

Dissolved N₂O and CH₄ in seepage and stream water in Yakutsk

Takuji Sawamoto^{1,*}, Fumiaki Takakai², Alexey R. Desyatkin^{2,3}, Roman V. Desyatkin³, and Ryusuke Hatano^{2,4}

¹ *Rakuno Gakuen University, Ebetsu, Hokkaido, 069-8501, JAPAN*

² *Graduate School of Agriculture, Hokkaido University, Sapporo, Hokkaido, 060-8589, Japan*

³ *Institute for Biological Problems of Cryolithozone, Siberian Branch, Russian Academy of Science, 41 Lenin Avenue, Yakutsk 677891, Republic of Sakha, Russia*

⁴ *Field Science Center for Northern Biosphere, Hokkaido University, Sapporo, Hokkaido, 060-8589, Japan*

*Corresponding author: sawataku@rakuno.ac.jp

1. INTRODUCTION

From a viewpoint of nutrient cycling, carbon budget, and greenhouse gases (GHGs) emission, degassing (or outgassing/evasion) of carbon dioxide (CO₂), methane (CH₄), and nitrous oxide (N₂O) from surface water to the atmosphere have been investigated.

Kling et al. (1991) measured the partial pressure of CO₂ in aquatic ecosystems (lakes and streams) across arctic Alaska. In most cases CO₂ was released to the atmosphere. They suggested that the arctic terrestrial sink for atmospheric CO₂ would be overestimated without taking CO₂ degassing from aquatic ecosystems into account.

Hope et al. (2001) investigated the dissolved CO₂ and CH₄ concentrations in a first-order stream located in northeast Scotland. Both CO₂ and CH₄ concentration showed super-saturation and decreased in the downstream site. They estimated the degassing of CO₂ and CH₄ from the stream surface and exports to downstream site. The degassing (dominantly in the form of CO₂) was estimated to represent at least 34% of the total stream-mediated exports of carbon. Their results show the degassing of CO₂ and CH₄ are significant in terms of carbon budget in watershed scale as well as GHGs emission.

Richey et al. (2002) demonstrated that the degassing of CO₂ from rivers and wetlands of the central Amazon basin constitutes an important carbon loss process, equal to $1.2 \pm 0.3 \text{ Mg C ha}^{-1} \text{ y}^{-1}$. They claimed CO₂ degassing from aquatic ecosystems is one of essential process in the global carbon cycle as well as in the terrestrial ecosystems in the humid tropics.

Sawamoto et al. (2002) measured the dissolved N₂O, CH₄, and CO₂ concentration in pipe drainages, seepages, and stream waters on a livestock farm located in a watershed in north Japan. All gaseous concentrations in pipe drainages and seepages were super saturation, indicating the degassing of all gases occurred. However, in contrast to the results of Hope et al. (2001), the dissolved CH₄ concentration in the stream increased in the downstream site, suggesting strong CH₄ influx and degassing occurred in the lower part of the farm.

Sawamoto et al. (2005) reviewed available data of N₂O and nitrate (NO₃) concentration in leached waters that were considered to be affected by human activity (mainly agriculture). They found a rough relationship between them and proposed a new emission factor (N₂O-N /NO₃-N) of 0.0024.

These previous studies show that observation of the behavior of GHGs in surface waters is essential for understanding nutrient cycling, carbon budget, and greenhouse gases emission. In Yakutsk region (far eastern Russia), however, such observation have not been investigated. Objectives of this study are to obtain preliminary data of dissolved CH₄ and N₂O concentration and water quality, and to compare with previous studies.

2. MATERIALS AND METHODS

2.1 Site description and Sampling

The field sampling was done twice in May and August 2005. All sampling sites located eastern side of River Lena (Fig. 1). Twelve stream waters were sampled along upper to down stream which was a tributary river of River Lena. The direct distance between S1 and S8 was about 90 km, and that between S8 and the floodplain of River Lena was about 1 km. Two seepages were also sampled. The direct distance between B1&B2 and the floodplain of River Lena was about 1 km.

