Assessment of winter fluxes of CO_2 and CH_4 in boreal forest soils of central Alaska estimated by the profile method and the chamber method: a diagnosis of methane emission and implications for the regional carbon budget

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ABSTRACT

This research was carried out to estimate the winter fluxes of CO_2 and CH_4 using the concentration profile method and the chamber method in black spruce forest soils in central Alaska during the winter of 2004/5. The average winter fluxes of CO_2 and CH_4 by chamber and profile methods were 0.24 ± 0.06 (SE; standard error) and 0.21 ± 0.06 gCO_2-C/m^2/d, and 21.4 ± 5.6 and 21.4 ± 14 μgCH_4-C/m^2/hr. This suggests that the fluxes estimated by the two methods are not significantly different based on a one-way ANOVA with a 95% confidence level. The hypothesis on the processes of CH_4 transport/production/emission in underlying snow-covered boreal forest soils is proven by the pressure differences between air and in soil at 30 cm depth. The winter CO_2 emission corresponds to 23% of the annual CO_2 emitted from Alaska black spruce forest soils, which resulted in the sum of mainly root respiration and microbial respiration during the winter based on the $\delta^{13}CO_2$ of $-22.5‰$. The average wintertime emissions of CO_2 and CH_4 were 49 ± 13 gCO_2-C/m^2/season and 0.11 ± 0.07 gCH_4-C/m^2/season, respectively. This implies that winter emissions of CO_2 and CH_4 are an important part of the annual carbon budget in seasonally snow-covered terrain of typical boreal forest soils.

1. Introduction

Winter CO_2 flux is a large carbon source in snow-covered ecosystems, such as arctic tundra (Zimov et al., 1993; Oechel et al., 1997; Fahnesock et al., 1999), alpine and subalpine regions (Solomon and Cerling, 1987; Sommerfeld et al., 1993; Brooks et al., 1996; Mast et al., 1998; Wickland et al., 1999, 2001), boreal forests (Winston et al., 1997) and temperate forests (Hirano, 2005; Takagi et al., 2005). Winter flux measurements have rarely been observed due to the difficulty of making measurements in snow-covered ecosystems; furthermore, winter CO_2 emissions through the snowpack were assumed to be negligible or were underestimated (Raich and Schlesinger, 1992; Raich and Potter, 1995). The magnitude of the flux through the snowpack to the atmosphere is smaller than that estimated in the growing season. However, the contribution of winter flux corresponded to 10–30% of the annual soil respiration in alpine and tundra regions, where the snow-covered period is more than 200 days (Oechel et al., 1997; Mast et al., 1998; Wickland et al., 1999, 2001). These findings suggest that the winter CO_2 emission to the atmosphere might contribute a significant portion of the annual carbon budget.

Winter CH_4 flux also acts as a large carbon source in temperate peatlands (Dise, 1992; Shannon and White, 1994), subalpine wetlands (Mast et al., 1998; Wickland et al., 1999, 2001), boreal peat bogs and peatlands (Alm et al., 1999; Panikov, 1999; Panikov and Dedysh, 2000) and boreal forests (Whalen and Reeburgh; 1988; 1992). These ecosystems are well known as CH_4 sources under anaerobic environments. Conversely, the soils under aerobic environments have also become famous as CH_4 sinks (e.g. CH_4 oxidation) in various ecosystems (Born et al., 1990; Dörr and Münnich, 1990; Deñas et al.,

1999). However, winter CH_4 emission and production in the snow-covered boreal forest soils are still poorly understood. The winter emissions of CH_4 as well as CO_2 in boreal forest and tundra soils might be significant carbon sources on regional and global scales.

Within the anaerobic environments of Alaska bogs and tundra wetlands, Duddleston et al. (2002) and Ström et al. (2003) demonstrated that acetate was the primary organic terminal product of anaerobic decomposition, and acetate was ultimately produced by this reduction: ($CH_3COOH \rightarrow CH_4 + CO_2$; $CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$). The produced CO_2 and CH_4 gases were released to the atmosphere by three main pathways: molecular diffusion, bubble ebullition and through emergent vascular plants during the growing season. During the winter, when boreal forest soils were frozen with time, the frozen soils sometimes changed to suboxic/anoxic environments. CO_2 and CH_4 produced in the frozen soil may be emitted to the atmosphere only by vascular plants; in this case tussock tundra plants at the tops of tussocks. Here, I provide a novel report of net methane emission through the vascular plants over tussock soils to the snowpack in a boreal forest of central Alaska. This study has been conducted in parallel with tower observations that have monitored the fluxes of CO_2 and CH_4 between the boreal forest and the atmosphere since November 2002 on the wooded campus of the University of Alaska Fairbanks.

The purposes of this study are to (1) compare the profile method and the chamber method for the winter fluxes of CO_2 and CH_4 in snow-covered boreal forest soils, such as the tussock and sphagnum moss regime of typical boreal forest soils, central Alaska, (2) clarify the emission of CO_2 and CH_4 in soil, snowpack, and tower air samples by using stable carbon isotopes of CO_2 and CH_4, and (3) evaluate the contribution of winter carbon to the annual regional carbon budget in a typical boreal forest of interior Alaska.

2. Materials and methods

2.1. Sampling locations and methods

The study site was located at a tower station (64°52′N, 147°51′W) on the campus of the University of Alaska Fairbanks in central Alaska. The site represents typical boreal forest and the dominant vegetation there is black spruce, *Picea mariana*, which are 6 m high. The forest floor is a mixture of tussocks, vascular plants, shrubs, sphagnum and feather mosses, and lichen. Specifically, those plants include *Betula glandulosa*, *Ledum palustre*, *Vaccinium vitis-idaea*, *Carex lugens*, *Sphagnum spp.*, *Thuidium abietinum* and *Cladina stellaris* on the permafrost regime. The densities of black spruce, sphagnum and feather mosses are 4500 trees/ha, 4.4 individual/cm^2 and 0.8 individual/cm^2, respectively. The vegetation characteristics of the site are shown in Table 1.
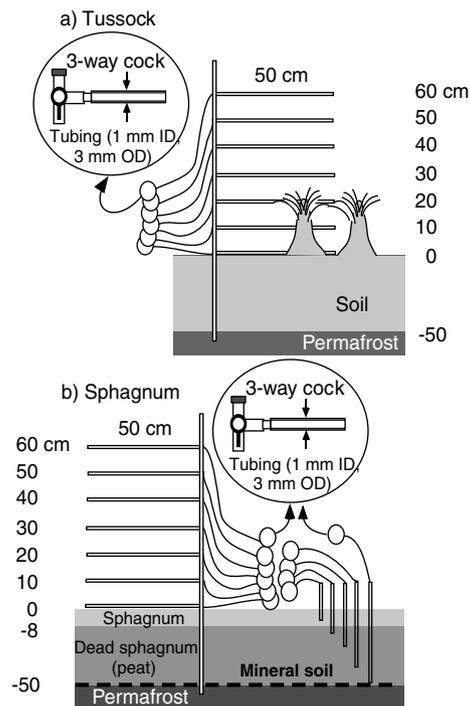

Fig. 1. Sampling scheme in (a) tussock and (b) sphagnum moss regimes of the boreal forest, central Alaska. Sampling depths in snowpack and soil were described in text.

