Title

A pulse-labeling experiment to determine the contribution of recent plant photosynthates to net methane emission in arctic wet sedge tundra

Journal

Soil Biology and Biochemistry, 34(2)

ISSN

0038-0717

Authors

King, JY
Reeburgh, WS

Publication Date

2002-02-01

DOI

10.1016/s0038-0717(01)00164-x

PERGAMON

Soil Biology & Biochemistry 34 (2002) 173–180

Soil Biology & Biochemistry

www.elsevier.com/locate/soilbio

A pulse-labeling experiment to determine the contribution of recent plant photosynthates to net methane emission in arctic wet sedge tundra

J.Y. King*, W.S. Reeburgh

Department of Earth System Science, University of California at Irvine, Irvine, CA 92697-3100, USA

Received 10 October 2000; received in revised form 7 June 2001; accepted 8 August 2001

Abstract

We conducted a ^{14}C pulse-labeling experiment under field conditions to estimate the contribution of recent photosynthates to methane (CH_4) emission in arctic wet sedge tundra dominated by *Carex aquatilis* and *Eriophorum angustifolium*. The average CH_4 emission rate from plant–soil mesocosms in this study was 0.45 g C m^{-2} d^{-1}. Carbon assimilated by plants via photosynthesis during pulse-labeling turned over rapidly and appeared as emitted CH_4 within 24 h. Integration of flux measurements made over a 2-week period shows that the contribution of recent photosynthates to mid-season CH_4 emission is relatively low. Less than 1% of the ^{14}C-labeled carbon dioxide taken up through photosynthesis was emitted as $^{14}CH_4$ during this study.

Keywords: ^{14}C pulse labeling; Photosynthates; Methane emission; Arctic wet sedge tundra

1. Introduction

Wetlands are recognized as the largest natural source of methane (CH_4) to the atmosphere. High-latitude wetlands represent nearly one-third of the source of CH_4 from wetlands and therefore represent an important part of the global CH_4 budget (Reeburgh, 1996). There has been limited success in modeling CH_4 emissions from a variety of ecosystems using only environmental factors. Current research efforts focus on process-based modeling which includes the influence of vegetation on CH_4 emission (Cao et al., 1996; Walter and Heimann, 2000). Radiocarbon measurements of CH_4 suggest that methane is produced primarily from recently-fixed carbon (Wahlen et al., 1989; Martens et al., 1992; Aravena et al., 1993; Chanton et al., 1995). Observed correlations between net ecosystem productivity (NEP) and CH_4 emission (Clymo and Reddaway, 1971; Svensson, 1983; Sebacher et al., 1986; Aselmann and Crutzen, 1989; Moore and Knowles, 1990; Whiting and Chanton, 1993; Klinger et al., 1994; Waddington et al., 1996) suggest that recent plant photosynthates may be an important source of carbon for CH_4 emissions. These findings also suggest that modeling CH_4 emissions based on primary productivity estimates may be useful in a variety of ecosystems. However, very few studies have been published which describe the underlying mechanism for this relationship, and no previous studies relating to the underlying mechanism have focused on arctic wetland ecosystems.

Only a few studies have been done in other ecosystems to examine photosynthate contribution to CH_4 emission. Minoda and Kimura (1994); Minoda et al. (1996) used ^{13}C pulse-labeling experiments in the laboratory to investigate the role of recently-photosynthesized compounds in CH_4 emissions from rice paddy fields. By using emission rates from a parallel set of unlabeled pots, Minoda et al. calculated the percent contribution of assimilated carbon dioxide (CO_2) to the total CH_4 emitted. The percentages of photosynthesized carbon contributing to emitted CH_4 differed in two studies covered a wide range of 13–110% depending on time of season (Minoda and Kimura, 1994) to a range of 3–52% when rice straw was added to the soil (Minoda et al., 1996). Measurements have shown that the contribution of photosynthates to CH_4 emission is lower when rice straw is applied to paddy soil (Chidthaisong and Watanabe, 1997; Kimura, 1997; Watanabe et al., 1998). Dannenberg and Conrad (1999) conducted a ^{14}C pulse-labeling laboratory experiment on rice plant microcosms and found that 3–6% of the ^{14}C label was emitted as CH_4 within 16 days of labeling. Megonigal et al. (1999) performed a ^{14}C pulse-labeling experiment on a single wetland plant, *Orontium*

* Corresponding author. Present address: University of Minnesota, Department of Soil, Water, and Climate, 1991 Upper Buford Circle, St Paul, MN 55108, USA. Tel. + 1-612-625-1244; fax: + 1-612-625-2208.
E-mail address: jyking@lamar.colostate.edu (J.Y. King).

0038-0717/02/$ - see front matter
PII: S0038-0717(01)00164-X

aquaticum, in a growth chamber and determined that emitted $^{14}CH_4$ made up less than 1% of the ^{14}C recovered 17 days after labeling.

These results from rice paddy systems and *O. aquaticum* suggest that carbon turnover occurs quickly and that recent photosynthates can be an important source of carbon for methanogenesis. The results also indicate a need for further investigation of the contribution of photosynthesized carbon to emitted CH_4 in order to constrain the percentages reported by Minoda and Kimura (1994); Minoda et al. (1996) and extend the measurements to other important methanogenic ecosystems and to field conditions.

Here we report the results of a ^{14}C pulse-labeling experiment conducted under field conditions in arctic Alaska. Our objectives were to investigate the movement of carbon through arctic wetland tundra ecosystems and to measure the contribution of recent plant photosynthates to net CH_4 emission under near-in situ conditions.

