Title
Effect of nitrogen fertilization on atmospheric methane oxidation in boreal forest soils

Journal
Chemosphere - Global Change Science, 2(2)

ISSN
1465-9972

Authors
Whalen, SC
Reeburgh, WS

Publication Date
2000-04-01

DOI
10.1016/s1465-9972(00)00003-9

PERGAMON

Chemosphere – Global Change Science 2 (2000) 151–155

CHEMOSPHERE – GLOBAL CHANGE SCIENCE

Effect of nitrogen fertilization on atmospheric methane oxidation in boreal forest soils

S.C. Whalen *, W.S. Reeburgh

Department of Earth System Science, University of California, Irvine, CA 92717-3100, USA

Received 7 May 1999; accepted 9 September 1999

Importance of this Paper: The "greenhouse" gas CH_4 is increasing in atmospheric concentration. Oxidation by methanotrophic bacteria in upland soils is the only biological sink for atmospheric CH_4. Methanotrophs are inhibited by high soil N, raising concern that the contemporary worldwide increase in atmospheric N deposition will decrease soil CH_4 oxidation. The boreal forest occupies 13% of the earth's continental surface and is important in atmospheric CH_4 oxidation. We studied atmospheric CH_4 oxidation in fertilized boreal forest plots to determine the N sensitivity of the methanotrophic community and conclude that increased N deposition will not alter the rates of CH_4 oxidation in these soils.

Abstract

Field plots of aspen and black spruce in the Alaskan boreal forest were fertilized repeatedly with nitrogen during the 1993 summer growing season, and weekly determinations of the influence of fertilization on atmospheric CH_4 oxidation were made with static chambers. Repeated fertilization with $(NH_4)_2SO_4$ solution or nutrient media used to culture methanotrophic or nitrifying bacteria gave a total addition of 140 or 580 kg N ha^{-1}. Time-integrated CH_4 oxidation was not significantly different in fertilized soils versus watered controls because CH_4 oxidation was localized in a subsurface soil zone that was probably not penetrated by surface-applied aqueous phase fertilizer. Insensitivity of CH_4 oxidation by these soils to a high rate of N fertilization and the low current rate of atmospheric N deposition suggest that future increases in atmospheric N deposition will not alter the sink strength of high latitude boreal forest soils in the atmospheric CH_4 budget.

Keywords: Methane oxidation; Forests; Nitrogen fertilization

1. Introduction

The atmospheric concentration of the radiatively important greenhouse gas CH_4 has shown a well-documented, contemporary increase (Houghton et al., 1996). Methane is second only to CO_2 as a greenhouse gas and contributes approximately 20% to global warming (Bouwman, 1990). Microbial oxidation in well-drained soils is the only identified biological sink for atmospheric CH_4 and accounts for 3% to 9% of total annual atmospheric CH_4 destruction (Prather et al., 1996). This is similar in magnitude to the current atmospheric increase of 37 Tg CH_4 yr^{-1} (Houghton et al., 1996). Consequently, alterations of the soil sink strength are a significant determinant of the rate of change in the atmospheric CH_4 concentration (Prather et al., 1996) and absence of this sink will cause the atmospheric CH_4 concentration to increase at 1.5 times the current rate (Duxbury, 1994).

Methanotrophic bacteria are generally considered to be responsible for atmospheric CH_4 oxidation (Conrad,

* Corresponding author. Present address: Department of Environmental Science and Engineering, CB #7400, University of North Carolina, Chapel Hill, NC 27599-7400, USA. Fax: +1-919-966-7911.

E-mail address: steve_whalen@unc.edu (S.C. Whalen).

1465-9972/00/$ - see front matter
PII: S 1 4 6 5 - 9 9 7 2 (0 0) 0 0 0 0 3 - 9

1996; King, 1997), although NH_4-oxidizing bacteria are capable of oxidizing CH_4 at rates lower than methanotrophs (Jones et al., 1984; Bedard and Knowles, 1989). Hence, soils with high nitrification rates show low CH_4 oxidation because CH_4 oxidation is dominated by nitrifiers (Castro et al., 1995; Steudler et al., 1996).