The annual average air temperature was –3.3 °C, and the average temperature in July was 16.4 °C during the last 40 years (1964–2004). Annual average precipitation at the site is 270 mm and the average precipitation in August is 47 mm during the last 40 yr.

The average height of each tussock is 20 cm. The soil surrounding tussocks is so compacted clayey that vascular plants only inhabit the top of tussocks (Fig. 1a). Hence, we could not collect soil air samples in the tussock regime. Sphagnum moss photosynthesizes and has a large water-holding capacity in the surface soil. Individual living moss strands are 7–8 cm long, and the dead moss in the peat phase exists to depths of 50 cm below the living moss. The soils are characterized by an organic matter mat (50 cm in depth), a loess parent material (<1cm depth) and permafrost. The mineral soil consists of loess that was blown from the Alaska Range and fluvial plains during the Holocene. The mineral loess soil extends at least to the top of the permafrost. Permafrost exists beneath the covered mineral loess soil (Fig. 1b). Tussock and sphagnum moss in Alaska's boreal forest completely freezes ordinarily in late January (V. Romanovsky, personal communication, 2003). The contents of biomass, carbon and nitrogen in aboveground and belowground vegetations in the black spruce forest of central Alaska are listed in Table 1.

The flux chamber was deployed within a few metres of the in-soil probes at sphagnum moss and the chamber diameter was 50 cm with a cross section area of 0.2 m^2 and height of 10 cm

Table 1. The contents of biomass, carbon, and nitrogen in abovegound and belowground of mass regime in the black spruce forest, central Alaska

Component	Blomass(kg/m^2)		Carbon content (kgC/m^2)		Nitrogen content (kgN/m^2)	
	Mean	SD	Mean	SD	Mean	SD
Black spruce (n = 3)	0.10	0.061	0.055	0.033	0.002	0.002
Moss (n = 5)	0.47	0.104	0.216	0.05	0.002	0.001
A0 layer (n = 4)	5.57	0.717	2.564	0.33	0.056	0.007

(plastic material). The chamber top has two fixed nozzles (1 mm ID and 3 mm OD; 300 cm long); one is used to collect air samples and the other is used to balance the pressure between the inside and outside chamber. The chamber was placed over the snowpack surface at the beginning of each measurement series as not to disrupt the surface snowpack at an interval of 2 m during a steady state condition. Four air samples were drawn from the headspace in the chamber at approximately 10 min intervals after the deployment. The 10 mL samples are collected using a 50 mL plastic syringe at a rate of 5 mL per minute. The flux was calculated from the gradient of the relationship between the concentration variations of a gas and sampling time. The correlation coefficient was above 0.99 and the curve was linear (Kim and Tanaka, 2003).

Stainless steel tubes (1 mm ID, 3 mm OD; 50 cm long; Swagelok, USA) used for concentration profiles of CO$_2$ and CH$_4$ were horizontally set on the ground between the tussocks (Fig. 1a) before the snow fell. The tubes were installed in sphagnum moss in March 2005. The stainless steel tube was connected with polyethylene tubing (1 mm ID and 3 mm OD; 150 cm long; Nalgene, USA), and the end of the tubing was attached to a three-way stopcock. The sampling depths were 0, 10, 20, 30, 40, 50 and 60 cm above the soil surface.

The tubes under the soil were vertically built at the sphagnum moss regime. The stainless steel tube, the polyethylene tubing and the three-way stopcock were connected using the same method executed at the tussock area (Figure 1b). The sampling depths were 5, 10, 20, 30 and 40 cm below the surface. The sampling period was an interval of 10 d and consisted in part of snow pit-wall observations, where we examined the snow crystals, snow density and snow temperature. However, the sampling strongly depended on local weather conditions. The monitoring of soil temperature and moisture has been carried out since June 2004 for the sphagnum moss regime. The monitoring of pressure at 2 m above the soil surface and at soil 30 cm depth using loggers (HOBO Pressure, Onset Instrument Corp., USA) and of temperature (HOBO 4-Channel External, Onset Instrument Cop.) at 5, 10, 30, 40 and 50 cm depth below the soil surface and at 85 cm above the surface have been conducted since October 2005 at the sphagnum moss site.

A tower site was built at the typical black spruce forest on the discontinuous permafrost in November 2002; the tower is 10 m high and the sampling inlets are at the 2 and 8 m levels.

The height of the canopy is 6 m. The ambient air samples were collected to a pre-vacuum vial (10 mL, GL Science, Japan) using the pump from two levels.

Ten millilitres of air sample from soil and snowpack were collected in the vials with a plastic syringe at a rate of 5 mL per minute at the tussock and sphagnum moss regimes. The vials were transferred to the laboratory for the analyses of CO$_2$ and CH$_4$ concentration. The air samples of soil, snowpack and tower were separately collected for the stable carbon isotope of CO$_2$ and CH$_4$ every 2 to 3 weeks.

2.2. Analyses of the concentrations and stable carbon isotopes of CO$_2$ and CH$_4$

These gas-filled vials were transported to the laboratory for later CO$_2$ and CH$_4$ analyses. Concentrations of CO$_2$ were measured with a gas chromatograph equipped with a thermal conductivity detector (TCD-GC, GC-14B, Shimadzu Ltd., Japan) with a column packed with Porapack Q (80/100 mesh). Calibration was done with a series of standard gases containing 338 ± 7, 491 ± 10 and 5000 ± 100 ppmv of CO$_2$ in synthetic air (Scotts Gas Co., USA). Precision with the calibration range was usually better than 1%.

Concentrations of CH$_4$ were measured with a gas chromatograph equipped with a flame ionization detector (FID-GC, GC-14B, Shimadzu Ltd., Japan) using a column packed with Molecular Sieve 13X (80/100 mesh). Calibration was done with a series of standard gases containing 0.79 ± 0.016, 1.61 ± 0.020 and 2.43 ± 0.048 ppmv of CH$_4$ in synthetic air (Scotts Gas Co.). Precision with the calibration range was usually better than 2%.

