Pulse-labeling studies of carbon cycling in Arctic tundra ecosystems: The contribution of photosynthates to methane emission

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[1] We investigated a possible mechanism underlying observed correlations between net ecosystem carbon dioxide exchange and methane emission. Using the technique of ^{14}C pulse-labeling, we traced the movement of carbon fixed by photosynthesis as it moved through wet sedge and moist tussock tundra plant-soil mesocosms and was emitted as methane to the atmosphere. The ^{14}C tracer provided a definitive way of quantifying the fate of recently fixed carbon. Carbon fixed by photosynthesis was measured as emitted methane from both moist tussock and wet sedge tundra mesocosms within 2 hours after labeling. Integration of time series measurements of methane emission showed that recent photosynthates are an important source of carbon for methane production. Approximately 2% of carbon fixed by photosynthesis at peak growing season was subsequently emitted as methane from moist tussock tundra, and approximately 3% was emitted as methane from wet sedge tundra. Measurements of soil pore water carbon pools demonstrate rapid transfer of ^{14}C from plant carbon to dissolved forms and subsequently to the atmosphere as carbon dioxide or methane. *INDEX TERMS:* 1615 Global Change: Biogeochemical processes (4805); 1030 Geochemistry: Geochemical cycles (0330); 1040 Geochemistry: Isotopic composition/chemistry; 9315 Information Related to Geographic Region: Arctic region; *KEYWORDS:* ^{14}C pulse-labeling, arctic tundra, methane biogeochemistry

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1. Introduction

[2] Attempts to estimate current methane (CH_4) emissions to the atmosphere and to predict future CH_4 emissions from natural and managed ecosystems have been hampered by a lack of information about the key controls on CH_4 production and oxidation which interact to control net emission. Many studies have examined factors that are correlated with CH_4 emission, such as water table depth, soil temperature, water chemistry, and vegetation cover [*Frolking and Crill*, 1994; *Moore et al.*, 1994; *Shannon and White*, 1994; *Bubier et al.*, 1995; *Mikkelä et al.*, 1995; *Johnson et al.*, 1996; *Moosavi et al.*, 1996]. However, modeling efforts to predict net CH_4 emissions are complicated by the fact that numerous factors influence net CH_4 emission, and no single-factor approach can fully explain the high spatial and temporal variability in observed CH_4 emission [*Whalen and Reeburgh*, 1992]. Models based only on environmental factors such as water table depth and soil temperature cannot capture observed CH_4 emission dynamics, and more recent process-based models [*Cao et al.*, 1995, 1996; *Walter et al.*, 1996; *Walter and Heimann*, 2000] appear to match observations only under a limited set of environmental conditions.

[3] Identifying the carbon sources of CH_4 emissions is key to understanding spatial and temporal variations and to predicting future changes in the global CH_4 budget. The primary substrates for microbial production of CH_4 are acetate (CH_3COOH) and carbon dioxide (CO_2). These substrates become available to methanogens through a variety of pathways, including decomposition of soil organic matter ("old carbon") and root exudation and root respiration ("new carbon," also referred to as recently fixed carbon or recent photosynthates). Quantifying the contributions of old versus new sources of carbon is important in modeling the current and future global carbon budgets [*Trumbore et al.*, 1995] and global CH_4 budgets. Radiocarbon measurements of CH_4 in soil pore water and emitted CH_4 indicate that CH_4 is produced mainly from new carbon

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[*Chanton et al.*, 1988; *Wahlen et al.*, 1989; *Aravena et al.*, 1993; *Chanton et al.*, 1995]. The purpose of this study was to directly measure the contribution of recently fixed carbon to CH_4 emission.

[4] Several researchers have also observed correlations between net ecosystem CO_2 exchange and CH_4 emission in a variety of ecosystems [*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 and Roulet*, 1996; *Christensen et al.*, 2000]. Plants influence CH_4 emission, and the importance of plant transport of CH_4 is well-established [e.g., *Dacey and Klug*, 1979; *Chanton et al.*, 1992; *Torn and Chapin*, 1993]. Because plant transport alone does not account for the differences in CH_4 emission between vegetated and nonvegetated sites [*Schimel*, 1995; *King et al.*, 1998], observed correlations between CH_4 emission and plant biomass or CO_2 exchange rates are likely best explained by a combination of plant influences on CH_4 transport, CH_4 oxidation in the rhizosphere, and substrate availability for methanogenesis. However, the mechanism underlying a relationship between carbon assimilated by plants through photosynthesis and carbon emitted as CH_4 has not been identified in natural ecosystems. In order to look for a mechanistic explanation for the observed correlations between net ecosystem CO_2 exchange and CH_4 emission, we conducted a pulse-labeling tracer study to observe the conversion of plant carbon to emitted CH_4.