Influences on rates of atmospheric CH_4 oxidation in well-drained soils include soil temperature, moisture and N status (reviewed by King, 1992, 1997; Mancinelli, 1995) as well as land use patterns (Ojima et al., 1993; Willison et al., 1995; Hudgens and Yavitt, 1997; Boecyx et al., 1998). Forest plots experimentally fertilized with N frequently show depressed atmospheric CH_4 oxidation relative to unfertilized controls (Steudler et al., 1989; King and Schnell, 1994; Sitaula et al., 1995). Consequently, the well-documented worldwide increase in atmospheric N deposition on terrestrial and aquatic environments (Matthews, 1994; Galloway et al., 1995) may result in a decrease in atmospheric CH_4 oxidation by upland forest soils (Castro et al., 1995).

The boreal forest occupies 13% of the earth's continental surface (Schultz, 1995), and upland boreal forest soils are important in atmospheric CH_4 consumption (Whalen et al., 1991). Agriculture is expanding worldwide (Adams et al., 1990) and agricultural soils have lower area-based rates of CH_4 consumption than aerated forests, shrublands and savannah (reviewed by King, 1997). Since boreal forests are generally unsuitable for this land use, upland soils of this region will be critical in slowing future increases in atmospheric CH_4. Accurate prediction of future trends in soil CH_4 oxidation in these high latitude soils requires an improved understanding of the microbial community response to global change, including increased atmospheric N deposition.

This study was aimed at determining the impact of increased N input on atmospheric CH_4 oxidation by boreal forest soils and to qualitatively determine the relative importance of methanotrophs and nitrifiers in oxidizing CH_4 in these soils. We experimentally fertilized plots with $(NH_4)SO_4$ solution to simulate increased atmospheric N input. Further, we attempted to enhance populations of nitrifiers or methanotrophs in additional plots by fertilizing with liquid media used to culture these microbial groups. The rate of CH_4 oxidation in treated plots was compared with that of watered controls.

2. Materials and methods

2.1. Site description

Field sites were located along a successional sequence in Bonanza Creek experimental forest (64°N, 148°W) near Fairbanks, Alaska. The intermediate successional stage was represented by a south-facing aspen (*Populus tremuloides*; site AS2) community with a 3–5 cm floor of leaf litter and an insignificant understory. The advanced successional stage was represented by a north-facing black spruce (*Picea mariana*; site BS2) stand with a continuous ground cover of feather mosses (*Pleurozium* spp. and *Hylocomium* spp.) invaded by lowbush cranberry (*Vaccinium vitis-idaea*) and lichens. The organic horizon extends to about 10 cm. These sites have no soil zone of methanogenesis and are atmospheric CH_4 sinks (Whalen et al., 1992).

2.2. Experimental

Methane flux determinations were made using the static chamber technique (Whalen et al., 1991). Each chamber consisted of a skirted aluminum base permanently seated in the soil and a removable aluminum lid that utilized a water-filled channel for a seal. Lids were equipped with an o-seal fitting to allow syringe sampling of headspace gas and fitted with a capillary bleed to equilibrate with atmospheric pressure. Eight bases were deployed in June 1993 at each site. Methane fluxes were determined at each chamber on 8 July (Calendar day (CD) 189). Soils enclosed within two randomly selected bases at each site were then amended with 2-L of one of the following: (a) a growth medium for methanogens (King and Adamsen, 1992); (b) a growth medium for nitrifiers (De Boer et al., 1991); (c) $(NH_4)_2SO_4$ in deionized water; or (d) deionized water only. Methane oxidation was measured weekly from CD 194 through CD 243. Fertilization with the assigned treatment immediately followed each CH_4 flux determination. Fertilization added 73 kg NO_3^--N ha^{-1} (methanogen medium) or 18 kg NH_4^+-N (nitrifier medium and $(NH_4)_2SO_4$ treatment) to these soils on a weekly basis.

2.3. Methane analysis

Methane determinations were made by flame ionization detection gas chromatography with a precision of <1% (Whalen et al., 1991). Calibration gases are relatable to mixtures obtained from the National Institute of Standards and Technology.