In order to analyze the contents of carbon and nitrogen from above- and below-ground vegetations, after each bulk sample was dried at 85 °C and the sample was homogeneously ground into powder using agate mortar and pestle, the contents of biomass, carbon and nitrogen in each sample were determined using a CN analyzer (SUMIGRAPH NC-1000, Japan). The analytical error was usually less than 0.1%.

The stable carbon isotope compositions of CO$_2$ and CH$_4$ were measured using a GC/C/IRMS [Gas Chromatography (Varian 3400)/Combustion/Isotope Ratio Mass Spectrometry (Finnigan MAT delta S)] system (Nakagawa et al., 2002; Ishimura et al., 2004). In this system, air samples for determination of ^{13}C of CO$_2$ were injected directly without any pre-treatment. Components

in the injected air samples were separated through a capillary column (Pora Plot Q, Chrompack). Column-separated CH_4 was oxidized in a CuO furnace (800 °C) to CO_2 and H_2O. After removal of H_2O from the mixture by a −100 °C cryogenic trap, the purified CO_2 was introduced into a mass spectrometer for $\delta^{13}C$ measurements. To determine ^{13}C in CO_2, CuO was not heated; the CO_2 was separated using a gas chromatograph and then introduced into the IRMS. The precision of the techniques was estimated to be ±0.10‰ for $^{13}CO_2$ and ±0.30‰ for $^{13}CH_4$.

Carbon isotope ratios in delta notation are calculated as:

$$\delta = \left[\frac{R_{sample}}{R_{standard}} - 1 \right], \tag{1}$$

where R is the molar ratio of heavy to light isotope ($^{13}C/^{12}C$), and the international standard for air samples of soil, snowpack and tower is CO_2 and CH_4 from the Vienna Pee Dee Belemnite (VPDB) standard. The δ values are conveniently presented in parts per thousand (‰).

CH_4 flux measured by tower observation was estimated by the aerodynamics flux method of Ueyama et al. (2006), who modified the various parameters that were suitable for the Alaska boreal forest, while the vertical wind is weak and the atmospheric stability is near neutral during the winter season.

2.3. Flux measurements of CO_2 and CH_4 by the chamber method

The flux of CO_2 and CH_4 from the snowpack surface can be calculated as the rate of concentration increases in the chamber:

$$J = \frac{dC}{dt} \cdot \frac{V}{A}, \tag{2}$$

where J is the flux of a gas, V is the volume of air in the chamber, m^3, A is the area covered by the chamber, m^2, C is the chamber concentration of a gas, ppm and dC/dt is the rate of concentration change in the chamber air for each gas, ppm/m.

The fluxes of CO_2 and CH_4 estimated by the chamber method were calculated with eq. (2). The average fluxes of CO_2 and CH_4 were 0.24 ± 0.05 $gC/m^2/d$ [coefficient of Variation (CV): 22%] and 0.023 ± 0.009 $mgCH_4$-$C/m^2/h$ (CV: 41%) during the winter period (from DOY 284 to DOY 484), respectively. The fluxes of CO_2 and CH_4 were measured at an interval of 2 hr for October 8, as is shown in Figure 2. The fluxes of CO_2 and CH_4 determined by the chamber method were found to be emitted through the snowpack to the atmosphere, suggesting that the soil may play a significant role as a source of CO_2 and CH_4 in the boreal forest. The fluxes of CO_2 and CH_4 showed similar diurnal variations during a 24 hr flux measurement. The diurnal variation of CH_4 flux similarly showed continuous CH_4 flux measured by the tower (shown in Fig. 2b during 24 hr CH_4 flux observation), indicating that the black spruce forest soil may be a CH_4 source during the winter. CH_4 oxidation was observed at a sphagnum moss carpet of black spruce forest soils and at deciduous aspen and birch forest soils (Whalen et al., 1991; Kim and Tanaka,
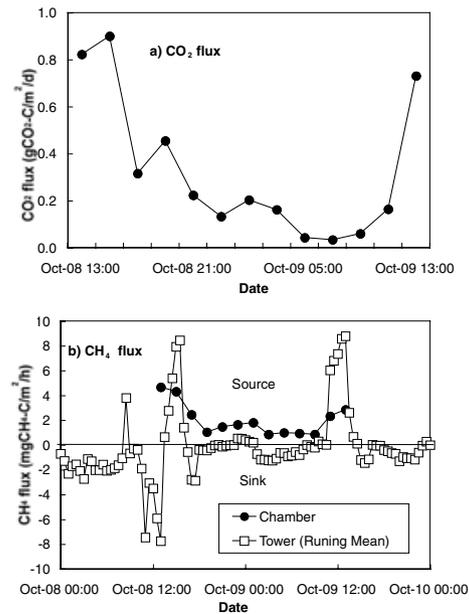

Fig. 2. Diurnal variations of (a) CO_2 flux by the chamber method and (b) CH_4 fluxes by the chamber method and tower observation in the early winter. These suggest that boreal forest soils play a significant role as a source during the winter.

2003) during the growing season, such as during annual CH_4 flux monitoring by the tower observation. Ueyama et al. (2006) reported the diurnal variations of CH_4 flux measured by tower in black spruce forest of central Alaska; a weak emission on snowpack, an obvious emission around spring thaw, and CH_4 uptake in the growing season.

2.4. Flux measurements of CO_2 and CH_4 by the profile method

The fluxes of CO_2 and CH_4 through snowpack to the atmosphere under the assumption that the surface soil was the top of a tussock were obtained by applying the following equation under a steady state condition described by Fick's first law:

$$J = D \cdot \frac{dC}{dt} \cdot \frac{V}{A} \cdot \tau \cdot \theta, \tag{3}$$

where J is flux of a gas, D is the diffusivity of a gas corrected only for the in-situ temperature within the snowpack, cm^2/s, dC/dt is the vertical concentration gradient of a gas observed within the snowpack, ppmv/cm, τ is tortuosity and θ is the snow porosity. Porosity was calculated from the density of ice ($\sigma_{ice} = 0.91$) and the water contents of the snowpack over the gradient interval. Tortuosity is difficult to measure and, in soils, usually is described as a function of porosity with values ranging from $\theta^{1/3}$ to $\theta^{2/3}$ (Striegl, 1993). In this study, the tortuosity of the snowpack was estimated from the theoretical relation $\tau = \theta^{1/3}$ (Millington, 1959), which yielded values ranging from 0.74

to 0.92. These values are similar to the range of 0.68 to 0.90 through the whole observation period for a boreal forest snowpack in central Alaska. Sommerfeld et al. (1993) and Mast et al. (1998) reported similar data (0.74–0.90) in a subalpine and alpine snowpack, Wyoming. The effective diffusivities of CO_2 and CH_4 through the whole snowpack calculated in this study were 0.042 to 0.074 cm^2/s and 0.062 to 0.110 cm^2/s under an assumption of 1 atm at site during the winter, respectively. Mast et al. (1998) calculated the effective diffusivities of both gases to be 0.061 to 0.144 cm^2/s for CO_2 and 0.086 to 0.203 cm^2/s for CH_4 at subalpine soils of Colorado, during 3-year study. These data are much higher than data in our study. This suggests the different physical factors at the observed site, such as snow transformation with time, snow depth, temperature gradient through the snowpack and so on.

