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Contribution of winter fluxes to the annual CH_4, CO_2 and N_2O emissions from freshwater marshes in the Sanjiang Plain

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Abstract: Wetlands at the interface of the terrestrial and aquatic ecosystems are intensive sites for mineralization of organic matter, but the contribution of winter season fluxes of CH_4, CO_2 and N_2O from wetland ecosystems to annual budgets is poorly known. By using the static opaque chamber and GC techniques, fluxes of CH_4, CO_2 and N_2O at two freshwater marshes in the Sanjiang Plain were measured during the winter seasons of 2002/2003 and 2003/2004 with contrasting snow conditions and flooding regimes. The results showed that there were significant interannual and spatial differences in CH_4, CO_2 and N_2O fluxes. The *Carex lasiocarpa* marsh emitted more CH_4 and CO_2 while absorbed less N_2O than the *Deyeuxia angustifolia* marsh during the winter seasons. Over the winter season, emissions of CH_4, CO_2 and N_2O ranged from 0.42 to 2.41 gC/m^2, from 24.13 to 50.16 gC/m^2, and from -25.20 to -148.96 mgN/m^2, respectively. The contributions of winter season CH_4 and CO_2 emission to the annual budgets were 2.32%—4.62% and 22.17%—27.97%, respectively. Marshes uptake N_2O during the freezing period, while release N_2O during the thawing period. The winter uptake equaled to 13.70%—86.69% of the growing-season loss. We conclude that gas exchange between soil/snow and the atmosphere in the winter season contributed greatly to the annual budgets and cannot be ignored in a cool temperate freshwater marsh in Northeast China.

Keywords: freshwater marsh; winter flux; greenhouse gas emission; Sanjiang Plain

Introduction

Carbon dioxide, methane and nitrous oxide play important roles in the radiation balance of the earth contributing to the greenhouse effect (Rodhe, 1990). N_2O also takes part in the destruction of stratospheric ozone (Wang, 1999). Natural wetlands are globally important sources for atmospheric CH_4 and sinks for atmospheric CO_2(Tyler, 1991). In the past decades, the processes of soil freezing and thawing have been identified as important sources of trace gas emissions from soils (Panikov and Dedysh, 2000; Teepe *et al.*, 2001). Some findings show that carbon sequestration could be overestimated when the winter fluxes are not included in budget calculations (Charlotte and Nigel, 2003). It is therefore crucial to take the trace gas emissions during the cold season into account when one tries to accurately estimate the emissions of CH_4, CO_2 and N_2O from wetlands.

Wetlands are the interface between terrestrial and aquatic components of the landscape. These areas have unique characteristics due to the mixture of terrestrial and aquatic factors that influence their structure and function (Yang, 2002). Wetland ecosystems have received attention in several contexts including biodiversity, water quality maintenance and habitat protection (Lu, 2002). The area of mires in the Sanjiang Plain is about 1.04×10^4 km^2, being second only to the Qinghai-Tibet Plateau in China (Zhao, 1999). Marshes in this area are mainly divided into four types according to plant type, viz. *Carex lasiocarpa*, *Carex psudocuraica*, *Carex meyeriana* and *Deyeuxia angustifolia* (Zhao, 1999). Over the last several years, scientists have made considerable efforts on quantifying the emissions of CH_4, CO_2 and N_2O in the plant-growing seasons (Ding *et al.*, 2004; Song *et al.*, 2003), while few studies have been dedicated to the emissions during winter season and the periods of freezing and thawing. The objective of this paper is to determine the magnitude and the significance of the winter CH_4, CO_2 and N_2O fluxes to the net annual emissions from marsh in the Sanjiang Plain, Northeast China.

1 Materials and methods

1.1 Study sites

Filed observations were made in the Sanjiang Mire Wetland Experimental Station(47°35′ N, 133°31′E), Chinese Academy of Sciences, located in the eastern part of Heilongjiang province, China. More detailed information of the site was described by Ding *et al.*(2004).

From the edge to the center of the billabong, the types of vegetation vary from *D. angustifolia* to *C. lasiocarpa* as standing water depth increases. Vegetations of *C. lasiocarpa* and *D. angustifolia* were selected, which are continuously flooded and seasonal flooded marshes, respectively. Characteristics of the marsh soils in the experimental plots are listed in Table 1.