[5] Past pulse-labeling studies have focused on measuring the carbon isotopic tracer in organic carbon compounds in the soil and plants and also in CO_2 in the soil [*Martin and Kemp*, 1986; *Milchunas and Lauenroth*, 1992; *Swinnen et al.*, 1994]. The flux of carbon in the form of CH_4 remains relatively poorly characterized. Two research groups have investigated the contribution of photosynthates to CH_4 emission from rice agroecosystems using pulse-labeling techniques. Using ^{13}C as a tracer, *Minoda and Kimura* [1994] and *Minoda et al.* [1996] estimated that photosynthesized CO_2 contributed 29% of the total CH_4 emissions during a growing season and 22% of the total CH_4 emissions if rice straw was applied to the soil. *Dannenberg and Conrad* [1999] performed a ^{14}C pulse-labeling study on rice plants and found that 3–6% of the assimilated $^{14}CO_2$ was emitted as $^{14}CH_4$ within 16 days after labeling. *Watanabe et al.* [1999] measured contributions of carbon to CH_4 emission from sources other than plants and estimated by difference that carbon supplied by rice plants made up approximately 80–85% of the total CH_4 emission when rice straw was not applied to the soils. In the only published study of a natural ecosystem, *Megonigal et al.* [1999] conducted a ^{14}C pulse-labeling study on a single wetland plant and found that $^{14}CH_4$ represented less than 1% of the ^{14}C recovered 17 days after labeling. This paper reports the results of the first replicated mesocosm study of a natural wetland system to examine carbon fluxes in the form of CH_4.

[6] High-latitude wetland ecosystems currently represent approximately 30% of the natural wetland source of CH_4 to the atmosphere [*Reeburgh et al.*, 1998]. Because of their high soil moisture and carbon contents, arctic ecosystems are potentially important as future sources of atmospheric CH_4. Developing an understanding of the controls on carbon cycling in arctic tundra is critical to estimating current and future global CH_4 budgets, as well as current and future arctic carbon budgets. This work was part of an integrated study using ^{14}C pulse-labeling of tundra mesocosms to investigate C cycling in moist tussock and wet sedge tundra ecosystems [*Loya et al.*, 2002; G. W. Kling et al., Gaseous and dissolved carbon production in Arctic ecosystems: The role of belowground processes, manuscript in preparation, 2002; K. J. Nadelhoffer et al., Pulse-labeling studies of carbon cycling in Arctic tundra ecosystems: Photosynthate allocation in moist tussock and wet sedge tundra vegetation, manuscript in preparation, 2002]. In this paper we present the results of measurements aimed at tracing the contribution of recent plant photosynthates to CH_4 emission. We address the following questions: How quickly is the carbon fixed by photosynthesis emitted as CH_4? How much carbon fixed by photosynthesis is emitted as CH_4 in the same growing season?

2. Methods

2.1. Experimental Setup and Design

[7] Intact cores of soil plus vegetation were taken from moist tussock and wet sedge tundra sites at the Toolik Lake Long Term Ecological Research site on the North Slope of Alaska (68°38′N, 149°36′W) during August 1997. Each core was 27 cm in diameter and approximately 31 cm deep. The wet sedge tundra mesocosms were dominated by *Carex chordorrhiza*, and the moist tussock tundra mesocosms were dominated by *Eriophorum vaginatum*. Further description of these tundra types is provided by *Shaver and Chapin* [1991]. The cores (plant-soil mesocosms) were placed in 20 L polyethylene containers and transported to Woods Hole, Massachusetts.

[8] We conducted the experiment in growth chambers (model PVG36, Controlled Environments, Inc.) at the Ecosystems Center of the Marine Biological Laboratory over the period of a simulated arctic growing season. To induce plant senescence we gradually reduced the air temperature to −4°C and decreased the photoperiod from 12 hours of light to complete darkness. After one week of continuous darkness at −4°C, we simulated the start of the growing season. Over the period of one week, we gradually increased the air temperature to 10°C and increased the photoperiod to 24 hours of full light. The average flux of photosynthetically active radiation (PAR) at the leaf level under full light conditions was 800–1000 μmol photons m^{-2} s^{-1}. The average soil temperature under 10°C and full light conditions was 15.2°C. The water table was kept constant at approximately 5 cm below the soil surface in moist tussock tundra mesocosms and at 2 cm above the soil surface in wet sedge tundra mesocosms through addition of distilled water. After 9.5 weeks under these conditions we began to simulate the end of the growing season by lowering the temperature and light levels. Over a period of three weeks we gradually decreased the photoperiod to 10 hours of light and reduced the air temperature to 6°C, with overnight freezes.