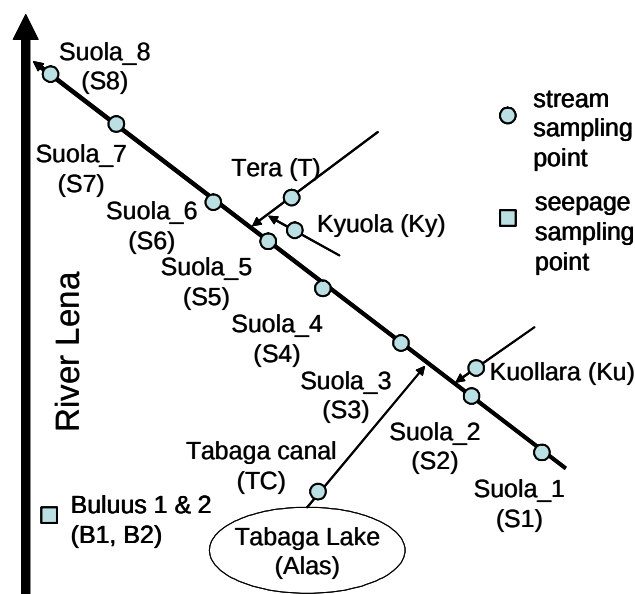


Fig. 1. The schematic diagram of stream flow and sampling sites (N61°92', E130°57' at S4). The arrow indicates flow direction.

2.2 Water quality

Water temperature was measured in situ. After the sampling, water samples were filtered through 0.2-μm membrane filters within a day. Total dissolved N (TDN) was determined by the method of alkaline persulfate digestion and HCl-acidified UV detection using UV-VIS spectrophotometer (UV mini 1240; Shimadzu, Kyoto, Japan). Nitrate (NO₃-N) was determined by ion chromatography (QIC Analyzer; Dionex Japan, Osaka, Japan). Ammonium (NH₄-N) was determined by the indophenol blue method using UV-VIS spectrophotometer (UV mini 1240; Shimadzu, Kyoto, Japan). DON was calculated by subtracting the concentration of inorganic N (NO₃-N, NH₄-N) from TDN. Total dissolved C (TDC) and dissolved inorganic C (DIC) were determined by TOC analyzer (TOC-5000A; Shimadzu, Kyoto, Japan) and dissolved organic C (DOC) was calculated by subtracting DIC from TDC.

2.3 Dissolved CH₄ and N₂O concentration

Head space method with single equilibration was used for determination of the dissolved CH₄ and N₂O concentration. A 30-mL vial bottle with butyl rubber stoppers was filled with N₂ gas (free of CH₄ and N₂O) in the laboratory, in which 15-mL of sample water was injected in situ. Immediately, the vial was shaken vigorously for 3-min and a 15-mL of headspace sample was transferred into another 10-mL vacuumed vial bottle with butyl rubber stoppers. Three replications were applied, and blank test was done. The CH₄ and N₂O concentrations were determined by gas chromatograph (GC-14B; Shimadzu, Kyoto, Japan) equipped with FID and

ECD, respectively. Bunsen absorption coefficient and Ostwald solubility coefficient (The Chemical Society of Japan, 1984) were used for the calculation.

2.4 Statistical analysis

Paired *t*-test for seasonal difference was performed using SigmaStat (SPSS 1997).

3. RESULTS AND DISCUSSION

3.1 The water quality and the dissolved CH₄ and N₂O concentration

Fig. 2 shows the water temperature, pH, and EC in the seepages and the stream waters. Significant seasonal change was observed in temperature and EC (higher in August). The temperature in seepage was about 1 °C in both May and August. Temperature in stream was about 10 and 15 °C in May and August, respectively. pH values were around 8.