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Table 1 Characteristics of the marsh soil in the experimental plots

Vegetation	Depth, cm	Organic carbon, g/kg	Total N, g/kg	Bulk density, g/cm^3
C. lasiocarpa	0—5	321.8±19.1	14.7±2.3	0.35±0.08
	5—10	266.4±38.4	12.5±1.7	0.54±0.13
D. angustifolia	0—5	148.1±51.0	9.5±2.5	0.56±0.21
	5—10	76.3±7.2	8.2±2.5	0.79±0.17

1.2 Measurements of CH_4, N_2O and CO_2 fluxes

Three neighboring plots at each marsh were subjectively established to permanently measure CH_4, CO_2 and N_2O emission rates. Wooden boardwalks were installed for minimizing the disturbance during sampling. Three stainless steel bases(0.5 m×0.5 m× 0.2 m) with a water groove on top were installed in the soil prior to sampling and were kept throughout the experimental period. Gas samples were taken between 0900 and 1100 LST. During sampling, an open-bottom stainless steel chamber(0.5 m×0.5 m×0.5 m, equipped with two fans) was placed over the base and filled with water in the groove to ensure tightness. Air sample inside the chamber was taken for every 10 min over a 30-min period by using 60 ml plastic syringes (total of four samples). Air temperature inside the chamber and atmospheric pressure were measured during taking samples. Concentrations of CH_4, CO_2 and N_2O were simultaneously analyzed in the laboratory(within a period of at most 12 h) using a gas chromatograph (Wang and Wang, 2003). The rates of CH_4, CO_2 and N_2O emission were calculated from a linear regression of the changes in the concentration. Corrections were made for air temperature and pressure. The data deviating significantly from linearity($r^2 < 0.95$ for CH_4 and CO_2, $r^2 < 0.90$ for N_2O) were discarded.

The gas sample was taken every other day during the thawing period(April—June), twice a week during the growing season(May—October) and once or twice a month in the winter (November—March next year). Daily fluxes were estimated by a linear interpolation for the days without measurements. The total amounts of annual and winter-season CH_4, CO_2 and N_2O emissions from both sites were computed from the daily fluxes.

Data analyses were performed with SPSS 11.0 and Microsoft Excel for Windows 2000. The tested factors and the hypothesized relationship were considered statistically significant when $p<0.05$.

2 Results and discussion

2.1 Temperature and precipitation

In the Sanjiang Plain, the marshes are generally ice covered from early November to late March next year. We define the ice-covered period as winter season. The mean air temperatures in the winter seasons of 2002/2003 and 2003/2004 were -15.1 and -14.2℃, respectively. One-way ANOVA indicated that there was no significant difference in the air temperatures between the two winter seasons(p=0.82). The average air temperature in January was the lowest, -20.3℃ and -21.0℃ in 2003 and in 2004, respectively. The air temperature from April onward was above zero and the frozen soil began to thaw.

About 10% of the annual precipitation falls as snow in this region. In the winter season of 2002/2003, the precipitation was only 20.0 mm and less than the long-term average. In contrast, the precipitation in the winter season of 2003/2004 was 91.7 mm and much higher than the long-term average. The difference in precipitation between the two winter seasons was significant (p=0.03). Winter season of 2003/2004 was warmer and wetter by contrast with that of 2002/2003. During the soil thawing period, the monthly mean air temperature in 2004 was lower while total amount of monthly precipitation was higher than that in 2003.

2.2 CH_4 emission

CH_4 emission from the soil/snow to the atmosphere during the entire winter period is given in Fig.1. At the *C. lasiocarpa* site, CH_4 fluxes were slightly lower in the winter season of 2002/2003 than those in 2003/2004. Average CH_4 fluxes in these two seasons were 0.32 and 0.89 mg/(m^2·h), respectively. On the contrary, CH_4 fluxes in the winter season of 2002/2003 at the *D. angustifolia* site were slightly higher than those in the winter season of 2003/2004. Average CH_4 fluxes were 0.25 and 0.15 mg/(m^2·h), respectively. The spatial averages ±standard deviations of methane emission flux were 0.29±0.18 and 0.52 ± 0.29 mg/(m^2·h) during the two respective winter seasons in the Sanjiang Plian. These values fall among the results reported from other wetland ecosystems. Northern Minnesota bogs and fens emitted 0.19—1.29 mg(m^2·h) during this time of