2.4. Statistical analyses and calculations

Area-based rates of CH_4 oxidation were calculated from the time-linear decrease in CH_4 concentration in the chamber headspace during a 0.75 h deployment. Day rates of CH_4 oxidation for each chamber were time-integrated over the experimental period and treatment means within each site were compared by single factor analysis of variance. Data satisfied assumptions of normality and homoscedasticity without transformation. A significance level of $\alpha = 0.05$ was used.

3. Results and discussion

Day-rates of CH_4 oxidation at AS2 varied over a factor of about 1.6 (0.46–0.76 mg m^{-2} d^{-1}) during the study period (Fig. 1). Time-integrated rates of CH_4 oxidation varied from 29.2 to 32.3 mg m^{-2} and were not significantly different across treatments. Day-rates of CH_4 oxidation at BS2 were somewhat higher and less variable than for AS2 (Fig. 1). Methane oxidation rates varied over a factor of 1.3, from 0.69 to 0.89 mg m^{-2}. Time-integrated rates of CH_4 oxidation varied from 36.8 to 40.1 mg m^{-2} and were not significantly different across treatments. The temporal pattern of CH_4 oxidation was generally similar across treatments within each site. This probably reflects changes in soil moisture, which controls diffusion of CH_4 to the zone of oxidation, and hence is a primary determinant of rates of atmospheric CH_4 oxidation by soils (King, 1997). Increased N input to forests can increase the fine root biomass (Magill et al., 1997), which may decrease air-filled porosity and limit rates of diffusion and atmospheric CH_4 oxidation. Any increase in fine root biomass in response to fertilization here had no impact on soil–atmosphere gas exchange, as time-integrated rates of CH_4 oxidation did not differ significantly between control and fertilized plots at either site. The generally higher rates of CH_4 oxidation at BS2 than at AS2 is consistent with the higher gas-filled porosity at BS2 (Whalen et al., 1992), which allows for more rapid diffusion of atmospheric CH_4 into the soil.

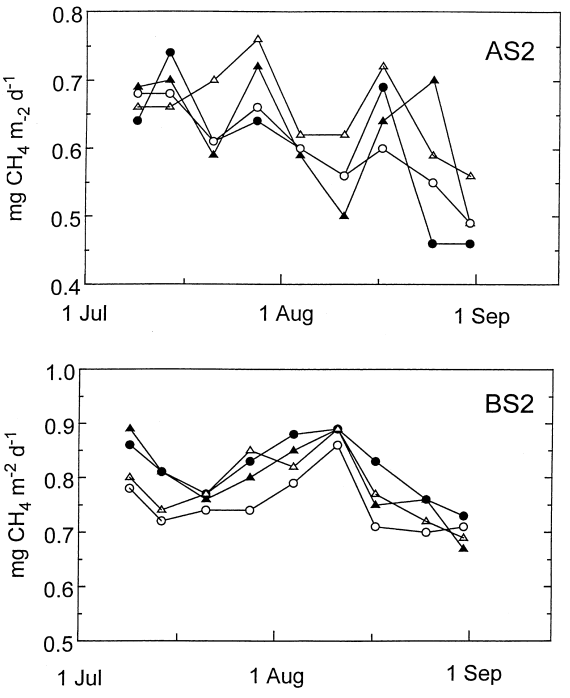

Fig. 1. Methane oxidation at sites AS2 and BS2 in response to fertilization with nitrifier medium (▲), methanotroph medium (○), $(NH_4)_2SO_4$ solution (△) or deionized water (●). Each point represents the mean rate of CH_4 oxidation from duplicate chambers for each treatment. Standard deviations averaged 0.06 and 0.17 mg CH_4 m^{-2} d^{-1} at AS2 and BS2, respectively. Error bars are eliminated for clarity.