2. Materials and methods

2.1. Site description and experimental design

The experiment was conducted at Toolik Field Station on the North Slope of Alaska (68°38′ N, 149°39′ W). Plant–soil mesocosms were taken from an area on the edge of Toolik Lake where many measurements of CO_2 exchange and CH_4 emission have been made in the past (King et al., 1998; Verville et al., 1998). A mesocosm is an intact portion of an ecosystem taken out of the ground as a soil core plus above-ground vegetation. The dominant vegetation in the mesocosms was *Carex aquatilis* and *Eriophorum angustifolium*. *Carex aquatilis—Eriophorum angustifolium* communities are representative of wet tundra in the Arctic (Walker et al., 1989). The circumpolar distribution of these species (Hultén, 1968) makes it possible for results of this study to be extrapolated to other areas of the Arctic. Four mesocosms were established; two of them were pulse-labeled with ^{14}C and two of them served as controls. Measurements were also made on two plots in situ for comparison between in situ and mesocosm conditions. Our interest in obtaining high time resolution time series data limited the number of replicate mesocosms we could measure. The soil cores (29 cm diameter, 35 cm height) and associated vegetation were collected from their natural environment in early July using a stainless steel corer designed specifically for this purpose in order to produce equivalently-sized mesocosms and to minimize the inclusion of void spaces. The soil cores were placed in 20 l stainless steel containers of the same dimensions and were allowed to equilibrate in the mesocosm containers for a few days before the pulse-labeling experiment began. The stainless steel containers minimized the amount of diffusion of oxygen and other gases into the soil through the sides of the container. During the study, lake water was added to each mesocosm to keep the water table constant.

The Nuclear Regulatory Commission (NRC) granted permission for a ^{14}C-labeling field experiment in closed containers inside of a fenced enclosure that minimized loss of ^{14}C to the surrounding environment. The mesocosms at Toolik were kept in a fenced cage outdoors and were exposed to natural climate conditions. The average daily minimum air temperature was 8.7°C, and the average daily maximum air temperature was 20.3°C. During the experiment, 6.4 mm of rainfall was recorded. Records of the climate conditions are available from the Arctic LTER database (J. Laundre, pers. comm.; http://ecosystems.mbl.edu/arc). We performed this experiment at Toolik Field Station in order to conduct the experiment under the same natural climate conditions as the mesocosms would experience in situ and to avoid disturbance of the mesocosms caused by transport from the field site to the laboratory.

The mesocosms in their stainless steel containers were placed together in a water jacket cooled by circulation of lake water. The cooling water was not in direct contact with the soil. This cooling system prevented the soil temperatures from exceeding maximum ambient air temperatures and also increased the temperature gradient between the soil and the ambient air. During the experiment, the average air temperature was 15.2°C. The average soil temperature (0–30 cm depth) was 14.9°C in the mesocosms and 6.5°C in situ. This difference is due to a steep gradient in soil temperature close to permafrost which we could not duplicate in the mesocosms. Although soil temperatures were warmer than natural, the lake water-cooling system kept soil temperatures cooler than ambient air temperatures, unlike growth chamber conditions in which soil temperatures often exceed air temperatures because of absorption of incident radiation by the dark soil surface.

2.2. ^{14}C pulse-labeling

We used the technique of ^{14}C pulse-labeling to trace the movement of recently photosynthesized carbon. Two mesocosms (referred to as mesocosms 3 and 4) were pulse-labeled with $^{14}CO_2$. The stainless steel mesocosm containers were designed with a channel at the top to provide a water seal for the clear Plexiglas headspace chamber (32 l) used for labeling and for CO_2 and CH_4 gas exchange measurements. The ^{14}C label (14.7 MBq from sodium bicarbonate solution (2.1 GBq/mmol; ICN Radiochemicals, Irvine, CA)) was added as $^{14}CO_2$ to the headspace chamber sealed to each mesocosm. The $^{14}CO_2$ was taken up by the plants through photosynthesis, and additional unlabeled CO_2 was added as necessary to maintain the CO_2 mixing ratio inside the chamber within the range of atmospheric levels (between 300 and 400 ppmv). Each mesocosm was labeled for 2 h, which ensured sufficient ^{14}C-label uptake. After 2 h, during which time unlabeled CO_2 had been added several times to replenish assimilated

CO_2, the chamber was removed, and the plants continued to grow under natural conditions. Syringe samples taken from the chamber headspace at the end of the labeling period for each mesocosm showed that the amount of ^{14}C remaining in the chamber was less than 1% of the ^{14}C initially added. Two additional mesocosms (1 and 2) were kept as controls alongside the two labeled mesocosms in the outdoor enclosure. Two sites (5 and 6) were established in the field as in situ controls.

2.3. Gas exchange and soil porewater measurements and analysis

We traced the movement of the ^{14}C label in the plant–soil mesocosms by taking frequent gas flux and soil porewater measurements. A chamber was temporarily placed over each mesocosm for gas flux measurements. A portable CO_2 analyzer (LI-6200, LI-COR, Inc., Lincoln, Nebraska) was used to make measurements of net ecosystem production (NEP) and respiration over a period of 3 min. Following the CO_2 exchange measurements, a reflective cloth was placed over the chamber to minimize temperature increases inside the chamber during the CH_4 flux measurement. Previous measurements in this ecosystem have shown that shading does not affect CH_4 flux rates (King, unpublished data). A series of four headspace syringe samples taken over a 30-min period from the chamber headspace was analyzed at the field station by gas chromatography and used to determine net CH_4 emission rate (Whalen and Reeburgh, 1988). A headspace gas sample collected in a Tedlar bag at the end of the gas flux measurements was analyzed for ^{14}C activity in CO_2 and CH_4. These measurements reflected net CH_4 emission and total ecosystem respiration.

