



Concentrations, sources, and export of dissolved CH₄ and CO₂ in rivers of the permafrost wetlands, northeast China



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ABSTRACT

This study, conducted at the Emuer River in northern China during the growing season of 2014, investigated the concentrations and sources of dissolved CH₄ and CO₂ in wetland rivers in the permafrost region, as well as the total export of dissolved gaseous carbon (C) transported by streamflow. CH₄ and CO₂ concentrations in these rivers, ranging from 0.410–1.045 to 107.838–190.925 μmol L⁻¹, respectively, were among the highest levels of gaseous C concentrations reported from studies around the world. The concentrations of the C gases varied greatly with streamflow, but the influence of hydrological, as well as meteorological parameters, was not significant, except for the influence of the thaw depth of the soil active layer of adjacent wetlands on CH₄ concentrations. δ¹⁸O‰ values suggest the observed dissolved gases originated mainly from soil pore water in the active layers of the wetlands, as opposed to organic-rich sediments in river beds, which serves as a common source for riverine CH₄ and CO₂ according to many other studies. A total of 444.30 tC was exported by the 3rd order main Emuer river during the growing season, and an export coefficient of 4.65 mg C m⁻² day⁻¹ was calculated for the wetland ecosystem. The dataset in this study is significant with regard to the global carbon balance because it is representative of the extensive wetlands occurring throughout the Eurasian permafrost.

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1. Introduction

Greenhouse gases such as carbon dioxide (CO₂) and methane (CH₄) originating from rivers have been recognized as important components of the global carbon (C) cycle (Richey et al., 2002; Battin et al., 2009; Bastviken et al., 2011; Raymond et al., 2013). Recent estimates show that inland waters including rivers and lakes discharge around 2.1 Pg C yr⁻¹ as CO₂ (Raymond et al., 2013) and 0.65 Pg C yr⁻¹ as CH₄ (Bastviken et al., 2011). These estimates are relatively uncertain, as the data needed for gas export calculations are sparse with high spatial and temporal variability. Therefore, detailed data regarding river-exported greenhouse gases from globally representative regions are needed to provide accurate estimates of the global carbon balance.

Wetlands ecosystems can affect greenhouse gas exchange between terrestrial habitats and the atmosphere. Wetlands are the dominant natural sources of global atmospheric CH₄

(Walter et al., 2001; Dlugokencky et al., 2011). According to global estimations, there is a potentially “missing” wetland CH₄ source of 87 Tg C yr⁻¹ (Walter et al., 2001). Recent studies indicate that there is still large uncertainty in the global CH₄ budget due to the potential overestimates from bottom-up approaches (Kirschke et al., 2013). One reasonable explanation for the “missing” CH₄ source is that additional CH₄ sources such as freshwater ecosystems, and source pathways such as river transport and emissions, are not well accounted for (Crawford et al., 2014). Similarly, the C fluxes in form of CO₂ transported by land rivers are often neglected in the global estimations, which will lead to an underestimation of land carbon emissions, especially in the boreal wetlands where the hydrological processes have been greatly changed due to climate warming (Vetrov and Romankevich, 2004; Striegl et al., 2012).

Wetlands, usually drained by streams and rivers, could play a key role in greenhouse gas budgets as they connect huge carbon pools that are sequestered in wetland soil or peat (Cao et al., 1998; Hopkinson et al., 2012). One study has indicated that drainage rivers could reach a very high saturation level of CH₄ of up to 11,560% (Wang et al., 2009) and a level of CO₂ of up to 126.5 μmol L⁻¹ higher than that in the atmosphere (Striegl et al., 2012). These dissolved C

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gases are not only locally significant to greenhouse gas budgets of emissions to the atmosphere in situ, but they may also affect atmospheric concentrations over the whole watershed scale because of continued emissions when transported downstream by streamflow. Therefore, to fully evaluate the influence of greenhouse gases from wetlands on the atmosphere, it is necessary to understand the lateral export of dissolved CH_4 and CO_2 .

Recent warming of the boreal permafrost area has resulted in substantial research regarding the lateral exports of dissolved organic and inorganic carbon from permafrost to the northern seas and Arctic Ocean (Vetrov and Romankevich, 2004; Prokushkin et al., 2009). However, gaseous C exports by rivers remain poorly quantified. From an ecosystem carbon balance perspective, whole riverine accounting of particulate, dissolved, and gaseous carbon exports is almost entirely lacking in the literature. Therefore, linkages between wetland sources and sinks of C, including net ecosystem exchange, remain poorly quantified. The objectives of this study were to: (1) characterize the seasonal and spatial variation of dissolved CH_4 and CO_2 concentrations in a boreal permafrost river and wetland landscape; (2) identify sources of the dissolved gases; and (3) estimate the downstream export of dissolved gaseous C.

2. Approach and methodology

2.1. Study area

The northern parts of the Great Xing' an Mountains in China form the southern margin of the continuous permafrost zone in Eurasia. Most of the cold temperate wetlands in China are located within this region, which, at present, contains $82.45 \times 10^4 \text{ km}^2$ of natural wetlands (Liu, 2005; Zhou et al., 2005). The Emuer River is a main tributary of the Amur River, which is a boundary river between China and Russia (Fig. 1). The annual average temperature in the region is approximately -3°C , and the average annual precipitation amounts to 400 mm. Wetlands are distributed extensively across the flat river valleys through which the rivers meander. The wetland are covered with peat soil with an organic carbon content in the range of 30–60% in the top 30 cm. The growing season lasts from May until early October, and the dominant plants are *Ledum palustre*, *Vaccinium uliginosum*, *Sphagnum* spp., and *Larix gmelini* Rupr. The maximum thaw depth of the active layer in the wetlands is about 50–60 cm, and generally is reached by late August. The mountains located on both sides of the valley are covered by newly planted *Pinus sylvestris* var. *mongolica*. Continuous logging and frequent fires during the past 60 years have caused extensive damage to the original coniferous forest, thereby resulting in very low accumulations of organic matter at the surface.