Fig. 3 shows the dissolved CH₄, DOC, and DIC concentrations in the seepages and the stream waters. The dissolved CH₄ concentration ranged from 0.097 to 23 µg CH₄-C L⁻¹, which is super saturation (1.9–680 times higher than ambient concentration). This result indicates that CH₄ degassing occurred from water surface to the atmosphere. Unlike with the case of Hope et al. (2001) and Sawamoto et al. (2002), the dissolved CH₄ concentration showed almost constant, suggesting continuous influx of dissolved CH₄ into the stream water. These degassing and influx should be quantified in terms of estimate of GHGs emission. Seasonal change in the dissolved CH₄ concentration was not observed, but that in sum of DOC and DIC was observed.

Fig. 4 shows the dissolved N₂O, DON, and DIN concentrations in the seepages and the stream waters. The dissolved N₂O concentration (ranged from 0.09 to 0.29 µg N₂O-N L⁻¹) was ambient level or lower than ambient level. This result indicates N₂O emission did not occur from water surface to the atmosphere. The dominant form of dissolved N was organic form. The average value (and the range) of DON / [DON + DIN] was 0.99 (0.88–1.00) for stream and 0.69 (0.62–0.75) for seepage. Both NO₃-N concentration (<0.02 and 0.06–0.11 mg N L⁻¹ for stream and seepage, respectively) and NH₄-N concentration (<0.31 and <0.00 mg N L⁻¹ for stream and seepage, respectively) were very low.

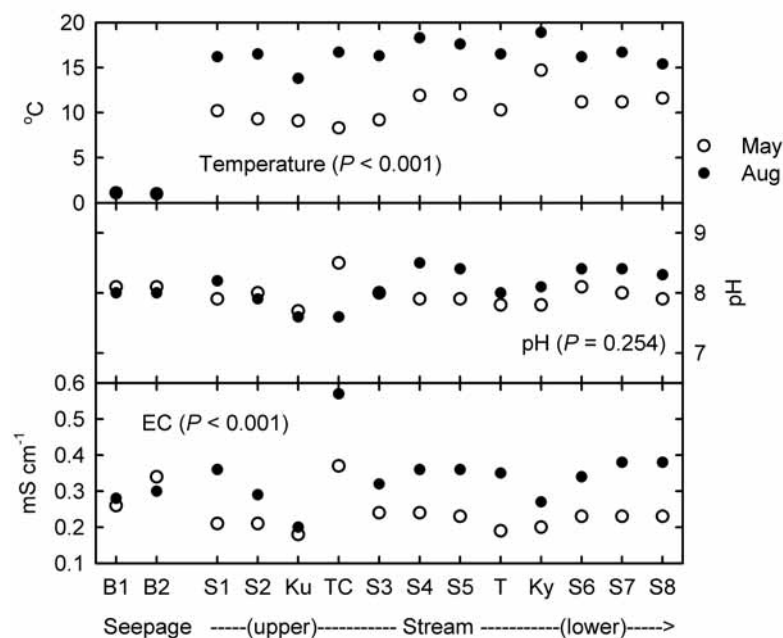


Fig. 2. Water temperature, pH, and EC in the seepages and the stream waters. *P* values show the results of paired *t*-test for seasonal difference.