Soil porewater samples were collected using stainless steel probes, made from 0.3 cm diameter stainless steel tubing and perforated at one end, inserted into the soil. Samples integrated for the whole soil profile were collected by sampling equal volumes of soil porewater at four different depths (approximately 5, 10, 15 and 20 cm depths). Individual samples were also collected at each depth to obtain measurements of porewater carbon pools in the soil profile. The porewater samples were extracted through equilibration with a nitrogen gas headspace for dissolved CH_4 and dissolved CO_2, which includes $CO_{2(aq)}$, carbonate, and bicarbonate (Kling et al., 2000). The remaining water sample was filtered and analyzed for dissolved organic carbon (DOC) on a total organic carbon analyzer (TOC-5000, Shimadzu Corp., Kyoto, Japan). Gas samples were analyzed for CH_4 on a gas chromatograph (GC-8A, Shimadzu Corp.) equipped with a flame ionization detector and a 1-m molecular sieve 5A column. Gas samples from the porewater extractions were analyzed for CO_2 on a gas chromatograph (GC-8A, Shimadzu Corp.) with a thermal conductivity detector and Porapak N column and for CH_4 on a gas chromatograph as described above. Gas standards relatable to National Institute of Standards and Technology (NIST) standards were used to calibrate the gas chromatographs.

To analyze the gas samples for ^{14}C we separated CO_2 and CH_4 on an oxidation line (Whalen and Reeburgh, 1990; King, 1999). Each air sample first passed through a sodium hydroxide solution (1 M NaOH, 15 ml) which trapped CO_2. The air sample then passed through a combustion tube which oxidized CH_4 to CO_2. The CO_2 produced from oxidation of the air sample was trapped in a second 1 M NaOH solution (15 ml). A subsample of each NaOH solution was combined with scintillation cocktail (Cytoscint, ICN Biomedicals, Inc., Costa Mesa, California) for analysis of ^{14}C activity. Porewater DOC samples were directly combined with scintillation cocktail and analyzed for ^{14}C activity. All samples were analyzed for ^{14}C activity by liquid scintillation spectroscopy (Rackbeta 1215, LKB/Wallac, Sweden; LS 3801, Beckman Instruments, Inc., Fullerton, California). Background and ^{14}C standards were analyzed with the samples, and corrections were also made for quenching of counts caused by the mixture of NaOH with scintillation cocktail.

Flux measurements of CO_2 and CH_4 were linearly interpolated and integrated using the trapezoidal rule to calculate the total net fluxes of CO_2 and CH_4 over the 15-day experiment. We assumed that net fluxes of $^{14}CO_2$ and $^{14}CH_4$ were linear at each measurement point if the total net CO_2 and CH_4 fluxes were linear. The method of integrating the total net CO_2 and CH_4 fluxes was also used to estimate the total net fluxes of $^{14}CO_2$ and $^{14}CH_4$ over the 15-day experiment.

Fifteen days after ^{14}C-labeling, the mesocosms were harvested. Total above-ground plant material was clipped at the soil surface and sorted by species. Three soil cores (35 cm depth, 4.2 cm diameter) were collected from each mesocosm and separated for roots and rhizomes. All plant and soil samples were dried, weighed, ground, and then analyzed for total carbon and nitrogen on an elemental analyzer (2400 CHN, Perkin Elmer, Norwalk, Connecticut) and for ^{14}C activity through the use of a biological oxidizer (OX-500, R.J. Harvey Instrument Corp., Patterson, New Jersey).

3. Results

3.1. Mesocosm characteristics

The mesocosms used in this experiment were similar in most respects to the in situ sites monitored simultaneously (Table 1). The two ^{14}C-labeled mesocosms (mesocosms 3 and 4) were similar to each other in biomass (24.1 and 21.7 g biomass per mesocosm, respectively) and in plant density, measured as the number of tillers (plant branches) per mesocosm (33 and 46 tillers per mesocosm, respectively). There was no significant difference among control and ^{14}C-labeled mesocosms and in situ sites in number of

Table 1
Mesocosm characteristics

Measured	^{14}C labeled *mesocosms*[a]	Unlabeled *mesocosms*[a]	In situ sites[a]	Literature value[b]	Reference
Average NEP[c] ($g\ C\ m^{-2}\ d^{-1}$)	4.82	4.60	5.47	2	Shaver et al., 1998[d]
Average RESP[c] ($g\ C\ m^{-2}\ d^{-1}$)	−4.40	−2.82	−3.39	−2; −1	Verville et al., 1998; Shaver et al., 1998[d]
Average GEP[c] ($g\ C\ m^{-2}\ d^{-1}$)	9.22	7.42	8.86	3	Shaver et al., 1998[d]
Above-ground biomass ($g\ m^{-2}$)	399.9	233.0	342.9	438	Shaver & Chapin, 1991; Shaver et al., 1992
Mean leaf number (m^{-2})	2626.5	2277.5	3071.6	3576.8	Shaver & Billings, 1975
Tiller density (m^{-2})	689.4	872.6	1108.2	1145.0	Shaver & Billings, 1975
Average CH_4 flux ($g\ C\text{-}CH_4\ m^{-2}\ d^{-1}$)	0.45	0.26	0.13	0.05–0.06	King et al., 1998; Schimel, 1995; Verville et al., 1998

[a] $n = 2$, see Section 2 for frequency of measurements.
[b] Literature values are provided for comparison of values of individual variables (horizontal) and not intended for comparison of whole system characteristics.
[c] NEP, net ecosystem production; RESP, ecosystem respiration; GEP, gross ecosystem production calculated as the sum of NEP and RESP.
[d] Data collected from different wet sedge community.

leaves or number of tillers ($P = 0.4$ and $P = 0.5$, respectively). The tiller densities were close to previous measurements in a similar wet sedge community (Shaver and Billings, 1975). The average above-ground biomass was $400\ g\ m^{-2}$, and the root: shoot ratio was nearly 2:1. These values are consistent with previous measurements in wet sedge tundra (Shaver and Chapin, 1991; Shaver et al., 1992).