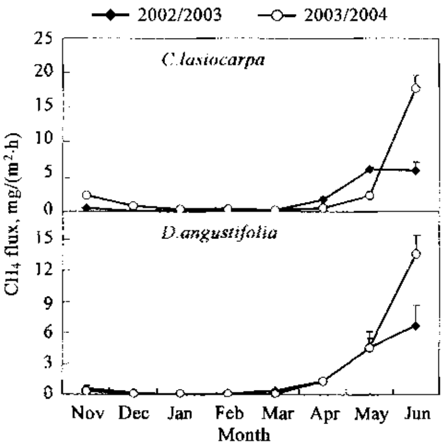

Fig.1 CH_4 flux from *C. lasiocarpa* and *D. angustifolia* marshes during freezing and thawing periods winter(Dise, 1993). In West Siberia(Tomsk), a smaller winter CH_4 flux(0.21 ±0.15 mg/(m^2·h)) was found and the average CH_4 fluxes for five winter seasons varied between 0.83 and 2.50 mg/(m^2·h) in New Hampshire (Panikov and Dedysh, 2000; Melloh and Crill, 1996).

The cumulative CH_4 emissions from the *C. lasiocarpa* site were 0.87 and 2.41 gC/m^2 in the two winter seasons, respectively, and those from the *D. angustifolia* site were 0.70 and 0.42 gC/m^2, respectively. The winter season CH_4 flux was about 2.32%—4.62% of the annual flux in both marshes, which is similar to the lower results reported from other wetland ecosystems. For example, winter seasonCH$_4$ emission was 4%—21% of annual fluxes in Minnesota, 2%—10 % in New Hampshire and 3.5%—11% in West Siberia(Dise, 1993; Panikov and Dedysh, 2000; Melloh and Crill, 1996).

CH_4 fluxes increased rapidly during the thawing period. There was a significant interannual difference in spring (through April to May) CH_4 release at the *C. lasiocarpa* site. The CH_4 released in 2004 was only about 37% of that in 2003 spring season. A study performed in boreal lakes during three consecutive winters showed that much greater snowfall in the winter reduced enormously the spring CH_4 release (Tuula *et al.*, 2004). The results of our study are consistent with their report.

2.3 CO_2 emission

Winter season CO_2 emission (Fig.2) was higher than CH_4. Significant difference in CO_2 flux(p=0.014) from the *C. lasiocarpa* site existed between the two winter seasons. The average CO_2 flux from the *C. lasiocarpa* marsh was 10.55±1.71 mg/(m^2·h) in the winter season of 2002/2003 and 44.66±5.99 mg/(m^2·h) in the 2003/2004, and 25.63 ±4.11 and 26.52 ± 3.76 mg/(m^2·h) from the *D. angustifolia* marsh in the two respective winter seasons. Note that most of the CO_2 fluxes during the winter season of 2002/2003 from the *C. lasiocarpa* marsh were below the detection limit of the gas chromatography system(Wang, 2003), we suspect that the values were erroneous and problematic, so they were not used in the determination of the annual carbon budget. The spatial averages ±standard deviations of CO_2 flux were 25.63 ±4.11 and 35.59±4.88 mg/(m^2·h) during the two winter seasons in the Sanjiang Plain. For comparison, the CO_2 flux in winter season was 22.92 mg/(m^2·h) in the Kolyma basin, Northeast Siberia (Zimov *et al.*, 1993), 45.83 and 12.22 mg/(m^2·h) from moist tussock tundra and coastal wet sedge ecosystems in the Alaskan Arctic respectively(Oechel *et al.*, 1997, 2000), and 43.2±10.8 mg/(m^2·h) in the Mirabel peatland, Quebec, Canada (Charlotte and Nigel, 2003).