During the winter of 2004/5, which featured a thicker snowpack, the average fluxes of CO_2 and CH_4 estimated by the profile method were 0.22 ± 0.05 gCO_2-C/m^2/d (CV: 23%) and 0.018 ± 0.005 mgCH_4-C/m^2/hr (CV: 28%), respectively.

3. Results and discussion

3.1. Temporal variation of CO_2 and CH_4 concentration in snowpack and soil

Temporal variations of CO_2 and CH_4 concentrations at the University of Alaska Fairbanks tower site in central Alaska within the snowpack at the tussock site (Figs. 3a and 3b) and at soil of sphagnum moss site (Figs. 3c and 3d) during the winter of 2004/5 (using Ocean Data View software of Schlizer, 2004) are shown in Figure 3. The data for above the surface (depth $= 0$) described the concentrations of CO_2 and CH_4 from the bottom soil of intertussocks. Because intertussock zones feature very hard and compact soil, the soil air probes could not be mounted in the soil. The vertically empty column in each figure indicates no observation due to extreme cold (> -20 °C), and the horizontally empty row in Figures 3c and 3d denotes no data because of frozen ground layers 10 and 30 cm in depth. However, the data at 5, 20 and 40 cm depths were shown in Figures 3c and 3d, suggesting the unfrozen layer may be due to a peat phase of thick dead Sphagnum moss, which had a higher pore space than soil.

The concentration gradients of CO_2 and CH_4 within the snowpack at the tussock site are shown in Figure 4, indicating that the depth that shows a maximum concentration is 20 cm at the top of the tussock and emissions of these gases occur through the vascular plant at the tussock tops during winter. The gradients and the depth of CO_2 and CH_4 have different patterns with time. While the maximum CO_2 emits through the vascular plants to the snowpack during the early winter (DOY 322 and 356), the maximum CH_4 emits during the early and middle of winter (DOY 312, 322 and 386). This suggests that organic matter (e.g. acetate) in the rhizosphere at the tussock is decomposed into CO_2 and CH_4 during the early winter and then CO_2 is reduced to CH_4 during the middle winter (Duddleston et al., 2002; Ström
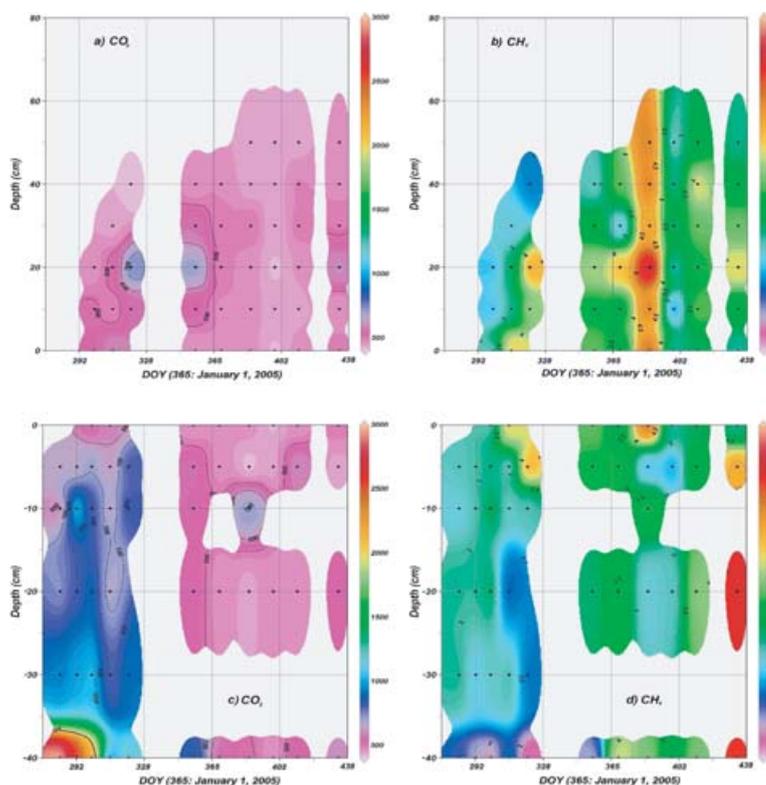

Fig. 3. Temporal variations of concentrations of (a) CO_2 and (b) CH_4 in snowpack above the tussock surface and of (c) CO_2 and (d) CH_4 in soil below the sphagnum moss mat. The vertically empty column indicates no observation due to the severe weather condition, and the horizontally empty row denotes no data owing to the frozen soil, which has no pore space. The contour interval is 100 ppmv within the snowpack for CO_2 profile, and 0.5 ppmv within the snowpack CH_4 profile during the winter season.
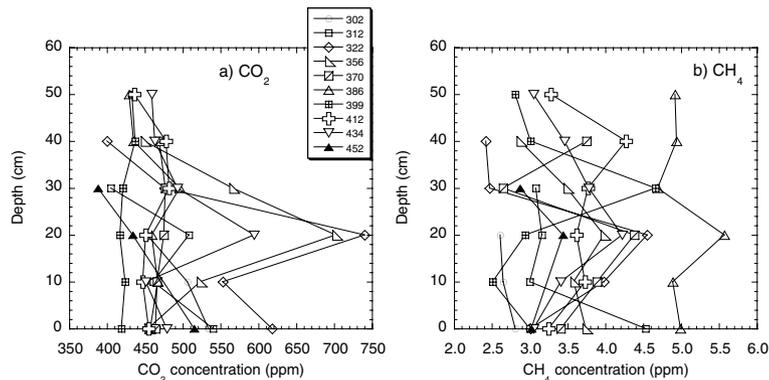
et al., 2003). Or this suggests that the pressured CO_2 and CH_4 that were produced at a sphagnum site may be transported to a tussock and emitted to the snowpack through tussock vascular plants.