Rates of net ecosystem production (NEP) and respiration (RESP) were measured approximately twice a day in each mesocosm. Gross ecosystem production (GEP) rates were derived from the sum of net ecosystem production and respiration rates. Mean net ecosystem production rates and respiration rates were not significantly different among control and ^{14}C-labeled mesocosms and in situ sites ($P = 0.8$ and $P = 0.2$, respectively). Comparison of the control and labeled mesocosms showed that the ^{14}C-labeling did not affect CO_2 exchange rates. The rates of net ecosystem production and respiration were slightly higher than rates previously reported for wet sedge tundra (Table 1), and the respiration rates were slightly higher than rates reported by Verville et al. (1998) in a field study at the same site. Comparison of the two ^{14}C-labeled mesocosms showed that there was no difference in rates of net ecosystem production ($P = 0.9$) but that mesocosm 4 had higher respiration rates than mesocosm 3 ($P \ll 0.05$). This difference is most likely due to differences in root biomass between mesocosm 3 and mesocosm 4 (37.7 and 49.7 g root biomass per mesocosm, respectively).

The average CH_4 emission rate from the ^{14}C-labeled mesocosms was $0.45\ g\ C\ m^{-2}\ d^{-1}$, which was significantly higher than simultaneous in situ measurements of CH_4 emissions ($P = 0.02$, Table 1). The CH_4 emission rates from the mesocosms were also higher than previous field observations in this wet sedge community (Schimel, 1995; King et al., 1998; Verville et al., 1998). Warmer than average temperatures during the middle of the 1998 growing season may have contributed to the higher CH_4 fluxes we observed. The CH_4 emission rates from mesocosms and in situ sites differed despite the fact that the field experiment was performed under natural climate conditions. Some of the discrepancy between mesocosm and in situ CH_4 emission rates can be attributed to differences in the water table levels between these sites. Additional water (lake water) was added to the mesocosms to keep the water table constant, but the water table at the in situ sites fell during the experiment due to an unusually dry period at the field site during the summer of 1998. Some of the discrepancy may also be attributed to the 8°C difference in soil temperature (Section 2.1). Literature Q_{10} values for CH_4 production range from 1.3 to 28 and depend on the temperature responses of various processes which lead to methane production (van Hulzen et al., 1999). However, most of the difference between mesocosm and in situ CH_4 emission rates was probably caused by the unnaturally stable water column in the mesocosms. Limited lateral and vertical movement of soil porewater caused changes in the soil environment which led to higher emission rates through increased CH_4 production and decreased CH_4 oxidation.

Porewater concentrations of DOC, dissolved CO_2, and dissolved CH_4 were similar in all mesocosms (averages were 1.8 mM, 7.3 mM, and 300 μM, respectively). The concentrations of these dissolved porewater components remained relatively constant throughout the experiment. Measurements of porewater concentrations of dissolved CO_2 and dissolved CH_4 in the ^{14}C-labeled mesocosms tended to be higher than those measured in situ (5.5 mM, $P = 0.15$; 100 μM, $P = 0.13$, respectively). Porewater concentrations of DOC were higher in ^{14}C-labeled mesocosms than in in situ sites (0.8 mM, $P = 0.05$). These concentration differences in porewater components between mesocosm and in situ measurements can also be attributed to the limited lateral and vertical exchange in the mesocosms.

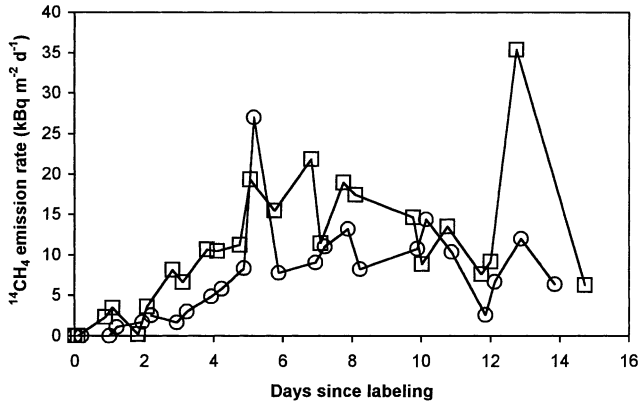

Fig. 1. Rates of emission of $^{14}CH_4$ from mesocosm 3 (squares) and mesocosm 4 (circles). The rates reached a maximum in both mesocosms approximately 5–7 days after labeling. A second maximum at 13 days after labeling is probably due to slight increases in air temperature which limited plant photosynthesis.

3.2. Contribution of recent photosynthates to methane emission

The conversion of photosynthate carbon to emitted CH_4 occurred quickly. Recently-assimilated carbon was translocated to the roots and available to the soil microbial community as root exudates or $^{14}CO_2$ from root respiration and emitted as $^{14}CH_4$ within 24 h (Fig. 1). Emission rates of $^{14}CH_4$ reached a maximum approximately 5–7 days after labeling. Both mesocosms exhibited similar patterns of $^{14}CH_4$ emission. The ^{14}C pulse-label mainly appeared as $^{14}CH_4$ over the course of 7–8 days beginning 2 days after labeling. After this time, the emission rate of $^{14}CH_4$ declined. The second maximum in $^{14}CH_4$ emission at 13 days after labeling followed a slight increase in air temperature. It also corresponded with an observed increase in the specific activity of DOC in the porewater that may have been caused by increased translocation of carbon between shoots to roots on warmer days that cause the plants to limit their photosynthesis.

The maximum emission rate of $^{14}CO_2$ occurred within the first day after labeling (Fig. 2). The emission of $^{14}CO_2$ immediately following labeling could be the result of leaf respiration of the ^{14}C-labeled CO_2 that had just been assimilated. Emission rates of $^{14}CO_2$ decreased very quickly in the first 2 days after labeling and remained relatively constant during the rest of the experiment.