The N load from repeated fertilization totaled about 140 kg NH_4^+-N ha^{-1} for plots treated with $(NH_4)_2SO_4$ and the nitrifier medium and about 580 kg NO_3^--N ha^{-1} for plots fertilized with the methanotroph medium. Atmospheric CH_4 oxidation at both sites was unaffected by fertilization at these levels, which is consistent with a similar observation for a boreal spruce forest fertilized for three years with NH_4NO_3 at 60 kg N ha^{-1} (Gulledge et al., 1997). However, most studies report inhibition of atmospheric CH_4 oxidation in N-fertilized forest soils. Pine forests fertilized with NH_4NO_3 at 30 and 90 kg N ha^{-1} gave CH_4 oxidation rates that were 85% and 62%, respectively, of unfertilized controls (Sitaula et al., 1995), while a temperate softwood forest amended with the same fertilizer at 120 kg N ha^{-1} showed a CH_4 oxidation rates that were 67% of unfertilized controls (Steudler et al., 1989). Urea fertilization of a pine plantation (180 kg N ha^{-1}) resulted in a 5- to 20-fold decrease in atmospheric CH_4 oxidation relative to controls (Castro et al., 1995), while application of urea, KNO_3 or NH_4Cl to forested peatland soils at 100 kg N ha^{-1} significantly depressed CH_4 oxidation (Crill et al., 1994). Finally, fertilization of a mixed oak–pine forest with only 10 kg NH_4Cl-N ha^{-1} inhibited atmospheric CH_4 oxidation by about 20% relative to control plots (King and Schnell, 1994).

Sensitivity of atmospheric CH_4 oxidation in forest soils to N fertilization may relate to the position of the CH_4 oxidizing community in the soil column. Maximum rates of atmospheric CH_4 oxidation are found 10–20 cm below the soil surface at both sites, and 91% and 98% of depth-integrated CH_4 oxidation at AS2 and BS2 occurs at soil depths >10 cm (Whalen et al., 1992). Hence, it is likely that liquid fertilizer additions did not penetrate to the active CH_4 oxidizing zone. Other studies also point to the locus of the CH_4 oxidizing zone as a key determinant of soil sensitivity to N fertilization. Surface localization of fertilizer in conjunction with a subsurface zone of CH_4 oxidation was offered as a possible explanation for the lack of a fertilizer effect on CH_4 oxidation in a humisol amended with 100 kg ha^{-1} urea-N (Dunfield et al., 1995), while addition of NH_4Cl, KNO_3 and urea-N at this same level all inhibited CH_4 oxidation in forest peat soils where CH_4 oxidation was largely restricted to the 0–5 cm zone.

Nitrifiers are frequently located in the organic, NH_4^+-rich surface layers of forest soils (Lensi et al., 1991; Schnell and King, 1994; Castro et al., 1995), suggesting that the NH_4^+-N and nitrifier medium solutions

contacted this component of the microbial community. Methane is co-oxidized along with NH_4^+-N by nitrifying bacteria (Bedard and Knowles, 1989) and urea-N fertilization (rapidly hydrolyzed to NH_4^+-N; Zhengping et al., 1996) in pine forest soil has been demonstrated to reduce atmospheric CH_4 oxidation by shifting the relative importance of methanotrophs and nitrifiers (Castro et al., 1995). Hence, insensitivity of CH_4 oxidation at AS2 and BS2 to amendment with NH_4^+-N or the nitrifier medium suggests that methanotrophs are primarily responsible for CH_4 oxidation in these soils. This is consistent with the observation that nitrifying activity is low in other boreal forest soils (Gulledge et al., 1997). In contrast to the nitrifier medium, the methanotroph medium probably never reached the target organisms. Hence, the rate of CH_4 oxidation was uninfluenced by this treatment.

Increased atmospheric N deposition is not likely to reduce rates of atmospheric CH_4 oxidation in boreal forest soils. The loading rate of 140 kg N ha^{-1} for repeated fertilization with NH_4^+-N solution or nitrifier medium is 70-fold higher than the estimated annual deposition (wet plus dry) of 2 kg N ha^{-1} for remote continental areas (Galloway, 1993). Moreover, experimental conditions were considerably more favorable than natural rainfall for delivery of nutrients to the active methanotrophic zone located at >10 cm below the soil surface. Each of the eight fertilizations simulated a 2.7 cm, high N rainfall while total annual precipitation at these study sites is 30 cm equally divided between rain and snow, and no single rain event exceeded 2 cm during the year this study was conducted (NOAA, 1993). Additionally, chamber bases extended to a depth of about 15 cm below the soil surface, ensuring that fertilizer did not move laterally following application. In concert, low annual precipitation and subsurface localization of the zone of atmospheric CH_4 oxidation ensure that any realistic increase in atmospheric N deposition will not alter the sink strength of upland boreal forest soils in the atmospheric CH_4 budget.