[9] Twelve each of moist tussock and wet sedge tundra mesocosms were pulse-labeled with 8 MBq (216 μCi) of ^{14}C. We began to pulse-label the mesocosms on the 52nd day of our simulated growing season when the plants were close to maximum biomass. Three mesocosms (referred to as long-term mesocosms) each of moist tussock and wet sedge tundra were sampled intensively for gas fluxes for approximately 10 weeks after they were labeled. By the end of the experiment, the plants had senesced, and the mesocosms were harvested for analysis of ^{14}C remaining in each mesocosm. The nine other mesocosms (short-term mesocosms) of each vegetation type were analyzed to examine the allocation of ^{14}C carbon in all carbon pools, including plant and soil pools, at several intermediate times during the experiment. Three mesocosms of each vegetation type were destructively harvested at 1 day, 1 week, and 3 weeks after labeling. The ^{14}C label distributions in the short-term mesocosms were used to examine changes in label distribution in plant and soil pools through time. The time series gas flux measurements from the original three long-term mesocosms of each vegetation type were used to estimate the gas fluxes from the short-term mesocosms harvested at intermediate time points in the experiment.

2.2. ^{14}C Pulse Labeling

[10] Each plant-soil mesocosm was sealed with a clear acrylic headspace chamber during labeling. A ^{14}C-labeled sodium bicarbonate solution (55 MBq/g C) was acidified with hydrochloric acid to produce $^{14}CO_2$, which was taken up by plants through photosynthesis. Additional unlabeled CO_2 was added as necessary to maintain the CO_2 concentration near 400 ppmv inside the chamber. Each mesocosm was pulse-labeled for a period of 1.5 hours during which samples of headspace air were collected to determine uptake efficiency. The headspace chamber was removed after labeling. On average, 93% and 80% of the ^{14}C label added (8 MBq) was taken up in moist tussock and wet sedge tundra mesocosms, respectively. Measurements in subsequent experiments showed that direct diffusion of label into the mesocosm soils during labeling was negligible (<4% of label added). Thus we assume that ^{14}C measured in carbon pools such as emitted methane, soil pore water, and plant biomass was originally taken up through photosynthesis. The mesocosms and labeling methodology are described in greater detail by *King* [1999].

2.3. Sampling and Analysis

[11] Gas fluxes of CO_2, $^{14}CO_2$, CH_4, and $^{14}CH_4$, and pore water samples were measured from each of the long-term mesocosms 1–2 times per day during the first week after labeling and 1–2 times per week thereafter. Gas fluxes were measured from the short-term mesocosms approximately every other week. Gas fluxes and pore water samples were also measured on each mesocosm immediately before it was destructively harvested.

[12] CH_4 fluxes were measured using the static chamber method and by calculating the increase in chamber headspace CH_4 concentrations over time [*Whalen and Reeburgh*, 1988]. Specifically, we sampled 10 mL of headspace air at 10-min intervals over 30 min (average chamber headspace volume = 35 L). A reflective covering was placed over the chamber to prevent excessive temperature changes during the flux measurement period. The chamber was placed over each mesocosm only during gas flux measurements. The air samples were analyzed for CH_4 concentration on a gas chromatograph equipped with a flame ionization detector (described below). CH_4 fluxes to the atmosphere, representing loss from the ecosystem, are considered positive.

[13] CO_2 flux measurements were made using a portable infrared gas analyzer (LI-6200, Licor, Inc.). The photosynthesis system was calibrated using National Institute of Standards and Technology (NIST)-analyzed CO_2 standards. The net ecosystem CO_2 exchange rates were determined using a clear acrylic headspace chamber over a 3-min period. The chamber was darkened using a reflective covering for ecosystem respiration measurements, also made over a 3-min period. A gas chromatograph (GC-14A, Shimadzu Corp.) equipped with two 1-m Porapak Q columns, a flame ionization detector (FID) for CH_4, and a thermal conductivity detector (TCD) for CO_2 was used to analyze gas samples collected during CH_4 flux measurements and gas samples extracted from soil pore water. Both gas chromatograph detectors were calibrated using standards that were referenced to NIST standards. Methane fluxes were calculated based on the linear regression of syringe samples that described the change in CH_4 concentration over time. A linear regression of $r^2 < 0.90$ was rejected due to sampling error or bias of flux measurements due to other processes, such as experimenter-caused ebullition. Flux measurements of CO_2 and CH_4 were linearly interpolated and integrated over time using the trapezoidal rule to calculate the overall fluxes of CO_2 and CH_4 in each experiment. The fluxes of $^{14}CO_2$ and $^{14}CH_4$ (analyses described below) were integrated in the same way. These integrated fluxes of $^{14}CH_4$ and the total amount of ^{14}C taken up as $^{14}CO_2$ (Section 2.2.) were used to calculate the fraction of assimilated $^{14}CO_2$ emitted as $^{14}CH_4$.