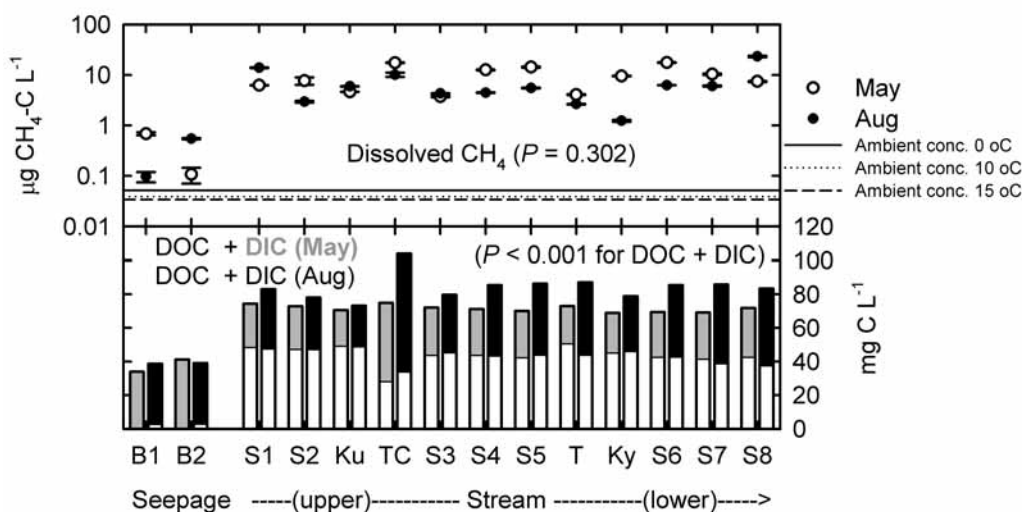


Fig. 3. Dissolved CH_4 , DOC, and DIC concentrations in the seepages and the stream waters. P values show the results of paired t -test for seasonal difference. Error bars denote standard deviation of three analytical replications.

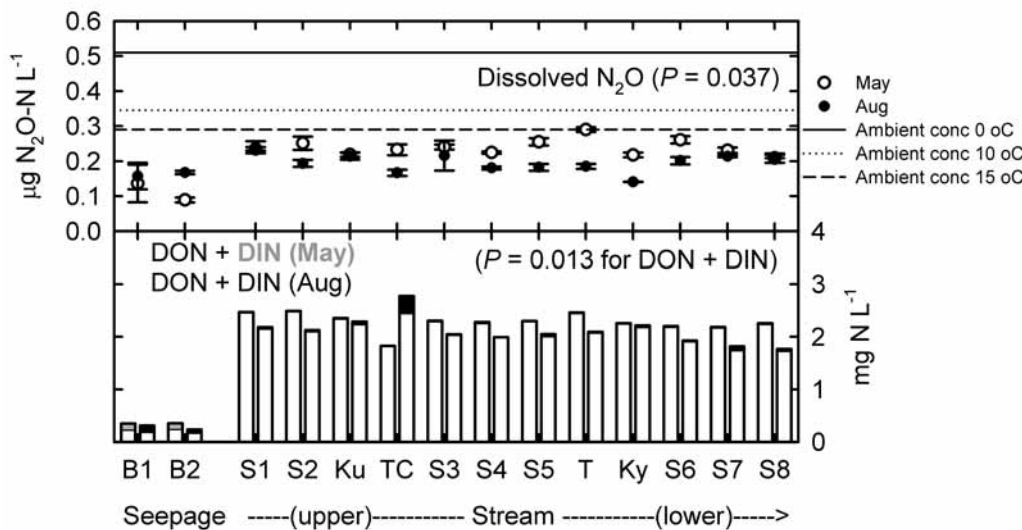


Fig. 4. Dissolved N_2O , DON, and DIN concentrations in the seepages and the stream waters. P values show the results of paired t -test for seasonal difference. Error bars denote standard deviation of three analytical replications.

3.2 Comparison with previous studies

Table 1 shows a comparison of the dissolved CH_4 concentration with previous studies. The concentration in the stream in this study was the same level as that in the previous studies. The concentration in the seepage in this study was lower than that in the results of Sawamoto et al. (2002). The seepage water did not flow into the stream in this study (Fig. 1). It may be possible that seepage and/or base flow, containing dissolved CH_4 as high concentration as the results of Sawamoto et al. (2002), flowed into this stream in Yakutsk.

Table 1. Comparison of the dissolved CH₄ concentration (range, µg CH₄-C L⁻¹).

