate models incorporating greenhouse gas increases predict higher temperatures and drier summer conditions for high northern latitudes 13-15. Coupled biological models predict increased CH₄ emission as a result of increased rates of methanogenesis³⁻⁶, but CH₄ oxidation was neglected in these models. Net consumption of atmospheric CH₄ has been reported in some soils of temperature forests¹⁶⁻¹⁸ and swamps¹⁹, tropical forests^{18,20-22}, savannah²³ tundra^{24,25}. The areal extent of the soil CH₄ sink is poorly understood, but it is estimated to account for <1-58 Tg yr or up to 11% of the global photochemical sink 16,17,21

Our observations were made in a moist tundra meadow on Unalaska Island, Aleutian Islands, adjacent to Skan Bay (53° N, 167° W). Plant cover was a continuous mat of mosses invaded by lichens, cottongrasses (Eriophorum sp.), low-lying ferns and dwarf shrubs (Vaccinium sp). The air temperature ranged between 5 and 8 °C; the soil temperature was 7 °C and uniform in the upper 15 cm.

We collected syringe samples of gas from three static chambers at 0.25-h intervals and analysed them by gas chromatography²⁶ on board the RV Alpha Helix. The chambers were fitted with a capillary bleed to equalize pressure following sampling. Chamber measurements vield net fluxes-in the measurements reported here, the methane concentration decreased with time, indicating that consumption exceeded production in the moist







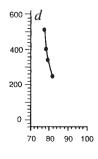
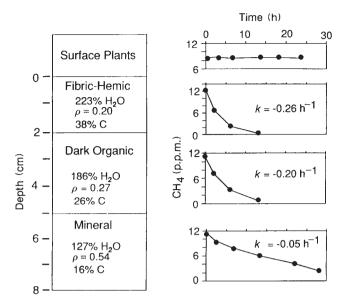


FIG. 1 Consumption of headspace methane in a field experiment using a static chamber. a, consumption of atmospheric CH4 to sub-ambient levels on 8 October 1987. b-d, Results of subsequent experiments (9-12 October 1987) in which the headspace of the same chamber was adjusted to initial CH4 concentrations of 10, 50 550 p.p.m., respectively. Chamber temperatures differed from ambient temperatures by <2 °C during the experiment.

FIG. 2 Soil characteristics and depth distribution of CH₄ consumption in a soil core. Maximum consumption rates occur in the top 5 cm of the core. No CH₄ was consumed by the vegetation. Water content ([wet weight/dry weight] -1), organic content ([loss on ignition]/[dry weight]) and bulk density (ρ represents [oven-dried mass]/[field volume] in g cm $^{-3}$) are given for each of the soil units.



soils studied. In all cases, the concentrations eventually level off to a small but non-zero value. Figure 1 shows the results of an experiment on one of two chambers where atmospheric methane (\sim 1.7 p.p.m.) was consumed, and subsequent experiments where the chamber headspace was adjusted to initial CH₄ concentrations of 10, 50 and 500 p.p.m. by injection of millilitre quantities of a 95% Ar: 5% CH₄ mixture. Headspace concentrations in chambers with 10 p.p.m. CH₄ became sub-ambient in 4–7 h and decreased to 0.14 p.p.m. in as little as 8 h.

Soil cores from the same site were collected in plastic tubes (6.7-cm inner diameter, 2-mm wall) for laboratory studies of CH₄ oxidation and the depth distribution of CH₄ oxidizing potential. We avoided sample disturbance by cutting around the tube perimeters with a serrated knife as the tubes were inserted. The cores collected were 2-12 cm long, and showed a 2-cm fibric-hemic layer below the surface vegetation followed by a 3-cm dark organic-rich layer that graded into a weathered mineral horizon extending below the sample (Fig. 2). These cores were capped at the base and were placed in ~1-1 Mason jars, the lids of which were equipped with valves and a septum. The jar headspaces were maintained at constant pressure by introducing ambient air following sample removal; this headspace dilution was negligible. Because only the soil surface was in contact with the jar atmosphere, the cores in jars were physically equivalent to the field chambers. The ratios of headspace volume to soil surface area for the chambers and jars agreed to within 5%, so CH₄ uptake kinetics for both are comparable. The response of whole cores to CH4 additions at ambient air temperatures was repeatable and identical to the field chambersthe first-order rate constants for methane consumption in both were $\sim -0.25 \, h^{-1}$ and headspace methane concentrations as low as 0.7 p.p.m. were observed in 6-13 h.