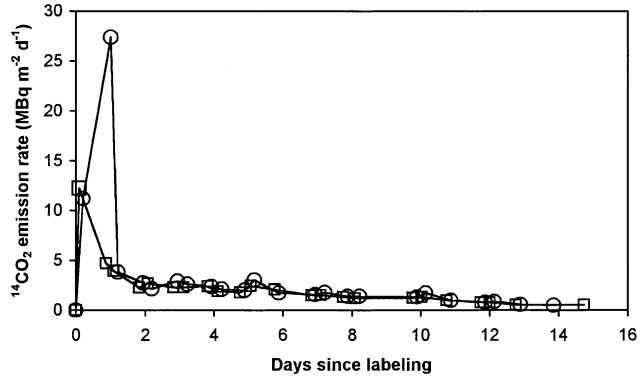

Fig. 2. Rates of emission of $^{14}CO_2$ from mesocosm 3 (squares) and mesocosm 4 (circles).

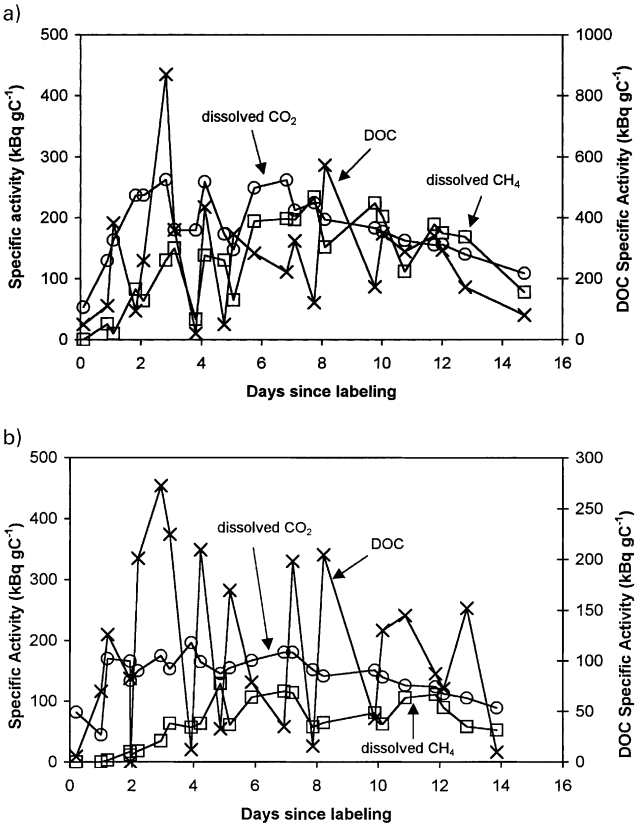

Fig. 3. Specific activities of porewater DOC (crosses), dissolved CO_2 (circles), and dissolved CH_4 (squares) over the course of the experiment in (a) mesocosm 3 and (b) mesocosm 4. Note the separate axis for the specific activity of DOC.

3.3. Pathway of recent photosynthates to methane emission

Time series measurements of ^{14}C in dissolved components in the soil porewater integrated over the entire soil profile showed transfer of recently assimilated carbon to soil porewater within one day of labeling (Fig. 3) and support our measurements of emitted $^{14}CH_4$. The highest values of specific activity were measured in the dissolved CO_2 pool of soil porewater initially. The specific activities of all porewater carbon pools increased over the first three days after labeling. Overall, the DOC pool exhibited higher values of specific activity than any other porewater carbon pools. The specific activity of DOC also showed the greatest variability (especially Fig. 3b). Patterns of changes in specific activity were similar in all porewater pools, indicating that transfers between pools in the porewater occurred rapidly (Fig. 3a). Emissions of $^{14}CH_4$ (Fig. 1) were coincident with changes in the specific activity of dissolved CH_4 in

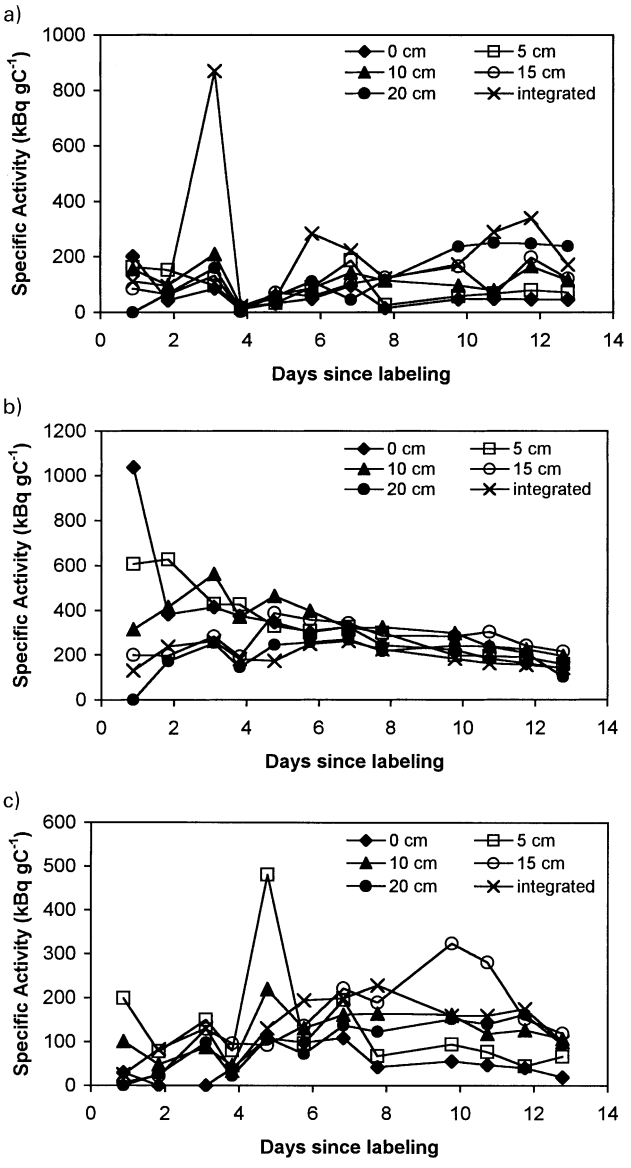

Fig. 4. Specific activities of (a) DOC, (b) dissolved CO_2, and (c) dissolved CH_4 for individual depths and the integrated samples in the soil profile of mesocosm 3 over the course of the experiment.

the porewater (Fig. 3). Because the most direct pathway for CH_4 to reach the atmosphere is through the plants, this suggests that transport of CH_4 from the soil to the atmosphere was plant-mediated.