Acknowledgements

This work was supported by the US Environmental Protection Agency and the National Institute for Global Environmental Change. The Bonanza Creek Long Term Ecological Research Program is supported by the National Science Foundation. Chad Staiger assisted in laboratory and field analyses.

References

Adams, R.M., Rosenzweig, C., Peart, R.M., Ritchie, J.T., McCarl, B.A., Glyer, J.D., Curry, R.B., Jones, J.W., Boote, K.J., Allen Jr., L.H., 1990. Global climate change and US agriculture. Nature 345, 219–224.

Bedard, C., Knowles, R., 1989. Physiology, biochemistry and specific inhibitors of CH_4, NH_4^+, and CO oxidation by methanotrophs and nitrifiers. Microbiological Reviews 53, 68–84.

Boecyx, P., Van Cleemput, O., Meyer, T., 1998. The influence of land use and pesticides on methane oxidation in some Belgian soils. Biology and Fertility of Soils 27, 293–298.

Bouwman, A.W., 1990. Introduction. In: Bouwman, A.W. (Ed.), Soils and the Greenhouse Effect. Wiley, Chicester, pp. 25–32.

Castro, M.S., Steudler, P.A., Melillo, J.M., Aber, J.D., Bowden, R.D., 1995. Factors controlling atmospheric methane consumption by temperate forest soils. Global Biogeochemical Cycles 9, 1–10.

Conrad, R., 1996. Soil microorganisms as controllers of atmospheric trace gases (H_2, CO, CH_4, OCS, N_2O and NO). Microbiological Reviews 60, 609–640.

Crill, P.M., Martikainen, P.J., Nykánen, H., Silvola, J., 1994. Temperature and N fertilization effects on methane oxidation in a drained peatland soil. Soil Biology and Biochemistry 26, 1331–1339.

De Boer, W., Gunnewiek, P.J.A.K., Veenhuis, M., Bock, E., Laanbroek, H.J., 1991. Nitrification at low pH by aggregated chemolithotrophic bacteria. Applied and Environmental Microbiology 57, 3600–3604.

Dunfield, P.F., Topp, E., Archambault, C., Knowles, R., 1995. Effect of N fertilizers and moisture content on CH_4 and N_2O fluxes in a humisol: measurements in the field and intact soil cores. Biogeochemistry 29, 199–222.

Duxbury, J.M., 1994. The significance of agricultural sources of greenhouse gases. Fertilizer Research 38, 151–163.

Galloway, J.N., 1993. Deposition of sulfur and nitrogen from the remote atmosphere. In: Galloway, J.N., Charlson, R.J., Andreae, M.O., Rodhe, H. (Eds.), The Biogeochemical Cycling of Sulfur and Nitrogen in the Remote Atmosphere. Riedel, Dordrecht, pp. 143–175.

Galloway, J.N., Schlesinger, W.H., Levy, H., Michaels, A., Schnoor, J.L., 1995. Nitrogen fixation: anthropogenic enhancement–environmental response. Global Biogeochemical Cycles 9, 235–252.

Gulledge, J., Doyle, A.P., Schimel, J.P., 1997. Different NH_4^+ inhibition patterns of soil CH_4 consumption: a result of distinct CH_4-oxidizer populations across sites? Soil Biology and Biochemistry 29, 13–21.

Houghton, J.T., Filho, L.G.M., Callandar, B.A., Harriss, N., Kattenberg, A., Maskell, K., 1996. Technical summary. In: Houghton, J.T., Filho, L.G.M., Callandar, B.A., Harriss, N., Kattenberg, A., Maskell, K. (Eds.), Climate Change 1995. The Science of Climate Change. Cambridge University Press, Cambridge, pp. 1–49.

Hudgens, D.E., Yavitt, J.B., 1997. Land-use effects on soil methane and carbon dioxide fluxes in forests near Ithaca, New York. Ecoscience 4, 214–222.

Jones, R.D., Morita, R.Y., Griffiths, R.P., 1984. Method for estimating in situ chemolithotrophic ammonium oxidation using carbon monoxide oxidation. Marine Ecology Progress Series 17, 259–269.