Methane consumption by core segments representing the various soil horizons was measured in jar experiments using initial headspace CH_4 concentrations of $\sim \! 10 \, \mathrm{p.p.m.}$ Microlitre quantities of the same $Ar: CH_4$ mixture that was used in the chamber experiments were added to the jars. The results are shown in Fig. 2. The surface vegetation consumed no CH_4 . Methane was consumed by all soil layers, with the highest rates in the 0-2 cm and 2-5 cm layers. A portion of each core segment was autoclaved; these samples consumed no CH_4 in 24 h, indicating that CH_4 consumption is biologically mediated.

We performed a tracer experiment to determine the products of methane oxidation and their distribution in soils. Two cores with nearly identical methane uptake kinetics (cores C and E) were used in this experiment, one to follow CH₄ decreases and the other to follow changes in headspace ¹⁴CH₄ and ¹⁴CO₂. The experiment was conducted in the dark to prevent photosynthetic uptake of ¹⁴CO₂. The headspaces of the jars containing both cores were adjusted to an initial CH₄ concentration of \sim 10 p.p.m., but stable CH₄ was added to core C whereas core E received CH₄ containing 14 CH₄ (2.8 × 10⁸ d.p.m. cm⁻³; d.p.m., disintegrations per minute). Core E was sampled at time intervals based on decreases in the stable CH₄ concentration of core C. Headspace samples (10 cm³) were injected into a metal and glass stripping/oxidation line²⁷ and were carried in a helium flow through bubbler traps and a quartz combustion tube filled with CuO heated to 800 °C. The ¹⁴CO₂ was trapped in 1N NaOH before combustion and the 14CH4 was trapped in Woeller's solution²⁸ as ¹⁴CO₂ following combustion. The jar headspaces were maintained at constant pressure by introducing ambient air following sample removal. The results are corrected for dilution (1.6% per sample). The experiment was terminated after 11 h when the CH₄ concentration reached 0.4 p.p.m. and 95% of the added ¹⁴CH₄ had been consumed. The remaining head-space ¹⁴CH₄ and ¹⁴CO₂ was recovered with a series of helium flushes; the jar and core were frozen and the ¹⁴C incorporated in the core was assayed by dry combustion of freeze-dried homogenized samples of core segments.

Figure 3 shows the results of the tracer experiment. Core C showed a decrease in headspace CH₄ that parallels the ¹⁴CH₄ decrease in core E. The headspace ¹⁴CO₂ activity indicated that part of the consumed ¹⁴CH₄ was retained by the core. We accounted for 96% of the ¹⁴CH₄ added to core E; 46% of this was respired as ¹⁴CO₂ and 54% was recovered by dry combustion. The fraction recovered by dry combustion represents ¹⁴CH₄ assimilated into microbial biomass, organic matter and inorganic matter. We made no attempt to independently assay the inorganic fraction; these soils are carbonate-free and soil pH values of 5.7 suggest that this fraction is small.

Interpretation of experiments involving amended atmosphere requires an understanding of the timescale of equilibration between the soil and headspace for added gases. The equilibration time was evaluated for core E with a relaxation technique²⁹ that involved inhibiting methane oxidation and following equilibration of added CH₄. Acetylene, a well-known inhibitor of CH₄ oxidation³⁰, was added to the headspace (30 p.p.m. final concentration) before sectioning core E for freeze-drying and dry combustion. Methane was then added and headspace concentrations were measured at 1-min intervals until the changes could not be resolved (6 min). This approach shows that equilibration is a two-step process: a fast $(k = -1.54 \, \text{min}^{-1})$