2.2. Water sampling and analysis

Between the 5th of May and the 5th of October 2014, water samples were collected in three sections of the watershed. These sections consisted of a first-order tributary (G1), a second-order tributary (G2), and the main course (G3) of the Emuer River (Fig. 1). The watershed areas controlled by sections G1, G2, and G3 (WG1, WG2 and WG3) are 30, 287, and 2590 km^2 in size, respectively, and each of these areas contains wetlands that correspond to 10.7%, 32.2% and 24.3% of the respective areas. WG1 is located mainly in a mountainous area with an average slope of 1/80, and the associated wetlands are distributed in the lowest part near the gauging profile G1. Therefore, the streamflow of G1 is mostly from newly planted mountain forest, and goes into the wetlands in WG2. As no direct hydrological connection with the mountain streamflows, the streamflows of G2 and G3 result mainly from the wetlands in the flat valleys (Fig. 1). Water samples were collected at the three sections

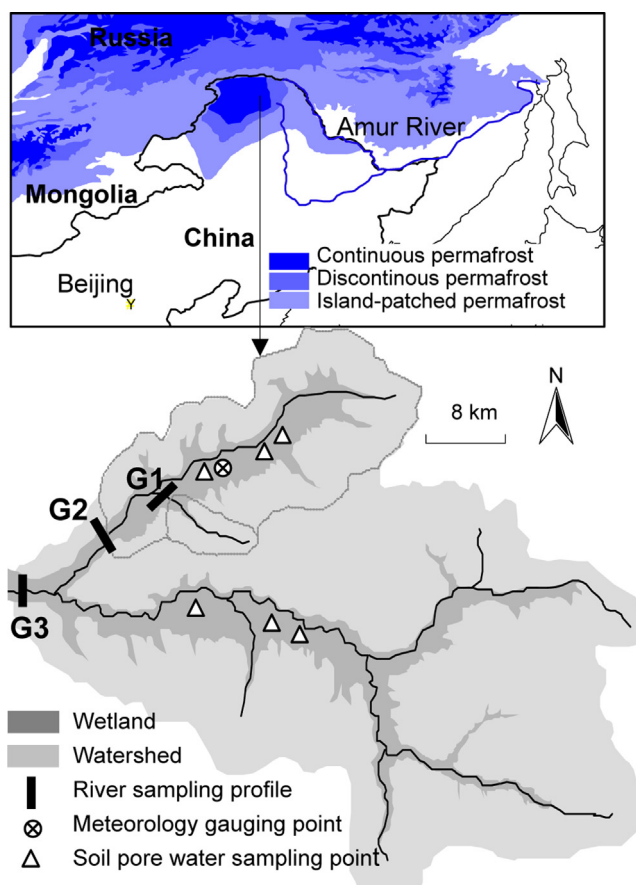


Fig. 1. Diagram of the geographic location of the study area and the sampling points.

every 3–7 days at noon, with a higher sampling frequency during flood periods and lower frequency during low water periods. Three duplicates samples were collected during each sampling event and values averaged. In G1, the last sample was collected on the 26th of July after when the streamflow ended.

During sampling, 30 ml of water at 20–50 cm depth from the center of the river was suctioned into a 100 ml glass bottle filled with high purity N_2 . Then, the bottle was oscillated vigorously on a shaking table for 5 min at 20°C and 5 ml of air from the headspace was collected with a 20 ml syringe. The concentrations of carbon dioxide (CO_2) and methane (CH_4) in the air samples were determined on a gas chromatograph (Agilent 7890A, USA). Routine calibration was done with mixed primary standard gases (2.06 ppmv CH_4 , 421.5 ppmv CO_2 ; certified accuracy $\pm 2\%$). The concentration and saturation level of the gases in the water samples can be calculated by the following equations:

$$C_L = C_G(\beta RT/22.4 + V_G/V_L) \quad (1)$$

$$S = (C_L/C_{eq}) \times 100 \quad (2)$$

where C_L and C_G ($\mu\text{mol L}^{-1}$) are the concentration of the gas in the water and headspace air, respectively; β ($\text{L L}^{-1} \text{atm}^{-1}$) is the Bunsen solubility; R ($\text{L atm mol}^{-1} \text{K}^{-1}$) is the gas constant; T (K) is the air temperature after balance; V_G and V_L (L) are the volume of the headspace air and water sample in the bottle, respectively; S (%) is the percent saturation; and C_{eq} ($\mu\text{mol L}^{-1}$) is the equilibrium concentration that is calculated by using Henry's first law.

To identify the source of river water dissolved CO_2 and CH_4 , the depletion of stable oxygen isotopes ($\delta^{18}\text{O}_\circ$) was evaluated for water from the three rivers, for wetland soil pore water and for local rainfall. The samples were analyzed with an isotope mass spectrometer (Finnigan Delta plus XP, USA) at the Key Laboratory of

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