[14] A sample of headspace air was collected in a glass bulb (175 cm^3) for ^{14}C analysis of the CO_2 and CH_4 at the end of each CH_4 flux measurement. Bulbs were flushed and filled using a small diaphragm pump (model 801501, Sensidyne Inc.) at a flow rate of 600 mL min^{-1}. The sample bulbs were sealed by two valves and were processed within 48 hours for analysis of $^{14}CO_2$ and $^{14}CH_4$.

[15] Soil pore water samples were collected using stainless steel needles inserted in the soil. Gas extraction from soil pore water was performed by a headspace equilibration of bubble-free pore water with nitrogen ($N_{2,gas}$), as described by *Kling et al.* [2000]. Dissolved CH_4 concentrations were calculated from measured headspace concentrations using solubility coefficients from *Yamamoto et al.* [1976]. Dissolved organic carbon content was determined using a TOC analyzer (TOC-5000, Shimadzu Corp.).

[16] Gas samples were separated and prepared for ^{14}C analysis on an oxidation line [*Whalen and Reeburgh*, 1990; *King*, 1999]. The gas samples were introduced to the oxidation line, and CO_2, including $^{14}CO_2$, was trapped in a 1 M sodium hydroxide (NaOH) trap. The rest of the gas sample passed through a combustion tube (800°C) containing CuO catalyst. Methane and other hydrocarbons, present

only in trace amounts, were converted to CO_2 and trapped in a second NaOH trap. Calibration tests of the oxidation line demonstrated that 99% of CH_4 passed through the combustion tube was oxidized to CO_2. Subsamples of the NaOH solutions were added to scintillation cocktail (Scintiverse II, Fisher Scientific) and analyzed in a liquid scintillation counter (LS 3801, Beckman Instruments, Inc.) for ^{14}C activity. Corrections were made for quench caused by NaOH.

3. Results and Discussion

3.1. Biomass, CO_2 Exchange, and Methane Emission Measurements

[17] The mean (± standard error, n = 3) aboveground biomass of the moist tussock tundra mesocosms harvested at the end of the experiment was 1047 ± 108 g m^{-2}; average aboveground biomass of the wet sedge tundra mesocosms harvested at the end of the experiment was 257 ± 20 g m^{-2}. The aboveground biomass measurements for moist tussock tundra are consistent with field observations of biomass [*Shaver and Chapin*, 1991; *Shaver et al.*, 1992; *Chapin et al.*, 1996]. The aboveground biomass values for wet sedge tundra were slightly lower than those reported from field observations [*Shaver et al.*, 1998]. The ratio of aboveground to belowground biomass was approximately 2.7:1 in the moist tussock tundra and 1:2.4 in the wet sedge tundra mesocosms. The ratio observed in moist tussock tundra mesocosms is high relative to field observations, perhaps due to incomplete separation of fine roots from the soil or to decreased root growth or increased root mortality (turnover) in the mesocosms. The ratio observed in wet sedge tundra mesocosms is similar to field observations [*Shaver and Chapin*, 1991].

[18] The mean (± standard error, n = 3) net ecosystem CO_2 exchange in moist tussock tundra was greater than in wet sedge tundra over the length of the experiment, (1.3 ± 0.2 versus 0.1 ± 0.2 g C m^{-2} d^{-1}, respectively). CO_2 fluxes directed to the atmosphere from the ecosystem are negative. The mean ecosystem respiration in moist tussock tundra was −4.4 ± 0.5 g C m^{-2} d^{-1}, and the mean ecosystem respiration in wet sedge tundra was −1.9 ± 0.1 g C m^{-2} d^{-1}. The rates of net ecosystem CO_2 exchange and ecosystem respiration were similar to rates observed in the field near the sites where the mesocosms were obtained [*Shaver and Chapin*, 1991; *Shaver et al.*, 1998]. The differences in CO_2 exchange between moist tussock tundra and wet sedge tundra are consistent with higher biomass and higher productivity of moist tussock tundra communities [*Bliss and Matveyeva*, 1992; *Oechel and Billings*, 1992; *Oechel et al.*, 1997].

[19] Rates of CO_2 exchange were relatively constant throughout the experiment because constant environmental conditions were maintained in the growth chamber, except for stepwise changes in light and temperature near the end of the experiment to simulate the end of the growing season. The net ecosystem CO_2 exchange rates measured at the time of labeling (data not shown) were not different (p = 0.8 and 0.5 for moist tussock tundra and wet sedge tundra, respectively) from the average net ecosystem CO_2 exchange rates during the experiment. There was no evidence that the ^{14}C-labeling had any effect on CO_2 exchange rates of the mesocosms. Mean CO_2 exchange rates in the long-term mesocosms tended to be higher than in the short-term mesocosms. This difference between the short-term and long-term mesocosms can be explained by differences in plant biomass; long-term mesocosms had higher biomass and were more productive.