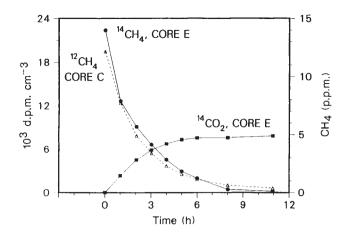


FIG. 3 14CH₄ oxidation as a function of time. This jar experiment was conducted on 12 October 1987, using two cores with almost identical CH₄ oxidizing characteristics. The headspace of core C was amended with CH4 and was used to guide sampling core E, which was amended with 14CH₄.

process, presumably involving the porous surface material or large voids, followed by a slower $(k = -0.75 \text{ min}^{-1})$ process involving more compact material or adjacent smaller voids. These first-order equilibration or relaxation constants show that equilibration is much faster than biological consumption (k = $-0.004 \,\mathrm{min}^{-1}$). Thus, CH₄ consumption is not controlled by transport in tundra soils. The initial samples in the chamber, jar and tracer experiments (taken 15 min after adjusting initial concentrations in the chamber and jar experiments; 5 min after tracer addition) contain no bias owing to incomplete equilibration.

The ability of these soils to respond to methane increases without a lag and to consume a wide (thousand-fold) range of concentrations with no indication of saturation (all follow firstorder consumption kinetics) indicates that they could be an important sink. These methane consumption measurements were made at the low end (7 °C) of the reported temperature range (2-30 °C). Methane consumption rates of 2.7 mg m⁻² d⁻¹ for our unamended chambers lie in the middle of reported ranges (0.2-4.2 mg m⁻² d⁻¹)¹⁶⁻²⁵, indicating that methane oxidation can be important in cold soils. Final methane concentrations as low as those observed in our chamber and jar experiments (0.1-0.4 p.p.m.) have not been reported in previous chamber experiments, but similar concentrations have been observed in soils¹⁶. Static chambers are rarely allowed to 'run down', so the high final concentrations in previous studies can be expected when static chambers are used for only short periods to avoid disturbing sub-surface gradients³¹. Our results demonstrate that soil oxidation of atmospheric CH₄ is microbially mediated and point to a population capable of oxidizing CH₄ at concentrations ten times lower than ambient atmospheric concentrations. Kinetic properties of cultured methanotrophs are not consistent with growth using atmospheric CH₄ as the sole carbon source, implying a threshold for growth well above atmospheric levels 32. Mixotrophic growth, co-oxidation by non-methanotrophs such as ammonium-oxidizing bacteria, or induction of high-methaneaffinity enzyme systems are processes that may be important in maintaining these low CH₄ concentrations 30,32,33

These measurements have implications important to tundra systems under the projected warmer drier conditions. The subsurface CH₄ oxidizing activity is important in controlling upward CH₄ fluxes at sites where vascular plants are absent. Vascular plants transport CH₄ and effectively bypass the CH₄ oxidizing zone. Tussock and low-shrub tundra (35-50% vascular plant cover) are expected to show larger consumption effects than wet meadow tundra (80-90% vascular plant cover)²⁶. Our previous observations at relatively wet permanent sites have not shown net oxidation²⁶. Methane fluxes from tundra³⁴ and swamp¹⁹ sites, however, are positively correlated with the level of the water table, and the 20% of the stations occupied during

a high-latitude transect study²⁴, all with a lowered water table, had zero or negative CH4 fluxes.

Consumption of atmospheric CH₄ by soils depends on transport to zones of consuming activity. Transport in waterlogged tundra soils is by aqueous molecular diffusion. Waterlogged tundra sites have a methane consumption maximum centred near the water table (the oxic/anoxic boundary), sustained by upward diffusion of dissolved CH₄. By contrast, transport in dry tundra soils is by gas-phase diffusion (10⁵ times faster than aqueous), so porous low-resistance tundra soils do not limit the supply of methane. Expected consequences of atmospheric warming (such as a lowered water table, increased seasonal thaw depth, or permafrost melting) will increase the vertical extent of oxidized tundra soil and enhance oxidation of CH₄. Our measurements demonstrate that the oxidation of atmospheric CH₄ occurs in oxic soils and is not limited by diffusion in non-waterlogged soils, so a negative feedback on atmospheric CH₄ concentration is possible. Furthermore, the CO₂ produced by oxidizing CH₄ is 20 times less effective as a greenhouse gas8.