The average tussock height is approximately 20 cm at the site, and the vegetation at the top of the tussocks is vascular plants. Precipitation levels were as follows: snow began to accumulate in early October 2004 and the maximum depth was 78 ± 5 cm (n = 10) in the middle of February 2005. The higher CO_2 concentration (>2000 ppm) showed at 40 cm depth of soil in the early winter (DOY 284 to DOY 302). In sphagnum moss, the CO_2 concentration varied from 420 ppm at 5 cm (DOY 386) to 1200 ppm at 40 cm (DOY 322) throughout the whole period. In the snowpack over tussocks, CO_2 concentration ranged from 370 to 740 ppm at 20 cm (DOY 322). In tower air, the CO_2 concentration varied from 471 ppm (DOY 357) to 554 ppm (DOY 388), which is much higher than that (380 ppm) of background air at the NOAA/CMD site in Barrow. Oechel et al. (1997) demonstrated the contribution of winter CO_2 emission to the atmosphere measured in tussocks and wet sedges of arctic tundra soil.

In the snowpack, a higher CH_4 concentration (6.4 ppm) was recorded at 20 cm depth on the top of tussocks (DOY 386). In the moss regime, the concentration ranged from 1.8 ppm at 40 cm (DOY 322) to 4.6 ppm at 5 cm (DOY 434). In tower observations, the CH_4 concentration varied from 2.7 ppm (DOY 436) to 3.6 ppm (DOY 357), which is higher than that (1.9 ppm) of background air at NOAA/CMD in Barrow. Because most of the winter CH_4 flux measurements were conducted in anaerobic environments such as bogs, fens, peatlands, and wetlands, the winter CH_4 emission is still poorly understood in typical boreal forest soils of central Alaska. Sommerfeld et al. (1993) indicated that CH_4 concentration profiles in alpine and subalpine snowpack tended to decrease with depth in Wyoming during a 2 yr winter season, demonstrating that alpine and subalpine aerated soils act as a sink of atmospheric CH_4. Whalen et al. (1991), Whalen and Reebugh (1996) and Kim and Tanaka (2003) reported CH_4 oxidation in typical boreal forest soils, central Alaska, during the growing season. Therefore, it is necessary to make clear the origins of CO_2 and CH_4 in boreal forest soils using the stable isotopic discrimination of $^{13}CO_2$ and $^{13}CH_4$ from the air
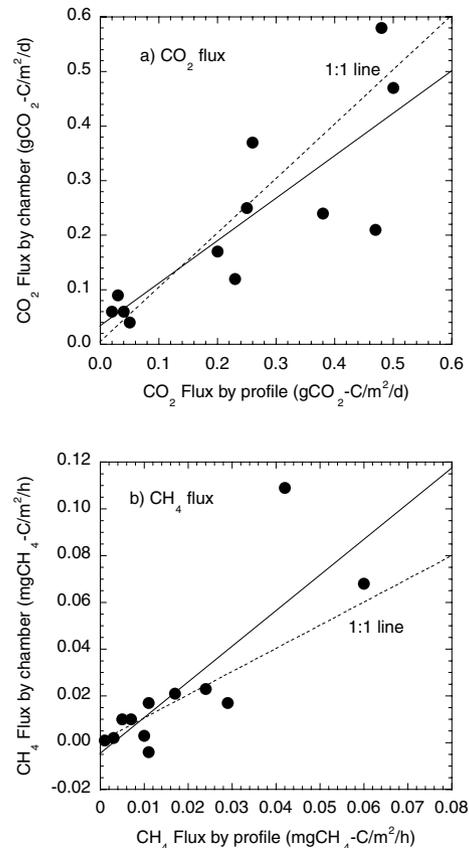

Fig. 5. Comparisons of (a) CO_2 flux and (b) CH_4 flux by the chamber method and the profile method, indicating that correlation coefficients (R) are 0.83 for CO_2 and 0.83 for CH_4, respectively.

samples of soil, snowpack and tower, as will be described in a chapter 3-3.

3.2. Winter fluxes of CO_2 and CH_4

Comparison of the winter fluxes of CO_2 and CH_4 measured by the chamber and the profile methods are shown in Figure 5. The fluxes estimated by two methods are not significantly different based on a one-way ANOVA with a 95% confidence level.

However, the flux ratios of profile to chamber methods were 1.28 ± 0.25 (CV 20%) for CO_2 (ratio range: 0.45–3.00) and 0.84 ± 0.22 (CV 21%) for CH_4 (range: -0.36–2.60) throughout the winter period, which means the CO_2 flux estimated by the profile method is somewhat higher than that estimated by chamber method, and CH_4 flux estimated by the chamber method is higher than that estimated by profile method. This indicates that the winter emissions of CO_2 and CH_4 estimated by chamber method explained 68% and 68% of the emissions of CO_2 and CH_4 evaluated by the profile method at boreal forest soils during winter. Mast et al. (1998) reported that the chamber fluxes were consistently lower than the gradient fluxes by nearly a factor of 3, resulting from the lower error in estimates of gradient fluxes, an inadequate seal between the chamber and the snow surface, and strong gust winds at the snow surface. However, because other studies were conducted at the Rocky Mountain sites, where season snowpacks typically reach several metres in depth, the snow surface did not reach a steady state condition due to strong winds, and thus deep snow depth and the estimates in chamber fluxes at subalpine sites might result in underestimations. In boreal forest soils, Winston et al. (1997) suggested that the winter CO_2 flux was a higher chamber flux than the gradient flux due to the presence of melt channels and tree wells.