[20] Mean (±standard error, n = 3) CH_4 emission rates over the length of the experiment were 0.1 ± 0.02 g CH_4-C m^{-2} d^{-1} (=133 mg CH_4 m^{-2} d^{-1}) in moist tussock tundra mesocosms and 0.3 ± 0.04 g CH_4-C m^{-2} d^{-1} (=400 mg CH_4 m^{-2} d^{-1}) in wet sedge tundra mesocosms (Figure 1). The mean emission rates for moist tussock tundra and wet sedge tundra are significantly different from each other ($p < 0.001$) as expected based on field observations [*Reeburgh et al.*, 1998].

[21] The CH_4 fluxes from moist tussock tundra mesocosms were higher than fluxes observed in the field [*Reeburgh et al.*, 1998] and consistent with other laboratory results [*Johnson et al.*, 1996]. Our moist tussock tundra mesocosms were more representative of tussock rather than intertussock areas. Tussock areas typically exhibit higher methane emissions than do intertussock areas [*Reeburgh et al.*, 1998], which is consistent with our observations. However, the CH_4 emission rates from the moist tussock tundra mesocosms were still 2–4 times higher than in situ measurements above tussock-only sites, probably because the mesocosms had higher soil moisture than in the field. The higher CH_4 emission rates measured in the moist tussock tundra mesocosms could be due to higher soil moisture in the mesocosms compared to in situ sites or to higher soil temperatures, as described below.

[22] The CH_4 emission rates from wet sedge tundra mesocosms were 2–4 times higher than emission rates observed in the field for these vegetation types [*Sebacher et al.*, 1986; *Crill et al.*, 1988; *Bartlett et al.*, 1992; *Schimel*, 1995; *King et al.*, 1998; *Reeburgh et al.*, 1998]. A possible explanation is that pore water movement was restricted within each mesocosm leading to more anaerobic conditions and a higher concentration of carbon compounds, which would increase CH_4 production, decrease CH_4 oxidation, and therefore increase net CH_4 emission. Higher soil temperatures in the mesocosms (15°C) compared to average field soil temperatures (~6°C) for 0–30 cm soil depth may also partially explain high CH_4 emission rates from the mesocosms of both vegetation types. The temperature dependence of CH_4 production is described by literature Q_{10} values of 1.3 to 28 [*van Hulzen et al.*, 1999] and a range of 5.3–16 specifically for temperate and subarctic peat soils [*Dunfield et al.*, 1993].

[23] The CH_4 emission rates from moist tussock tundra mesocosms increased gradually over the first part of the experiment probably due to increasingly anaerobic conditions and increased belowground plant productivity (Figure 1a). The same pattern of increasing CH_4 emission through the growing season has been observed from in situ measurements (W. S. Reeburgh, unpublished data, 1996). The rates of CH_4 emission declined in both moist tussock and wet sedge tundra mesocosms at the end of the experiment due to decreased

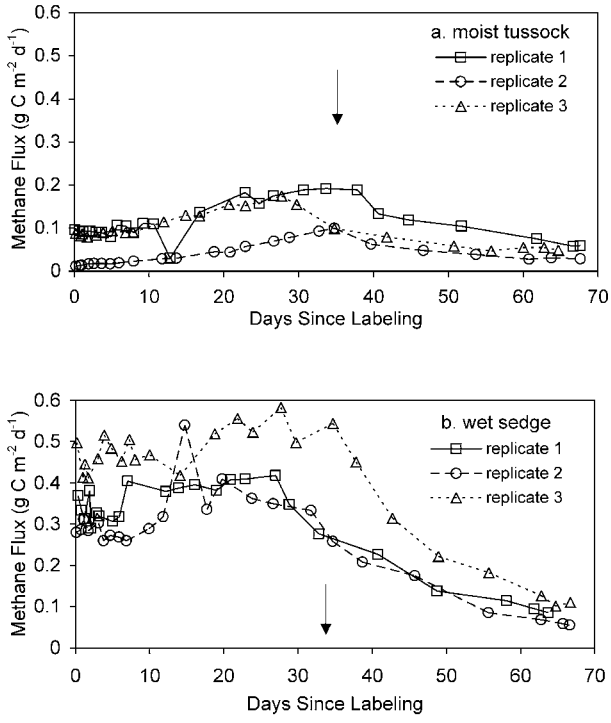

Figure 1. Methane flux measurements from (a) moist tussock tundra and (b) wet sedge tundra mesocosms. Measurements were made on three replicate mesocosms for 10 weeks following pulse-labeling. Arrows indicate time at which photoperiod and temperature decreased.

temperature and photoperiod in the growth chamber that simulated the end of the growing season.