Received 29 January; accepted 3 May 1990

- Cicerone, R. J. & Oremland, R. S. Global biogeochem. Cycles 2, 299-327 (1988)
- Post, W. M., Emanuel, W. R., Zinke, P. J. & Stangenberger, A. J. Nature 298, 156-159 (1982).
- Khalil, M. A. K. & Rasmussen, R. A. Tellus B41, 554-559 (1989)
- Lashof, D. A. Clim. Change **14**, 213-242 (1989). Guthrie P. D. J. geophys. Res. **91**, 10847-10851 (1986).
- Hameed, S. & Cess, R. D. Tellus B35, 1-7 (1983)
- Steele, L. P. et al. J. atmos. Chem. 5, 125-171 (1987) Blake, D. R. & Rowland, F. S. Science 239, 1129-1131 (1988).
- Dickinson, R. E. & Cicerone, R. J. Nature 319, 109-115 (1986).
- Mitchell, J. F. B. Rev. Geophys. 27, 115-139 (1989)
- Ramanathan, V. et al. Rev. Geophys. 25, 1441-1482 (1987).
- Enhalt, D. Tellus 26, 59-70 (1974).
- Manabe, S. & Wetherald, R. T. J. atmos. Chem. 44, 1211-1235 (1987).
 Schlesinger, M. E. & Zhao, Z-C. J. Clim. 2, 459-495 (1989).
- Wilson, C. A. & Mitchell, F. B. J. geophys. Res. 92, 13315-13343 (1987).
- Born, M., Dörr, H. & Levin, I. Tellus B42, 2-8 (1990).
- Steudler, P. A., Bowden, R. D., Melillo, J. M. & Aber, J. D. Nature 341, 314-316 (1989).
- Keller, M., Goreau, T. J., Wofsey, S. C., Kaplan, W. A. & McElroy, M. B. Geophys. Res. Lett. 12, 1156-1159 (1983)
- Harriss, R. C., Sebacher, D. I. & Day, F. P. Nature 297, 673-674
- Goreau, T. J. & de Mello, W. Z. Ambio 17, 274-281 (1988). Seiler, W. & Conrad, R. in Geophysiology of the Amazonia: Vegetation and Climate Interactions
- (ed. Dickinson, R. E.) 133-162 (Wiley, New York, 1987). Keller, M., Kaplan, W. A. & Wofsey, S. C. *J. geophys. Res.* **91**, 11791–11802 (1986). Seiler, W., Conrad, R. & Scharffe, D. *J. atmos. Chem.* **1**, 171–186 (1984).
- Whalen, S., C. & Reeburgh, W. S. Tellus B42, 237-249 (1990). King, S. L., Quay, P. D. & Lansdown, J. M. J. geophys. Res. 94, 18273-18277 (1989).
- Whalen, S. C. & Reeburgh, W. S. Global biogeochem. Cycles 2, 399-408 (1988).
- Alperin, M. J. & Reeburgh, W. S. Appl. envir. Microbiol. 50, 940-945 (1985).
- Woeller, F. H. Analyt. Biochem. 2, 508-511 (1961).
- Sparks, D. L. Kinetics of Soil Chemical Processes (Academic, San Diego, 1989).
- Bédard, C. & Knowles, R. Microbiol. Rev. 53, 68-84 (1989)
- 31. Hutchinson, G. L. & Mosier, A. R. Soil Sci. Soc. Am. J. 45, 311-316 (1981).
- Conrad, R. in Current Perspectives in Microbial Ecology (eds Klug, M. J. & Reddy, C. A.) 461-467 (Am. Soc. Microbiol., Washington, DC (1984)
- Ward, B. B. Arch. Mikrobiol. 147, 126-133 (1987).
- bacher, D. I., Harriss, R. C., Bartlett, K. B., Sebacher, S. M. & Grice, S. S. Tellus B38, 1-10 (1986).

ACKNOWLEDGEMENTS. This work was supported by the NSF and NASA.