The CH_4 fluxes estimated by the chamber and the profile methods and the tower observations were emitted through the snowpack to the atmosphere during the winter, as shown in Figure 6. Based on the monitoring CH_4 flux at the tower, these soils were a source of CH_4 during the winter of 2003/4, whereas the flux was a sink during the growing season in these typical black spruce forest soils. Moreover, in winter, CH_4, estimated by both methods, was also emitted to the atmosphere, suggesting that the soil-originated CH_4 is a source of atmospheric CH_4 during the winter in boreal forest soils of central Alaska. Conclusively, the boreal forest soils should be a significant carbon source
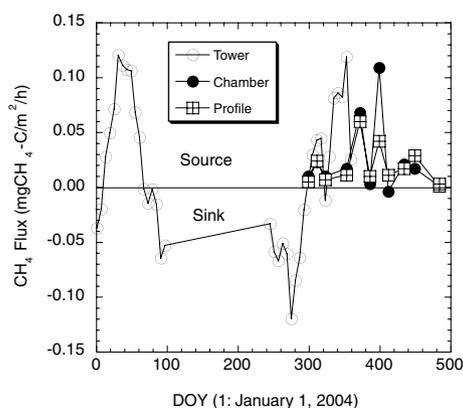
reservoir in the form of CH_4 as well as CO_2 during the winter. We suggest a possible transport mechanism of the soil-originated CH_4 through the snowpack to the atmosphere. In early winter, the moss surface temperatures at 5 cm and 100 cm dropped below freezing and rose above freezing on 25 August 2004 (DOY 237); the temperature ranged from -4.8 °C at 04:00 to 25.6 °C at 14:00 ALT (Alaska local time), as shown in Figure 7a. Also, the sphagnum moss carpet began to freeze on 29 September 2004 (DOY 272). The moss column was completely frozen to 30 cm below the surface on 19 October 19 2004 (DOY 290), as shown in Figure 7b. The soil began to freeze on 12 October 2004 (DOY 285) at 10 cm below the surface and on 17 October 2004 (DOY 290) to 30 cm below the surface, indicating that the freezing rate between 10 cm and 30 cm was 4 cm/d (Fig. 7c). Therefore, the cold ambient temperature freezes the moss carpet fromthe surface downward, while permafrost freezes the dead moss layer (peat) upward with time, indicating a possible hypothesis of soil-originated CH_4 transport and emission to the atmosphere: (1) the pressure in the carpet is steadily enhanced by the freezing from both directions with time, (2) the frozen moss surface seals off the air-filled pore space, (3) the atmospheric O_2 does not reach the soil any more, (4) the soil CO_2 begins to reduce to CH_4 under suboxic/anoxic environments in the sphagnum moss, (5) the pressured CH_4 (higher CH_4 concentration) moves to the tussock and finally (6) the higher concentration CH_4 releases to the vascular plants at a top of a tussock (20 cm) and then the atmosphere through the snowpack, as shown in Figure 3b. The comprehensive CH_4 transport at sphagnum moss and tussock regimes in the black spruce forest soils is schematically shown in Figure 8. This implies the possibility of soil-originated CO_2 and CH_4 emissions through the vascular plants to the snowpack and to the atmosphere during the winter. This finding of CH_4 emission through the tussock to the snowpack in boreal forest soils of central Alaska reveals another contribution to the carbon budget at a regional scale. However, this phenomenon of winter CH_4 emission is not continuous, but intermittent. The temporal variations of pressures at air and in soil 30 cm depth during a month of 2005, and of soil temperatures at soil 5, 10 and 30 cm depth below the surface, are shown in Fig. 9. The difference in pressure, the pressure at soil 30 cm subtracted from air pressure, during <30 d tends to decrease with time. This demonstrates that the pressure at soil 30 cm depth is enhanced. Also, the soil temperature at 5 cm below the surface is affected by colder air temperature and the time lag between soil 5 and 10 cm depths was approximately 3 hr. This suggests that the surface soil is downwardly affected by the extremely cold air temperature. Furthermore, the changes in soil temperatures below 30 cm (not described data) are almost similar, and the soil temperature at 30 cm was upwardly decreased to the end of January, when the whole soil column was frozen as described by Romanovsky (personal communication, Geophysical Institute, University of Alaska Fairbanks). Therefore, although these data are additional experiments, these data enable us to prove the

Fig. 6. Temporal variations of CH_4 fluxes estimated by the chamber, profile and tower observations. The discontinuation of tower CH_4 observation occurred due to problems with the CH_4 analyzer since January 2005.

hypothesis on the process of CH_4 transport/emission in boreal forest soils of central Alaska during the winter.

The average wintertime emissions of CO_2 and CH_4 were 49 ± 13 gCO_2-C/m²/(winter season) and 0.11 ± 0.07 gCH_4-C/m²/(winter season) for the winter period of 212 d. This sug-

Fig. 8. Schematic drawing of the soil-originated CH_4 flow in tussock and sphagnum moss regime. The moss layer began to freeze the end of September, and the layer was completely frozen to 30 cm depth below the surface since 19 October 2005 based on the Figure 6b. The curves in the moss layer denote the freezing depth with time, and the depth is affected downward by the cold atmospheric temperature and upward by the permafrost. It suggests that the pore space in dead moss is getting narrower and then the soil air is steadily pressured with time. The stars demote the frozen soil with time. See the text for more detail.
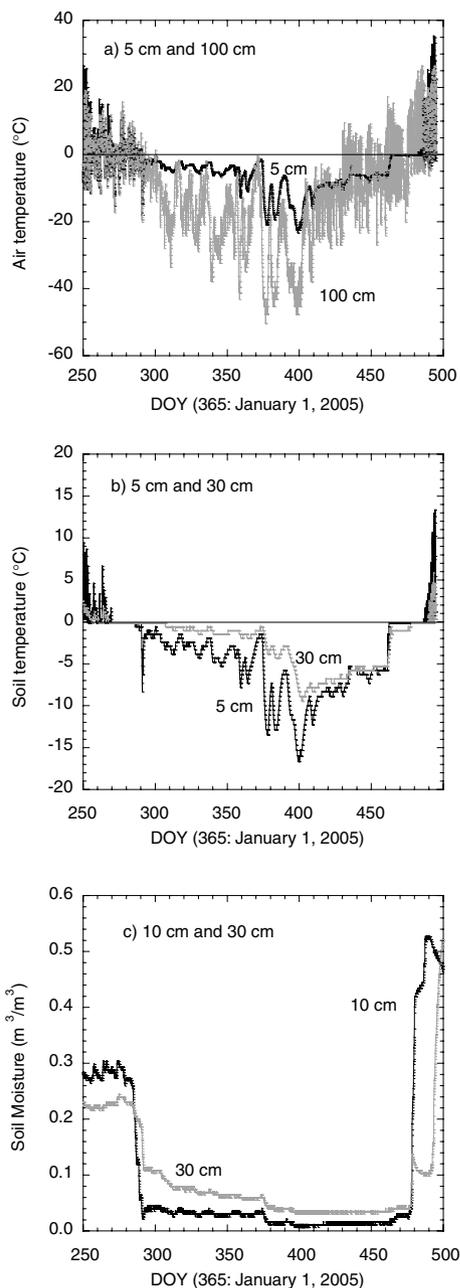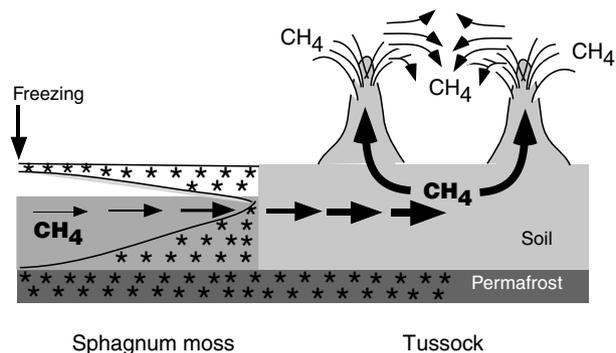