3.2. Emissions of $^{14}CO_2$ and $^{14}CH_4$

[24] Emissions of $^{14}CH_4$ were first detected in all long-term moist tussock and wet sedge tundra mesocosms within 2 hours after labeling (Figure 2). The maximum ^{14}C activity of CH_4 emitted from both vegetation types occurred within the first week after labeling (Figure 2). These results agree with results from rice plant and wetland plant studies which indicated detection of photosynthate-derived CH_4 within 12–24 hours after labeling and maxima in $^{14}CH_4$ emission rates within one week of labeling [*Minoda and Kimura*, 1994; *Minoda et al.*, 1996; *Dannenberg and Conrad*, 1999; *Megonigal et al.*, 1999]. Because the carbon assimilated during labeling still had to pass through several pools before emission as CH_4, it is not surprising that the timing of maximum $^{14}CH_4$ emission varied between mesocosms. The detection of $^{14}CH_4$ emission within 2 hours of labeling indicates rapid conversion of plant-derived carbon to CH_4 and rapid transport of CH_4 out of the soil.

[25] The time course of $^{14}CO_2$ loss from the mesocosms was the same in moist tussock tundra as in wet sedge tundra (data not shown). Large proportions of recently assimilated carbon were very quickly respired as CO_2, probably through leaf respiration initially and then through root respiration. Decomposition of root exudates by soil microbes also contributed to fluxes of $^{14}CO_2$. Detection of $^{14}CO_2$ indicated that the pulse-labeling resulted in plant incorporation of the ^{14}C label and that some photosynthates were immediately respired.

3.3. Proportion of ^{14}C Emitted as Methane

[26] Overall, 1–5% of the plant productivity taken up as ^{14}C during labeling was emitted as $^{14}CH_4$ during the same growing season in moist tussock and wet sedge tundra mesocosms (determined by integration of traces in Figure 2). Emissions of $^{14}CH_4$ from the long-term mesocosms were 1.3, 2.2, and 2.5% of ^{14}C assimilation by moist tussock tundra and 1.3, 2.5, and 5.1% of ^{14}C assimilation by wet sedge tundra. The mean percentage (± standard error, n = 3) of ^{14}C label emitted as $^{14}CH_4$ was 2.0 ± 0.4% from moist tussock tundra and 2.9 ± 1.1% from wet sedge tundra. These results suggest that recently fixed carbon makes up a large proportion of the total carbon emitted as methane from these ecosystems. Our measurements of respired CO_2, emitted CH_4, pore water dissolved organic and inorganic carbon, plant biomass, and soil carbon accounted for greater than 70% of the total ^{14}C originally taken up. Our results are similar to results from other ^{14}C pulse-labeling studies which reported that <1% to 6% of assimilated ^{14}C was emitted as $^{14}CH_4$ [*Dannenberg and Conrad*, 1999; *Megonigal et al.*, 1999].

3.4. Origin of Methanogenic Substrates

[27] The carbon sources for CH_4 production in soil pore water are the dissolved organic carbon (DOC) and dissolved

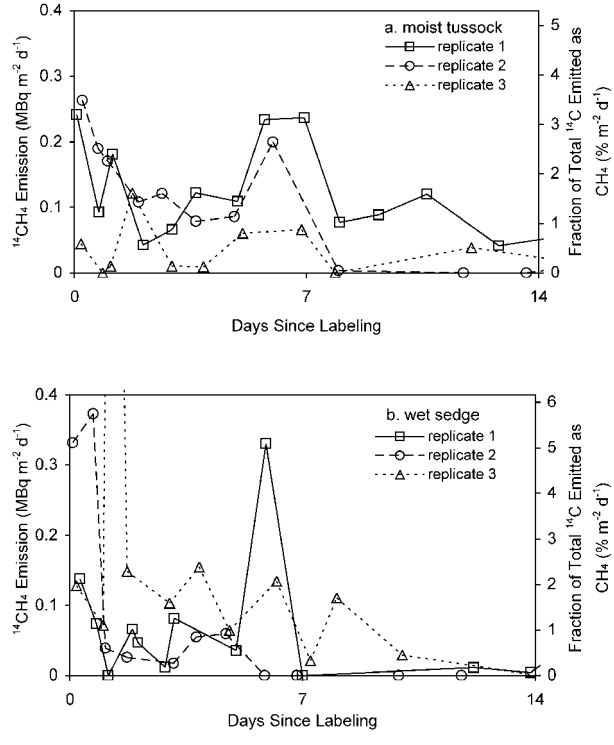

Figure 2. Emission of $^{14}CH_4$ from (a) moist tussock tundra and (b) wet sedge tundra mesocosms during the first 2 weeks following pulse-labeling. Measurements were made for 10 weeks (data not shown) on three replicate mesocosms of each vegetation type.