Measurements of dissolved components in the soil porewater profile showed that transport of recent photosynthates to the soil most likely occurred via the roots (Fig. 4). Immediately after labeling, the highest specific activity of porewater components usually appeared in the top 10 cm of soil, and the fastest, most direct pathway for carbon to reach up to 10 cm depth in the soil is through transport by roots. Previous measurements have shown that the highest density of roots occurs between 3 and 9 cm depth in the soil (King et al., 1998). Similarly, measurements in this experiment of root biomass separated

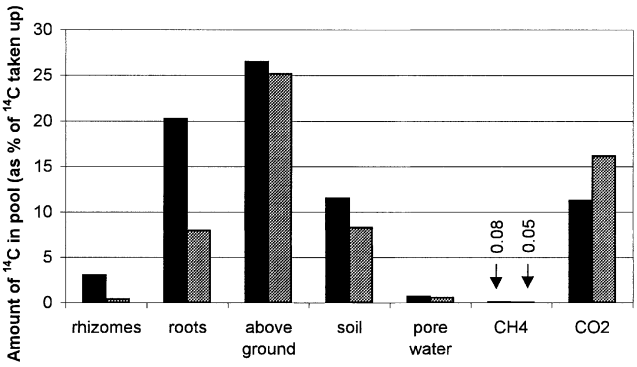

Fig. 5. Mass balance distribution of ^{14}C in mesocosm 3 (black bars) and mesocosm 4 (gray bars) at the time of harvest 15 days after labeling.

by 5-cm depth intervals at the time of harvest showed that most of the roots were located in the 5–10 cm depth interval of the soil profile. Therefore, the root–soil interface is an important site not only for transport of CH_4 from the soil to the atmosphere (King et al., 1998), but also probably for conversion of recently-fixed carbon to methane. Soil porewater samples that integrated over the whole soil profile (samples that include equal volumes from several depths) were not necessarily representative of the average of the soil column (Fig. 4).

3.4. Distribution of ^{14}C label at the time of harvest

Using measurements of plant material collected at harvest, measurements of porewater pools collected immediately prior to harvest, and integrations of the measured gas fluxes for the length of the experiment, we constructed a mass balance of the ^{14}C label. The distribution of the ^{14}C label at the time of harvest, 15 days after labeling, was similar in both mesocosms (Fig. 5). The values are presented relative to the original amount of ^{14}C taken up by each mesocosm. While a large proportion (25%) of the ^{14}C label assimilated at mid-season remained in the aboveground biomass, approximately 10–20% of the ^{14}C label had been allocated to roots, and approximately 10% of the ^{14}C label was found in the soil. Less than 5% of the ^{14}C label was in the rhizome and porewater pools at 15 days after labeling. Less than 1% (~0.1%) of the ^{14}C label was emitted as $^{14}CH_4$, and approximately 15% of the ^{14}C label was emitted as $^{14}CO_2$.

The overall differences in ^{14}C label distribution at the time of harvest between mesocosm 3 and mesocosm 4 are consistent with relative differences in the measured CO_2 exchange rates and total biomass of these mesocosms (Fig. 5, Table 1). Mesocosm 4 had higher root biomass and higher respiration rates. Therefore the proportion of loss of ^{14}C as CO_2 through respiration was higher from mesocosm 4 than mesocosm 3. Mesocosm 3 had slightly higher net ecosystem production rates and higher aboveground biomass. These plants sequestered more ^{14}C label in biomass and lost less ^{14}C as $^{14}CO_2$. Recovery of the total

amount of ^{14}C taken up in the mesocosms was greater in mesocosm 3 than in mesocosm 4 (73 vs 59%, respectively). This difference could be due to differences in the distribution of fine roots and incomplete separation of roots from soil. Recovery of ^{14}C could also be affected by variability in the measurement of porewater carbon pools which can cause large errors when the measurements are scaled to the mesocosm level. Continuous measurements of gas fluxes and ^{14}C activity of emitted CO_2 and CH_4 would improve those estimates and increase recovery of the ^{14}C label.

4. Discussion

This experiment allowed us to investigate the question of plant contribution to CH_4 emissions under near-in situ conditions. The mesocosms were exposed to natural climate conditions. Stainless steel containers prevented the diffusion of oxygen and other gases through the sides of the mesocosms into the soil, and the water jacket kept soil temperatures cooler than air temperatures and closer to natural conditions. We avoided severe disturbance of the mesocosms by performing the experiment in the field rather than transporting the mesocosms long distances to laboratory growth chambers.

The detection of $^{14}CH_4$ within 24 h after labeling shows that transfer of carbon fixed by photosynthesis to CH_4 emission occurred rapidly. These are the first measurements of this kind made under field conditions in any ecosystem. Although the amount of ^{14}C converted to CH_4 was small, the overall patterns of carbon allocation in the field experiment were similar to patterns observed in growth chamber experiments. Over the 15-day experiment, approximately 0.1% of the ^{14}C-label initially taken up was emitted as CH_4. Transfer of ^{14}C to the soil porewater appeared to correspond with daily variations in CO_2 exchange, and ^{14}C did not accumulate in the porewater carbon pools. Our results are similar to the results from laboratory studies of other ecosystems (Minoda and Kimura, 1994; Minoda et al., 1996; Dannenberg and Conrad, 1999; Megonigal et al., 1999). Similar rates of carbon cycling through plants have been measured in agroecosystems such as rye, maize, ryegrass, and wheat (Cheng and Coleman, 1990; Martens, 1990; Meharg and Killham, 1990; Swinnen et al., 1994).