Fig. 7. Temporal variations of (a) 5 and 100 cm above sphagnum moss mat, (b) 5 and 30 cm below the moss layer for temperature and (c) 10 and 30 cm below the moss for soil moisture in near tower site from June 2004 to May 2005. Snow melting and freezing time can be analyzed with soil moisture, relating to the melting and freezing of soil surface condition. It suggests that the soil surface environment provides a clue to the production and emission of CH_4 in boreal forest soils.

gests that the winter emissions of CO_2 and CH_4 are an important part of the annual carbon budget in seasonally snow-covered terrain of the boreal forest. The winter CO_2 flux corresponds to 23% of the annual CO_2 emitted from Alaska black spruce forest soils based on the CO_2 emission of 167 ± 89 gCO_2-C/m² during the growing season. Vogel et al. (2005) reported the winter respiration of 30–54 gCO_2-C/m²/season during the winter, defined as snow-cover season, in mature black spruce forest soils of Bonanza Creek, central Alaska, these results are similar to values in this study. In alpine and subalpine soils of Wyoming, the average winter CO_2 fluxes accounted for 8–23% of the gross annual CO_2 emissions (Mast et al., 1998). In a boreal peatland in Finland, the winter fluxes corresponded to 20 to 23% of the total CO_2 fluxes (Alm et al., 1999). During the winter season in arctic tundra ecosystems of the North Slope of Alaska, the seasonal emissions from moist tussock tundra and coastal wet sedge was 70 and 20 gCO_2-C/m², which is the main share of the total annual net emission of carbon (Oechel et al., 1997). The contribution of winter CO_2 fluxes measured in this study are similar to previously reported values in alpine, boreal forest and tundra ecosystems.

There are no reports for winter CH_4 emission in snow-covered boreal forest soils. Whalen and Reeburgh (1988) suspected that downward freezing toward an impermeable permafrost horizon played a role in concentrating the dissolved subsurface methane pool and that a void in the frozen moss matrix permitted an episodic methane release observed along a pond margin with standing water in Alaska. They reported that winter CH_4 flux from the moss sites corresponded to 40% of the mean annual CH_4 flux. However, their sites are not oxidized soils, but nearly anaerobic environments due to the existence of lake water. Considering the tussock and moss distribution areas in the Northern
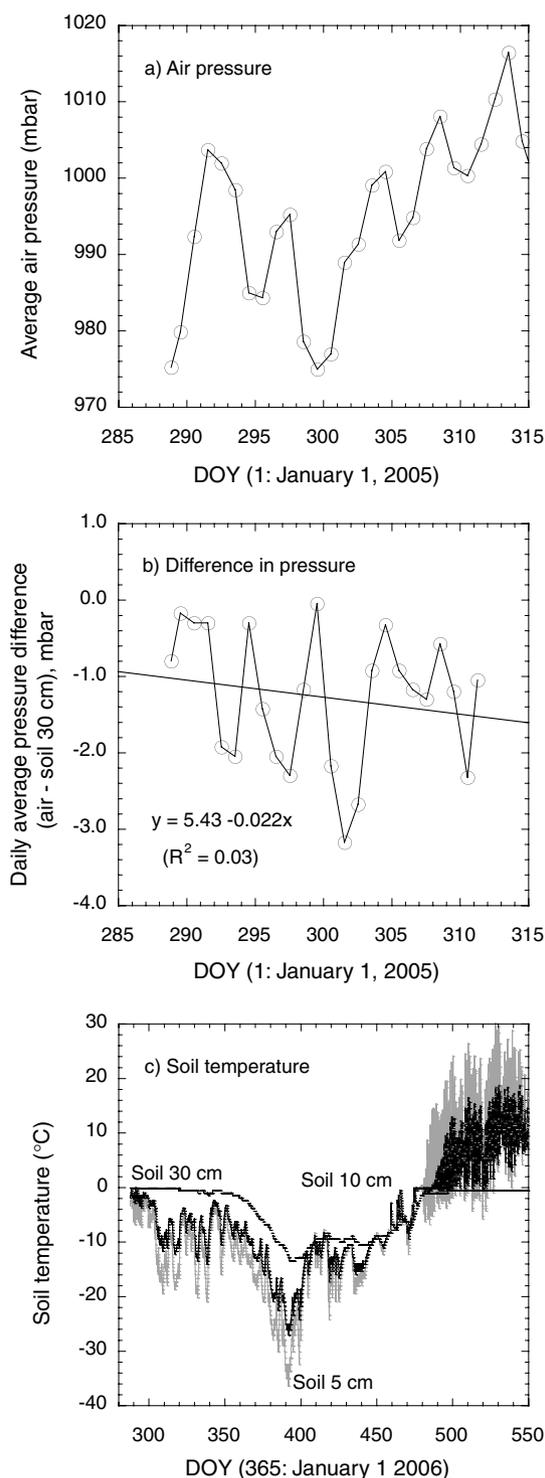
Fig. 9. Temporal variations of (a) pressure at 2 m high above the soil surface, (b) daily average pressure difference that subtracts pressure at soil 30 cm depth from that at 2 m high during October 2005 and (c) soil temperatures at 5 (grey), 10 and 30 cm depth below the surface during 2005/6 in sphagnum moss regime. These data are additionally conducted to prove the hypothesis on the transport process of methane in boreal forest soils of central Alaska during the winter.

Hemisphere (6.5×10^{12} m^2; Whalen and Reeburgh, 1998), the winter CH_4 emission measured in this study should not be overlooked in estimations of the regional/global carbon budget.