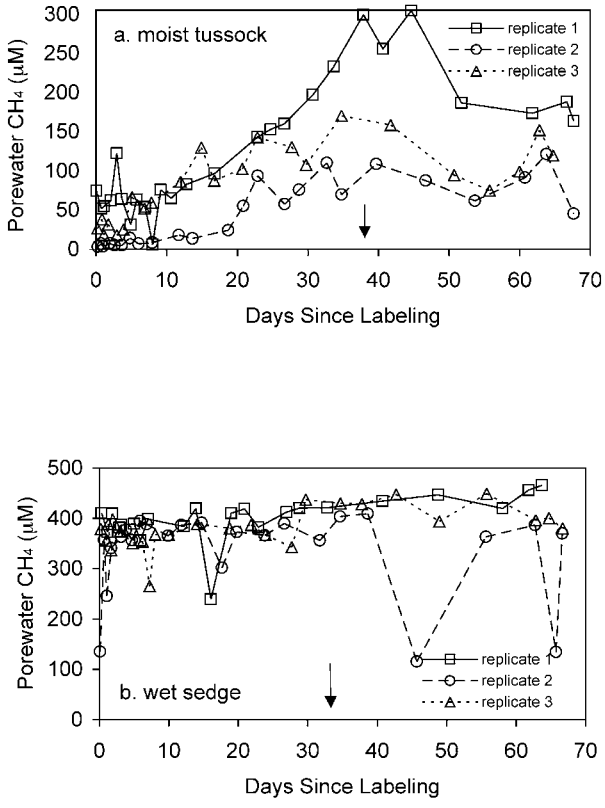

Figure 3. Pore water concentrations of dissolved CH_4 in (a) moist tussock tundra and (b) wet sedge tundra mesocosms. Arrows indicate time at which photoperiod and temperature decreased.

inorganic carbon (DIC) pools. Measurements of these pools were made throughout the experiment to monitor possible changes in overall methanogen substrate availability. Average pore water concentrations of DOC were approximately 9.3 mM and 1.6 mM in moist tussock tundra and wet sedge tundra mesocosms, respectively. Average pore water concentrations of DIC were approximately 8.8 mM and 10.2 mM in moist tussock tundra and wet sedge tundra mesocosms, respectively. Wet sedge tundra mesocosms had higher pore water concentrations of dissolved CH_4 (~400 μM) than did moist tussock tundra mesocosms (~100 μM; Figure 3), which is expected due to the anaerobic conditions in wet sedge tundra soils. Total pore water concentrations of DOC, DIC, and dissolved CH_4 were relatively constant throughout the experiment (data not shown).

[28] The ^{14}C activity of the DOC and DIC pools was measured to determine the availability of plant-derived carbon in the soil pore water for CH_4 production. The measurements of ^{14}C in DOC and DIC pools show that plant-derived carbon was quickly transferred to the soil pore water where it became available as a substrate for the soil microbial community [see *Loya et al.*, 2002]. The ^{14}C label was detected in all pore water carbon pools within hours of labeling. Dissolved $^{14}CH_4$ also appeared in the pore water of moist tussock tundra mesocosms within hours of labeling, indicating that conversion of plant-derived carbon to CH_4 occurred as soon as it was available in the soil. In moist tussock tundra mesocosms, the specific activity of pore water CH_4 reached a maximum within the first day after labeling and generally decreased thereafter (Figure 4a). The maximum specific activity of DIC occurred 4–8 days after labeling, and the maximum specific activity of DOC occurred 8 or more days after labeling. The specific activities of all dissolved pore water components in moist tussock tundra mesocosms did not change in response to changes in light and temperature at the end of the experiment.

[29] The specific activities of pore water components in wet sedge tundra were more variable than in moist tussock tundra. Detection of ^{14}C in DOC and DIC pools soon after labeling indicated that recently fixed carbon was quickly available as substrate for methanogenesis. The maximum specific activity of dissolved CH_4 occurred approximately 6 days after labeling (Figure 4b) and coincided with the maximum specific activity of DOC and DIC and with the highest emission of $^{14}CH_4$. These results suggest that fluxes between pore water carbon pools in wet sedge tundra occur rapidly. Transient increases in the specific activity of pore water carbon pools may have been a byproduct of plant translocation of previously fixed carbon from aboveground tissues to belowground tissues in response to lowered temperature and light levels at the end of the experiment.