It is well-recognized that plants play an important role in transporting CH_4 to the atmosphere in these wet sedge tundra communities as well as in other ecosystems (King et al., 1998). This carbon tracer experiment has shown that plants also influence CH_4 emissions by providing carbon substrates that are quickly consumed by microbes in the soil and emitted in part as CH_4. Previous work in this field suggests a relationship between plant productivity and CH_4 emission (Sebacher et al., 1986; Moore and Knowles, 1990; Whiting and Chanton, 1993; Waddington et al., 1996). The results of this ^{14}C pulse-labeling experiment describe a possible mechanism underlying those observed correlations and may help to explain the high variability in CH_4 emissions from tundra ecosystems (Reeburgh et al., 1998). Other recent measurements suggest that a relationship between CH_4 flux and respiratory CO_2 flux could be more applicable across different wetlands (Kim et al., 2000); however, no experimental work has been done to investigate this proposed relationship.

The results of this ^{14}C pulse-labeling experiment conducted under near-in situ conditions show that the conversion of photosynthate carbon to emitted CH_4 occurred quickly and suggest that recent photosynthates can be an important source of carbon for methanogenesis. While this study focused on one portion of the arctic growing season, we recognize that the contribution of plant photosynthates to CH_4 emissions may be dependent on plant phenology. Seasonal variation in total CH_4 emission related to growth stage has been observed in a prairie marsh (Kim et al., 1998), and variation in the contribution of photosynthates to CH_4 emission with respect to time of growing season was observed by Minoda and Kimura (1994) in rice paddy fields. These results underscore the need to understand both the short-term and long-term effects of plant productivity on CH_4 emission in order to make accurate predictions of the response of CH_4 emission to changes in plant productivity because of changes in climate or atmospheric CO_2 concentration. Future studies should also be aimed at understanding the contribution of CH_4 oxidation to overall CH_4 fluxes as well as the processes that control CH_4 oxidation.

Acknowledgements

Kate Su and An Tran were a great help in the lab and in the field. We thank G.W. Kling and K.J. Riseng for analysis of porewater samples for DOC concentration and K.J. Nadelhoffer and K.T. Thieler for CHN analysis of plant and soil samples and for use of the biological oxidizer. We also acknowledge the support of the Toolik Field Station Staff. This work was supported by NSF grant OPP 96-15942 through a subcontract from the Marine Biological Laboratory. We thank Steve Whalen, Arvin Mosier, and two anonymous reviewers for comments on earlier drafts of this manuscript.

References

Aravena, R., Warner, B.G., Charman, D.J., Belyea, L.R., Mathur, S.P., Dinel, H., 1993. Carbon isotopic composition of deep carbon gases in an ombrogenous peatland, Northwestern Ontario, Canada. Radiocarbon 35, 271–276.

Aselmann, I., Crutzen, P.J., 1989. Global distribution of natural freshwater wetlands and rice paddies, their net primary productivity, seasonality and possible methane emissions. Journal of Atmospheric Chemistry 8, 307–358.

Cao, M., Marshall, S., Gregson, K., 1996. Global carbon exchange and methane emissions from natural wetlands: Application of a process-based model. Journal of Geophysical Research 101, 14399–14414.

Chanton, J.P., Bauer, J.E., Glaser, P.A., Siegel, D.I., Kelley, C.A., Tyler, S.C., Romanowicz, E.H., Lazrus, A., 1995. Radiocarbon evidence for the substrates supporting methane formation within northern Minnesota peatlands. Geochimica et Cosmochimica Acta 59, 3663–3668.

Cheng, W., Coleman, D.C., 1990. Effect of living roots on soil organic matter decomposition. Soil Biology & Biochemistry 22, 781–787.

Chidthaisong, A., Watanabe, I., 1997. Methane formation and emission from flooded rice soil incorporated with ^{13}C-labeled rice straw. Soil Biology & Biochemistry 29, 1173–1181.

Clymo, R.S., Reddaway, E.J.F., 1971. Productivity of *Sphagnum* (bog–moss) and peat accumulation. Hydrobiology 12, 181–192.

Dannenberg, S., Conrad, R., 1999. Effect of rice plants on methane production and rhizospheric metabolism in paddy soil. Biogeochemistry 45, 53–71.

Hultén, E., 1968. Flora of Alaska and Neighboring Territories. Stanford University Press, Stanford.

Kim, J., Verma, S., Billesbach, D., 1998. Seasonal variation in methane emission from a temperate *Phragmites*-dominated marsh: Effect of growth stage and plant-mediated transport. Global Change Biology 5, 433–440.

Kim, J., Verma, S.B., Shurpali, N.J., Harazono, Y., Miyata, A., Yun, J.-I., Tanner, B., Kim, J.W., 2000. Diurnal and seasonal variations in CH_4 emission from various freshwater wetlands. In: van Ham, J. (Ed.). Non-CO_2 Greenhouse Gases: Scientific Understanding, Control and Implementation. Kluwer Academic Publishers, Netherlands, pp. 131–136.

Kimura, M., 1997. Sources of methane emitted from paddy fields. Nutrient Cycling in Agroecosystems 49, 153–161.

King, J.Y., 1999. Effects of Vegetation on Methane Emissions from Arctic Tundra Ecosystems. Doctoral Dissertation. University of California, Irvine, CA.

King, J.Y., Reeburgh, W.S., Regli, S.K., 1998. Methane emission and transport by arctic sedges in Alaska: Results of a vegetation removal experiment. Journal of Geophysical Research 103, 29083–29092.

Kling, G.W., Kipphut, G.W., Miller, M.M., O'Brien, W.J., 2000. Integration of lakes and streams in a landscape perspective: the importance of material processing on spatial patterns and temporal coherence. Freshwater Biology 43, 477–497.

Klinger, L.F., Zimmerman, P.R., Greenberg, J.P., Heidt, L.E., Guenther, A.B., 1994. Carbon trace gas fluxes along a successional gradient in the Hudson Bay lowland. Journal of Geophysical Research 99, 1469–1494.