3.3. Discriminations of $^{13}CO_2$ and $^{13}CH_4$

We used a simple mixing model developed by Keeling (1958, 1961) to calculate the isotope ratio of CO_2 respired by a forest ecosystem:

$$\delta_f = \frac{[CO_2]_0}{[CO_2]_f}(\delta_0 - \delta_R) + \delta_R, \qquad (4)$$

where $[CO_2]$ is concentration of CO_2 and δ is the stable carbon isotope ratio of CO_2, and the subscripts f and o represent the atmosphere within a forest boundary layer and the atmosphere above (outside) the forest boundary layer, respectively. It can be seen from eq. (4) that a plot of $1/[CO_2]_f$ versus δ_f gives a straight line relationship with a slope, $[CO_2]_o(\delta_o - \delta_R)$, and an intercept, δ_R. We used this model to estimate δ_R, the isotope ratio of CO_2 respired by soils. Estimates were obtained from the y-intercept of a geometric mean linear regression (Flanagan et al., 1999) between δ and $1/[CO_2]$ values measured on air samples collected at different heights above the forest floor during the night when photosynthesis was not active. In this study, the same mixing model (eq. 4) was used to calculate the isotope ratio of respired CO_2 from air samples collected during the change in CO_2 in soil, snowpack and from the tower. It is possible, therefore, to use the "Keeling plot" approach to calculate separately the isotope ratio of respired CO_2 at different spatial scales during the winter.

According to the Keeling plot for the relationship between CO_2 concentrations and $\delta^{13}C$ of CO_2, despite different air samples as shown in Figure 10a, the intercept was $-22.5‰$, which corresponds to the value composed by soil respiration (i.e. the sum of root respiration and microbial respiration in soil) in a black spruce forest in central Alaska. This value is similar to respired $\delta^{13}CO_2$ ($-23.56 \pm 1.50‰$) found in a moss regime in a black spruce forest during the BOREAS project (Flanagan et al., 1999). Also, when the ambient CO_2 level was 385 ppm, the isotopic value was $-8‰$, suggesting that the CO_2 source is isotopically of the same origin and is formed by the decomposition of soil organic matter, such as acetate. Vogel et al. (2005) demonstrated that proportional contribution of roots in mature black spruce forests varied from 81% to 85% of total soil respiration using the trench method during the winter. Because their sites are similar to our sites, the $\delta^{13}CO_2$ measured in this study may be mainly originated by root respiration rather than by heterotrophic respiration.

On the other hand, the relationship between CH_4 concentrations and $\delta^{13}C$ of CH_4 was sparse, as is shown in Figure 10b. The two groups are the heavier group ($-43‰$) and relative lighter group ($-51‰$) based on the Keeling plot on the relationship between CH_4 concentrations and $\delta^{13}C$ of CH_4. However, the CH_4 concentrations from tower air samples were not constant,
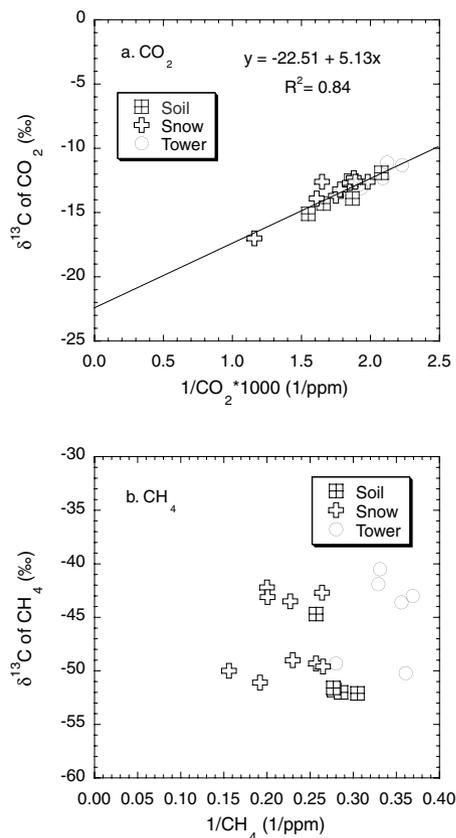
Fig. 10. Comparison of the relationships between (a) $1/CO_2$ concentration and the carbon stable isotope ratio of CO_2, (b) $1/CH_4$ concentration and the carbon stable isotope ratio of CH_4 in air samples collected soil, snowpack and tower in the boreal forest of central Alaska during the winter season of 2004/5.

ranging from 2.7 ppm ($\delta^{13}C$: $-43.0‰$) at the 8 m level (DOY 436) to 3.6 ppm ($\delta^{13}C$: $-49.3‰$) at the 8 m level (DOY 357); these are much higher than the ambient concentration of 1.8 ppm ($\delta^{13}C$: -47.2 ± 0.2 ‰) at NOAA/CMD in Barrow during the winter. This suggests that the origin of CH_4 could not be identified exactly due to relatively lower tower height and because the tower air samples might be contaminated by another air mass that has higher CH_4 concentration in this study area. The 1 d average wind speed and wind direction was monitored at the 8 m level of the tower. Generally, the wind direction during the winter was northwestern (NW). This may imply the influence ($\delta^{13}C$: $-35‰$ to $-40‰$) of the burning of fossil fuel. However, because the carbon isotopes are more enriched than the values from the tower air samples, these data could not definitely explain the origin of CH_4 emission. Moreover, Sugimoto and Wada (1993) demonstrated that $\delta^{13}C$ varied widely in methane from anaerobic environments, mainly because of changes in methanogenic pathways (acetate fermentation: $\delta^{13}C = -30‰$ to $-40‰$ and CO_2 reduction: $\delta^{13}C < -70‰$), comparing our two groups ($-43.0‰$ and $51‰$) in soil, snowpack and tower air samples. Therefore,

in order to demonstrate these results, we additionally need to (1) collect background level ambient air samples higher than 10 m by using a high volume balloon for the constant values of CH_4 concentrations and $\delta^{13}C$ of CH_4 and (2) monitor the O_2 concentration in the soil that is a significant key to understanding changes in soil environments with time during the winter.

4. Conclusions

The winter fluxes of CO_2 and CH_4 were measured in tussock and sphagnum moss sites of the boreal forest in central Alaska during the winter of 2004/5 using the chamber and the profile methods. While most of the winter CH_4 emissions were measured in anoxic environments such as fens, bogs, peatlands and wetlands, the CH_4 emission through the snowpack to the atmosphere is here first reported in boreal forest soils. Although the origin of CH_4 production was not discerned using the stable carbon isotope of methane, our findings schematically suggest how soil-originated CH_4 was transported to the atmosphere based on the CH_4 profiles in soil and snowpack, the tower observation, pressure in soil and in air during the winter of 2005/6, and soil temperature and moisture data. Because tussock areas are widely distributed in the Northern Hemisphere, the winter CH_4 emission observed in this study is of considerable significance in estimating the seasonally regional/global carbon budget along with the winter CO_2 emission that accounts for $>20\%$ of the annual soil respiration during the cold season.

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