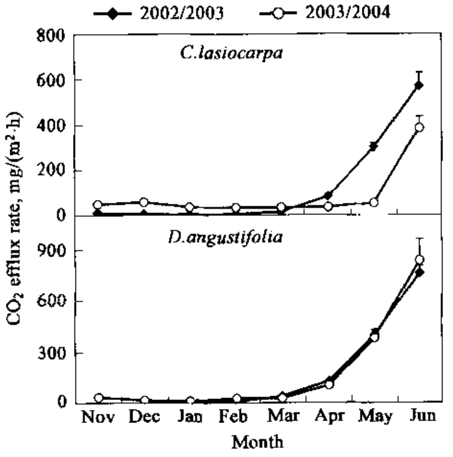

Fig.2 CO_2 efflux rate from *C. lasiocarpa* and *D. angustifolia* marshes during freezing and thawing periods

Some findings show that carbon sequestration could be overestimated if the cold season fluxes are not included in budget calculations (Charlotte and Nigel, 2003). In our study, the measured CO_2 emission using a dark chamber represents the ecosystem respiration rate, including nocturnal respiration of plant shoots and whole-day metabolism of all below-ground organisms, roots, microbes, and animals, as well as any other soil reactions degrading organic matter to CO_2. In winter, the CO_2 emission rate is equal to CO_2 flux owing to the lack of plant photosynthesis. Indeed, the net annual CO_2 flux is accounted for as a difference:

$$\text{Net C} - \text{flux} = \text{RH} - \text{NPP}$$

where RH and NPP stand for soil heterotrophic respiration and net primary production of the ecosystem, respectively. In the years of 2002—2004, we measured the CO_2 emission rates of bare soil(RH) and the plant biomass during the growing season (NPP) in the two marshes for each year (data not shown). The results of winter CO_2 flux, total annual respiration and net annual CO_2 flux are shown in Table 2.

The results demonstrated that winter CO_2 efflux comprised 2.60%—8.51% of the annual respiration and 22.17%—27.97% of the annual C budget. Current estimates of the winter CO_2 contribution to the annual C budget, based on nonconcurrent measurements of summer and winter exchange, are 21% in a Finnish mixed mire (Alm *et al.*, 1999), 25% in an Alaskan forest (Sommerfeld *et al.*, 1996) and 17% in an alpine wetland (Mast *et al.*, 1998).

C. lasiocarpa marsh emits more CH_4 and CO_2 in the winter than *D. angustifolia* marsh. Perhaps this results from the deeper upper organic layers of *C. lasiocarpa* marsh, which delays the time of soil freezing and reduces the depth of frost penetration.

Table 2 Contribution of winter to the annual CO_2 emissions and net annual C budget in the marshes, northeast China

Vegetation	Winter season	C emission, gC/m^2	Annual total respiration, gC/m^2 (winter of annual, %)	Annual net C budget, gC/m^2 (winter of annual, %)
C. lasiocarpa	2002/2003	ND	506.97	-278.26
	2003/2004	50.16	589.79 (8.51)	-226.27 (22.17)
D.angustifolia	2002/2003	24.13	745.22 (3.24)	ND
	2003/2004	26.05	1000.12 (2.60)	-93.12 (27.97)

Note: Minus indicates net uptake of CO_2; ND. no data

In general, there are two major sources of C emission during wintertime, the CO_2 and CH_4 produced *de novo* in winter by psychrophilic microorganisms(Flanagan and Bunnell, 1980), and the stored methane and carbon dioxide formed by mesophilic microbes during the warm period (Dise, 1993). In the study by Stadler(1996), 8%—20% of the soil water was not frozen although the soil temperature was -5℃ for several days. In the liquid water of frozen soils, the availability of labile C may be high as a consequence of microorganisms being killed by freezing or hygroscopic effects and organic matter in broken aggregates (Christensen and Tiedje, 1990; Christensen and Christensen, 1991). This creates a favorable condition for the activity of microorganisms and part of the trapped gas may escape through the frost-induced cracks. Panikov *et al.* (2000) studied winter CO_2 and CH_4 emissions in West Siberia and found that cold-resistant microorganisms located within the frozen peat layers were still active at -16℃ and could provide the observed instant winter CO_2 emission. In the Sanjiang Plain, according to the *in situ* observation for three years, the lowest temperatures of the marsh topsoil and subsoil(10—30 cm) were about -13℃ and -4 — -7℃, respectively. So we suggest that at least part of the C emission during the winter season in the Sanjiang Plain might be attributed to microorganism activities.

We found a significantly positive correlation between the emission rates of CH_4 and CO_2(p=0.01) during the winter season. This provides an indication that there should be common mechanisms controlling emissions of both gas species under the cold non-growing conditions. Such mechanisms may include the diffusion of gases through snow (Sommerfeld *et al.*, 1996).