	Hope et al. (2001) A first-order stream located in northeast Scotland	Sawamoto et al. (2002) A livestock farm located in a watershed in north Japan	This study Yakutsk region
stream	0.6 – 23	0.0 – 17	1.2 – 23
seepage	no data	0.0 – 3400	0.1 – 0.7

Perakis and Hedin (2002) investigated the nitrogen loss from stream water in unpolluted south American forests and in north America where nitrogen cycling have been affected and altered by human activities. The DON concentration range was the same (8–135 µg N L⁻¹) in both regions. But they found that the major nitrogen compound was quite different. In north America, NO₃-N was major nitrogen compound (NO₃-N / [DON + DIN] = 0.70–0.86). However, in unpolluted south American forests, DON was major nitrogen compound (DON / [DON + DIN] = avg. 0.80, range 0.61–0.97). They concluded that this nitrogen composition in stream waters is one of the important characteristic in unpolluted ecosystems. Our study also similar nitrogen composition (DON / [DON + DIN] was 0.99 (0.88–1.00) for stream), which may suggest indirect evidence of un-pollution in Yakutsk region. DON concentration in this study (range 1.7–2.5 mg N L⁻¹, Fig. 4) was remarkably higher than the result of Perakis and Hedin (2002), which are discussed by Nakahara et al. (in present volume).

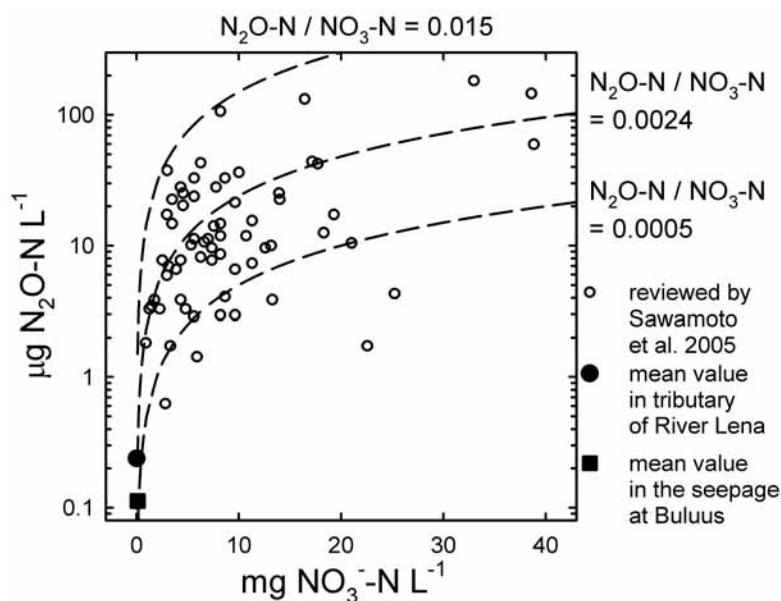


Fig. 5. Relationship between NO₃ and N₂O concentrations. Reviewed values by Sawamoto et al. (2005) were obtained from seepages, drain ditch waters, drain pipe waters, and well waters that were obtained in Japan, UK, USA, and Israel.

Fig. 5 shows the relationship between NO₃-N and N₂O concentrations. High NO₃-N concentration and super saturated N₂O concentration were observed in the data reviewed by Sawamoto et al. (2005), which probably due to anthropogenic impact, mainly agricultural activity. As comparison to these values, both NO₃-N and N₂O concentrations are very low level in Yakutsk region. This may be resulted from N cycle, including de-nitrification, and also from un-pollution in this region discussed in the previous paragraph.

4. CONCLUSION

- (1) Dissolved CH₄ concentration was super-saturation, indicating the degassing of CH₄ does occur from water surface to the atmosphere. It should be quantified in terms of estimate of GHGs emission.
- (2) Dissolved N₂O concentration was ambient level or lower than ambient level, indicating N₂O emission does not occur from water surface to the atmosphere.
- (3) NO₃ concentration was very low and DON was the major N compound, which resemble the composition in unpolluted south American forests. NO₃ and N₂O concentrations were very low as compared to reviewed values. These facts would be resulted from the N cycle (incl. de-nitrification process) and un-pollution in this region.

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