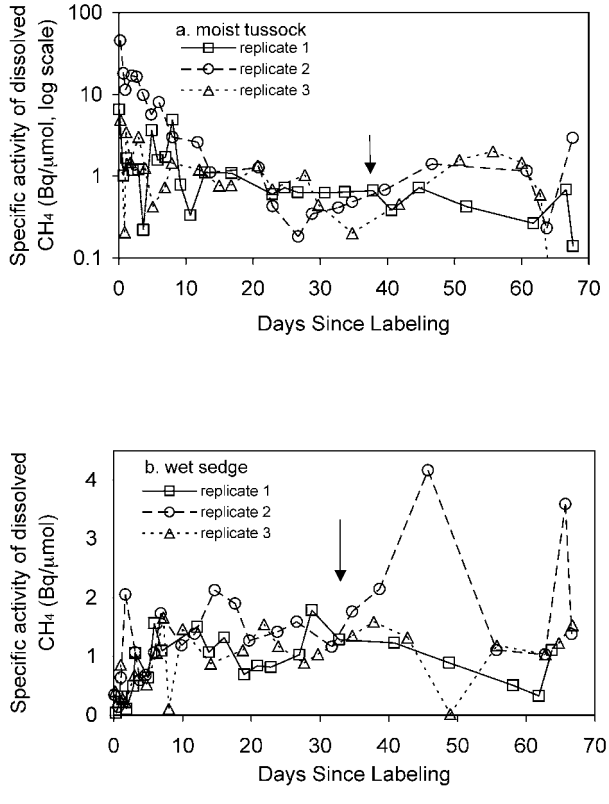

Figure 4. Specific activity of dissolved CH_4 in pore water in (a) moist tussock tundra (please note log scale of *y* axis) and (b) wet sedge tundra mesocosms. Arrows indicate time at which photoperiod and temperature decreased.

4. Summary

[30] The results of this pulse-labeling study show that transport of carbon through the plant-soil system and emission as CH_4 occurs quickly. The ^{14}C label first appeared as emitted CH_4 within 2 hours of labeling in both wet sedge and moist tussock tundra. The maximum flux of $^{14}CH_4$ occurred within one week after the pulse-labeling. Such fast turnover of carbon means that changes in plant uptake of carbon on short timescales (days to weeks) may be important controls on CH_4 emissions. Similar results were reported for rice systems [*Minoda and Kimura*, 1994; *Minoda et al.*, 1996; *Dannenberg and Conrad*, 1999], which suggests that the same processes leading to CH_4 emission are occurring in very different ecosystems and also suggests that it may be possible to model CH_4 emissions for various ecosystems using relationships between net ecosystem CO_2 exchange and CH_4 emission.

[31] Our results show that recent photosynthates are an important carbon source for emitted CH_4. Overall, approximately 2–3% of ^{14}C incorporated at mid season was emitted as CH_4 in wet sedge and moist tussock tundra. Carbon taken up by plants as $^{14}CO_2$ was traced through the plant-soil system of the mesocosms and was measured as emitted $^{14}CH_4$. However, we did not find a simple relationship between plant productivity and CH_4 emission, as described by *Whiting and Chanton* [1993]. That relationship predicts higher CH_4 emission with higher plant productivity, but our measurements show lower CH_4 emission from moist tussock tundra, which demonstrated relatively higher plant productivity. Artifacts caused by the laboratory setting of this study (e.g., stagnant water column) probably account for the observed pattern.

[32] By using a carbon tracer we have directly measured conversion of plant photosynthates to emitted CH_4. These are among the first experimental results to quantitatively demonstrate a mechanism linking plant productivity and CH_4 emission. The gas exchange measurements demonstrate that, although the amount of carbon exchanging with the atmosphere as CO_2 or CH_4 at any given point in time is small, the integrated amounts over the period of a growing season are significant. We measured the conversion of 2–3% of recent plant productivity to CH_4 emission in this study. The average measured CH_4 emission from wet sedge and moist tussock tundra was 12.75 g C m^{-2} for the growing season. Average plant productivity in these tundra ecosystems is 647 g biomass m^{-2} [*Shaver and Chapin*, 1991], and biomass is composed of approximately 50% carbon. Converting 3% of average plant productivity to CH_4 emission results in a flux of CH_4 originating from recent plant productivity of 9.71 g C m^{-2} during a growing season. Thus, using our results and average values for plant productivity in back-of-the-envelope calculations, we estimate that greater than 75% of average CH_4 emissions from these ecosystems originate from recently fixed carbon. These observations support radiocarbon evidence that recently fixed carbon is a significant carbon source of emitted CH_4 [*Wahlen et al.*, 1989; *Chanton et al.*, 1995; *King and Reeburgh*, 2002]. Our observations suggest that the relatively small pool of recently fixed carbon is a primary source of carbon for the CH_4 cycle in arctic tundra, whereas the large carbon stores in tundra soil (21.8 kg m^{-2}, [*Billings*, 1987]) appear to contribute minimally to CH_4 emission over seasonal timescales. Our results imply that recently fixed carbon represents a significant fraction of annual net CH_4 emission from these ecosystems and that environmental changes affecting ecosystem productivity may heavily impact the global CH_4 cycle.

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