Contrary to the winter period, CH_4 fluxes and CO_2 emission rates both correlated with air temperatures during the thawing period. But the coefficients in the Equations are somewhat different in different marshes (Table 3). This is perhaps due to different hydrological conditions. On the basis of experiments and observations in wetland ecosystems, many researchers have pointed out that the relationship between CH_4 flux and soil temperature can be described by the Arrhenius equation (Shangguan *et al.*,1993; Huang *et al.*, 2001) as follows:

$$F=A\cdot e^{-Ea/(R\cdot T)}$$

where F is CH_4 flux, Ea is apparent activation energy, R is the gas constant, T is the soil temperature(K), and A is an empirical coefficient. The change of CH_4 flux with temperature can also be expressed by the term of Q_{10} that denotes the factor of CH_4 flux increase with a soil temperature increase of 10℃. Generally, the Q_{10} of CH_4 flux is 2—3 (Khalil *et al.*, 1998). But during the thawing period, the very steep temperature response of microbial metabolism, which is especially high for methanogenic bacteria(the apparent values of Q_{10} are 5.79 and 9.12 in the two marshes, respectively) means that temperature not only stimulates microorganism activity, but also greatly improves the gas emission in soil. During the thawing period, the relationship between CO_2 release rates and air temperatures can also be described by the Arrhenius equation. The Q_{10} values are 5.21 and 5.44 in the two marshes respectively, while it is only 1.43 in the summer season and 3.93 in the autumn season. Obviously, the increase in air temperature stimulates more CO_2 production and emission during the thawing period. But compared to CH_4, CO_2 is less dependent upon temperature. This agrees with the results from a laboratory study that shows that spring thaw should considerably activate gas production, especially for the anaerobic formation of CH_4 (Panikov and Dedysh, 2000).

Table 3 Relationship between CH_4 fluxes, CO_2 release rates and air temperature during thawing periods

Vegetation	CH_4	CO_2
C. lasiocarpa	$Y = 0.26e^{0.18x}$ (p<0.001)	$Y = 10.73e^{0.17x}$ (p<0.001)
	$r^2 = 0.65$, Q_{10}=5.79	$r^2 = 0.78$, Q_{10}=5.21
D. angustifolia	$Y = 0.43e^{0.22x}$ (p<0.001)	$Y = 61.66e^{0.17x}$ (p<0.001)
	$r^2 = 0.54$, Q_{10}=9.12	$r^2 = 0.66$, Q_{10}=5.44

Notes: Y. CH_4 flux or CO_2 emission rate; x. air or soil temperature

There is a distinct difference between the CO_2 emissions from the two marshes in the freezing and thawing periods. During the freezing periods, the difference is very small (p>0.05), but during the thawing periods, CO_2 emission from the *D. angustifolia* marsh is significantly higher than that from the *C. lasiocarpa* marsh (p<0.01). This is mainly attributed to the differences of the hydrological conditions and plant growth.

2.4 N_2O emission

Higher N_2O emission was found during the soil freezing period in some studies (Papen and Butterbach, 1999; Teepe *et al.*, 2000). However, the measurements of this study indicated that the two marshes absorbed atmospheric N_2O in the two winter seasons (Fig.3). Perhaps this discrepancy in the research is due to the different ecosystems. The previous studies of winter N_2O flux have mainly focused on agriculture and forest ecosystem and the researchers have held the opinion that soil freezing can create an anaerobic environment for N_2O production (Papen *et al.*, 1999; Teepe *et al.*, 2000). The mechanism of N_2O absorption by marsh in the two winter seasons probably resulted from frozen soil absorbing atmospheric N_2O through frost-induced cracks and perhaps the anaerobic environment was too strong to reduce N_2O to N_2 by reductase enzymes. In the winter seasons of 2002/2003 and 2003/2004, the cumulative N_2O emissions were -29.49 and -25.20 mgN/m^2 from the *C. lasiocarpa* site, and -148.96 and -34.33 mgN/m^2 from the *D. angustifolia* site, while the marshes became a source of N_2O during the plant-growing season. Correspondingly, the contributions of winter uptake to the growing-season emission were 37.09%, 13.83%, 86.69% and 13.70%, respectively. The N_2O flux at the *D. angustifolia* site exhibited remarkable variability between the two winter seasons ($p<0.001$). This large interannual difference may be ascribed to the difference of precipitation. Precipitation in the winter season of 2003/2004 was significantly higher than that in the 2002/2003, so the snow cover on the soil during the former was much deeper than that during the latter. Deeper snow might baffle the absorption of atmospheric N_2O and then reduced the magnitude of the soil sink for atmospheric N_2O.