King, G.M., 1992. Ecological aspects of methane oxidation, a key determinant of global methane dynamics. Advances in Microbial Ecology 12, 431–468.

King, G.M., 1997. Responses of atmospheric methane consumption by soils to global climate change. Global Change Biology 3, 351–362.

King, G.M., Adamsen, A.P.S., 1992. Effects of temperature on methane consumption in a forest soil and pure cultures of the methanotroph *Methylomonas rubra*. Applied and Environmental Microbiology 58, 2758–2763.

King, G.M., Schnell, S., 1994. Effect of increasing atmospheric methane concentration on ammonium inhibition of soil methane consumption. Nature 370, 282–284.

Lensi, R., Lescure, C., Clays-Josserand, A., Gourbiere, F., 1991. Spatial distribution of nitrification and denitrification in an acid forest soil. Forest Ecology and Management 44, 29–40.

Magill, A.H., Aber, J.D., Hendricks, J.L., Bowden, R.D., Melillo, J.M., Steudler, P.A., 1997. Biogeochemical response of forest ecosystems to simulated and chronic nitrogen deposition. Ecological Applications 7, 402–415.

Mancinelli, R.L., 1995. The regulation of methane oxidation in soil. Annual Review of Microbiology 49, 581–605.

Matthews, E., 1994. Nitrogenous fertilizers: global distribution of consumption and associated emissions of nitrous oxide and ammonia. Global Biogeochemical Cycles 8, 411–439.

National Oceanographic and Atmospheric Administration (NOAA), 1993. Local climatological data, Fairbanks, Alaska. Annual Summary with Comparative Data. Asheville, NC, USA.

Ojima, D.S., Valentine, D.W., Mosier, A.R., Parton, W.J., Schimel, D.S., 1993. Effect of land use change on methane oxidation in temperate forest and grassland soils. Chemosphere 26, 675–685.

Prather, M., Derwent, R., Ehalt, D., Fraser, P., Sanhueza, E., Zhau, X., 1996. Other trace gases and atmospheric chemistry. In: Houghton, J.T., Filho, L.G.M., Callandar, B.A., Harriss, N., Kattenberg, A., Maskell, K. (Eds.), Climate Change 1995. The Science of Climate Change. Cambridge University Press, Cambridge, pp. 77–126.

Schnell, S., King, G.M., 1994. Mechanistic analysis of ammonium inhibition of atmospheric methane consumption in forest soils. Applied and Environmental Microbiology 60, 3514–3521.

Sitaula, B.K., Bakken, L.R., Abrahamsen, G., 1995. CH_4 uptake by temperate forest soil: effect of N input and soil acidification. Soil Biology and Biochemistry 27, 871–880.

Steudler, P.A., Bowden, R.D., Melillo, J.M., Aber, J.D., 1989. Influence of nitrogen fertilization on methane uptake in temperate forest soils. Nature 341, 314–316.

Steudler, P.A., Jones, R.D., Castro, M.S., Melillo, J.M., Lewis, D.L., 1996. Microbial controls of methane oxidation in temperate forest and agricultural soils. In: Murrell, J.C., Kelly, D.P. (Eds.), Microbiology of Atmospheric Trace Gases. NATO ASI Series, vol. 139, Springer, Berlin, pp. 69–84.

Whalen, S.C., Reeburgh, W.S., Barber, V.A., 1992. Oxidation of methane in boreal forest soils: a comparison of seven measures. Biogeochemistry 16, 181–211.

Whalen, S.C., Reeburgh, W.S., Kiser, K.S., 1991. Methane consumption and emission by taiga. Global Biogeochemical Cycles 5, 261–273.

Willison, T.W., Webster, C.T., Goulding, K.T.W., Powlson, D.S., 1995. Methane oxidation in temperate soils: effects of land use and the chemical form nitrogen fertilizer. Chemosphere 30, 539–546.

Schultz, J., 1995. The Ecozones of the World. Springer, Berlin.

Zhengping, W., Van Kleemput, O., Baert, L., 1996. Movement of urea and its hydrolysis products as influenced by moisture content and urease inhibitors. Biology and Fertility of Soils 22, 101–108.