Martens, R., 1990. Contribution of rhizodeposits to the maintenance and growth of soil microbial biomass. Soil Biology & Biochemistry 22, 141–147.

Martens, C.S., Kelley, C.A., Chanton, J.P., Showers, W.J., 1992. Carbon and hydrogen isotopic characterization of methane from wetlands and lakes of the Yukon-Kuskokwim Delta. Western Alaska. Journal of Geophysical Research 97, 16689–16701.

Megonigal, J.P., Whalen, S.C., Tissue, D.T., Bovard, B.D., Albert, D.B., Allen, A.S., 1999. A plant–soil–atmosphere microcosm for tracing radiocarbon from photosynthesis through methanogenesis. Soil Science Society of America Journal 63, 665–671.

Meharg, A.A., Killham, K., 1990. Carbon distribution with the plant and rhizosphere for *Lolium perenne* subjected to anaerobic conditions. Soil Biology & Biochemistry 22, 643–647.

Minoda, T., Kimura, M., 1994. Contribution of photosynthesized carbon to the methane emitted from paddy fields. Geophysical Research Letters 21, 2007–2010.

Minoda, T., Kimura, M., Wada, E., 1996. Photosynthates as dominant sources of CH_4 and CO_2 in soil water and CH_4 emitted to the atmosphere from paddy fields. Journal of Geophysical Research 101, 21091–21097.

Moore, T.R., Knowles, R., 1990. Methane emissions from fen, bog, and swamp peatlands in Quebec. Biogeochemistry 11, 45–61.

Reeburgh, W.S., 1996. 'Soft spots' in the global methane budget. In: Lidstrom, M.E., Tabita, F.R. (Eds.). Microbial Growth on C1 Compounds. Kluwer Academic Publishers, Boston, pp. 334–342.

Reeburgh, W.S., King, J.Y., Regli, S.K., Kling, G.W., Auerbach, N.A., Walker, D.A., 1998. A CH_4 emission estimate for the Kuparuk River basin. Journal of Geophysical Research, Alaska 103, 29005–29013.

Schimel, J.P., 1995. Plant transport and methane production as controls on methane flux from arctic wet meadow tundra. Biogeochemistry 28, 183–200.

Sebacher, D.I., Harriss, R.C., Bartlett, K.B., Sebacher, S.M., Grice, S.S., 1986. Atmospheric methane sources: Alaskan tundra bogs, an alpine fen, and a subarctic boreal marsh. Tellus 38B, 1–10.

Shaver, G., Billings, W., 1975. Root production and root turnover in a wet tundra ecosystem, Barrow, Alaska. Ecology 56, 401–409.

Shaver, G.R., Billings, W.D., Chapin III, F.S., Giblin, A.E., Nadelhoffer, K.J., Oechel, W.C., 1992. Global change and the carbon balance of arctic ecosystems. Bioscience 42, 433–441.

Shaver, G.R., Chapin III, F.S., 1991. Production: biomass relationships and element cycling in contrasting Arctic vegetation types. Ecological Monographs 61, 1–31.

Shaver, G.R., Johnson, L.C., Cades, D.H., Murray, G., Laundre, J.A., Rastetter, E.B., Nadelhoffer, K.J., Giblin, A.E., 1998. Biomass and CO_2 flux in wet sedge tundras: Responses to nutrients, temperature, and light. Ecological Monographs 68, 75–97.

Svensson. B.H. Carbon fluxes from acid peat of a subarctic mire with emphasis on methane No. 20. Swedish University of Agricultural Sciences, Department of Microbiology, 301 (1983).

Swinnen, J., Van Veen, J.A., Merckx, R., 1994. ^{14}C pulse-labelling of field-grown spring wheat: An evaluation of its use in rhizosphere carbon budget estimations. Soil Biology & Biochemistry 26, 161–170.

van Hulzen, J.B., Segers, R., van Bodegom, P.M., Leffelaar, P.A., 1999. Temperature effects on soil methane production: An explanation for observed variability. Soil Biology & Biochemistry 31, 1919–1929.

Verville, J.H., Hobbie, S.E., Chapin III, F.S., Hooper, D.U., 1998. Response of tundra CH_4 and CO_2 flux to manipulation of temperature and vegetation. Biogeochemistry 41, 215–235.

Waddington, J.M., Roulet, N.T., Swanson, R.V., 1996. Water table control of CH_4 emission enhancement by vascular plants in boreal peatlands. Journal of Geophysical Research 101, 22775–22785.

Wahlen, M., Tanaka, N., Henry, R., Deck, B., Zeglen, J., Vogel, J.S., Southon, J., Shemesh, A., Fairbanks, R., Broecker, W., 1989. Carbon-14 in methane sources and in atmospheric methane: The contribution from fossil carbon. Science 245, 286–290.

Walker, D.A., Binnian, E., Evans, B.M., Lederer, N.D., Nordstrand, E., Webber, P.J., 1989. Terrain, vegetation and landscape evolution of the R4D research site, Brooks Range Foothills, Alaska. Holarctic Ecology 12, 238–261.

Walter, B., Heimann, M., 2000. A process-based, climate-sensitive model to derive methane emissions from natural wetlands: Application to five wetland sites, sensitivity to model parameters, and climate. Global Biogeochemical Cycles 14, 745–765.

Watanabe, A., Yoshida, M., Kimura, M., 1998. Contribution of rice straw carbon to CH_4 emission from rice paddies using C-13-enriched rice straw. Journal of Geophysical Research 103, 8237–8242.

Whalen, S.C., Reeburgh, W.S., 1988. A methane flux time series for tundra environments. Global Biogeochemical Cycles 2, 399–409.

Whalen, S.C., Reeburgh, W.S., 1990. Consumption of atmospheric methane by tundra soils. Nature 346, 160–162.

Whiting, G.J., Chanton, J.P., 1993. Primary production control of methane emission from wetlands. Nature 364, 779–794.