Nitrous oxide flux in the winter season also showed an evident difference between the two marshes. The amount of N_2O uptake at the *D. angustifolia* site was much higher than that at the *C. lasiocarpa* site($p=0.005$). It is probable that N_2O fluxes were related to the actual density of frost-induced cracks on the ice or frozen surface soil in the measurement plot because it were cracks that provided a route for gas entry. Some studies showed that there existed significant power correlation between the volume of crack and the daily N_2O emission flux in rice field (Huang *et al.*, 2004). The shallower upper organic layer of the *D. angustifolia* site advanced the time of soil freezing and increased the area and density of cracks.

Both marshes were transformed from sinks into sources of atmospheric N_2O as the temperature rise during the thawing periods. These periods were very important for N_2O emission and accounted for nearly half of the annual N_2O emissions, which agrees with the results reported from other ecosystems (Teepe *et al.*, 2000). Several studies indicated that the thawing period was important for N_2O production and emission because the process of soil thawing could provide amounts of labile C and N to microorganisms, and thus stimulate N_2O substantive production in the soil (Sommerfeld *et al.*, 1993). Studies in rice fields have shown that either the soil moisture or the water table was a crucial factor for N_2O emission and that a deeper water table would restrict N_2O formation and emission during the flooding period (Zheng *et al.*, 2000). Surprisingly and interestingly, the period of flooding was longer and the water table was deeper in 2004 than that in 2003, while N_2O fluxes were much higher in 2004 than that in 2003. The reason is still unknown and a further investigation is necessary.

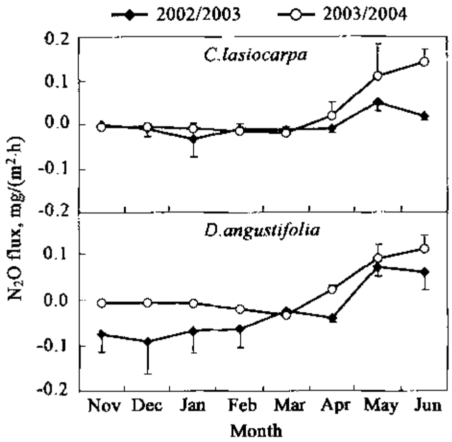

Fig.3 N_2O flux from *C. lasiocarpa* and *D. angustifolia* marshes during soil freezing and thawing periods

Considerable investigations demonstrated that process of soil thawing enhanced CH_4, CO_2 and N_2O emissions (Oeclel *et al.*, 1997; Teepe *et al.*, 2001; Tuula *et al.*, 2004). The possible explanations for this phenomenon come from several aspects, including the release of the stored CH_4, CO_2 and N_2O in the soil below the frozen soil surface(Burton and Beauchamp, 1994), the increased C and N supply to microorganisms originating from organic matter in broken aggregates or from soil microbes killed by freeze/thaw cycles (Panikov and Dedysh, 2000), the creation of an anaerobic, water saturated topsoil by melting ice and snow in the frozen subsoil(Teepe *et al.*, 2001), and the temperature activation of dormant microbial cells and plant roots(Tuula *et al.*, 2004).

3 Conclusions

The results of this study indicated that the gas exchange between soil/snow and the atmosphere in the winter season contributed greatly to the annual budgets and cannot be ignored in a cool temperate freshwater marsh in northeast China. Marshes uptake N_2O during the freezing period, while release N_2O during the thawing period. Significant temporal and spatial differences in the CH_4, CO_2 and N_2O fluxes existed in the winter season.

The mechanisms and processes of the production and emission of the trace gases during the winter season are complex in the freshwater marshes. To accurately quantify the contribution of winter season emissions of CH_4, CO_2 and N_2O to the annual budgets, it is crucial to pay more attention to the freezing and thawing actions.

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