

Methane transport from the active layer to lakes in the Arctic using Toolik Lake, Alaska, as a case study

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Methane emissions in the Arctic are important, and may be contributing to global warming. While methane emission rates from Arctic lakes are well documented, methods are needed to quantify the relative contribution of active layer groundwater to the overall lake methane budget. Here we report measurements of natural tracers of soil/groundwater, radon, and radium, along with methane concentration in Toolik Lake, Alaska, to evaluate the role active layer water plays as an exogenous source for lake methane. Average concentrations of methane, radium, and radon were all elevated in the active layer compared with lake water (1.6×10^4 nM, 61.6 dpm·m⁻³, and 4.5×10^5 dpm·m⁻³ compared with 1.3×10^2 nM, 5.7 dpm·m⁻³, and 4.4×10^3 dpm·m⁻³, respectively). Methane transport from the active layer to Toolik Lake based on the geochemical tracer radon (up to 2.9 g·m⁻²·y⁻¹) can account for a large fraction of methane emissions from this lake. Strong but spatially and temporally variable correlations between radon activity and methane concentrations ($r^2 > 0.69$) in lake water suggest that the parameters that control methane discharge from the active layer also vary. Warming in the Arctic may expand the active layer and increase the discharge, thereby increasing the methane flux to lakes and from lakes to the atmosphere, exacerbating global warming. More work is needed to quantify and elucidate the processes that control methane fluxes from the active layer to predict how this flux might change in the future and to evaluate the regional and global contribution of active layer water associated methane inputs.

methane | Arctic lakes | active layer | radioisotope tracers | permafrost

Methane is a powerful greenhouse gas estimated to be responsible for approximately one-fifth of man-made global warming, and its concentration has been increasing (1). The largest natural source of methane is wetlands (2), including a major component from northern high-latitude regions containing permafrost (3). Observations in the Siberian Arctic show high rates of methane release from both the coastal seabed and land-based permafrost soils (4, 5). Previous research has also documented extensive methane release from Arctic lakes, primarily via ebullition, with significant spatial variability within and between lakes (6, 7). Methane in Arctic lakes forms by microbial production (methanogenesis) in the water column and/or within anaerobic lake sediments (8). However, it is possible that some fraction of the observed methane is not produced within the lakes but is rather transported to the lakes from external, land-based sources through subterranean groundwater discharge (SGD) (9) (Fig. 1). Specifically, water in the active layer (the surficial layer of the soil system above the permafrost that thaws every summer) could be transported into lakes and rivers during the thaw season (May–August) (10). Groundwater in some temperate climate regions can be highly enriched in methane (11), and groundwater discharge contributions to the methane budget of the coastal ocean and lakes in temperate climate have been reported (12, 13). This conduit of methane transport may be particularly important in regions where organic-rich soils and anaerobic conditions promote methane production in the soil,

such as in the Arctic (14). SGD occurs everywhere at the land–water interface in lakes, rivers, and the ocean, and evidence of water flow through the active layer in permafrost-dominated regions is found in springs, in open water reaches and base flow of rivers, in talik, and in the distribution of halophytic vegetation (15, 16). Methane fluxes sourced from land into lakes and coastal waters in the Arctic have not been directly measured. The organic-rich soils and anaerobic conditions in the active layer in the Arctic are prime conditions for methane production, and this methane could be transported to lakes and coastal waters.

Results and Discussion

Toolik Lake is an Arctic lake (1.5 km²) situated in the continuous permafrost zone in the northern foothills of the Brooks Range, Alaska (68°38'N, 149°36'W; elevation 720 m). The region is affected by rapidly growing thermokarst features, a testimony to decaying near-surface permafrost due to recent climate changes (17). There are more than 2 million Arctic lakes, which cover a large fraction of the surface area north of 45°N (18, 19), and methane input from the active layer into these lakes could be important. Methane, radon, and radium concentrations were determined concurrently in lake water, river water entering the lake, and groundwater, in July 2011 and August 2012; limited sampling in a few other lakes was also conducted in the summer of 2014. Naturally occurring ²²⁴Ra ($t_{1/2} = 3.6$ d) and ²²²Rn ($t_{1/2} = 3.8$ d) isotopes are well recognized as valuable geochemical tracers of water that has been in contact with rocks and soil (20, 21) and have been used to quantify groundwater discharge

Significance

Methane, a greenhouse gas, contributes to global warming. We show that methane-rich water from the seasonally thawed active layer in the Arctic flows into Toolik Lake, Alaska. This may be an important previously unrecognized conduit for methane transport and emissions in Arctic lakes. The controls on methane input from the active layer are fundamentally different than those affecting methane production within lakes, and the response of these processes to climate and environmental change is also distinct. The accuracy of predictions of methane emissions and ultimately the extent of climate change that can be expected in the Arctic depend on a better understanding of methane dynamics in the region, including the controls over methane production and transport processes within the active layer.

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Table 1. The ^{224}Ra , ^{222}Rn , and methane average concentrations and range of concentrations measured in the active layer, lake water, and rivers at Toolik Lake

Tracer analyzed	Active layer water	Lake water	Primary inflow	Secondary inflow
		2011		
^{224}Ra , dpm·m ⁻³	58.5 12.3–161.7 <i>n</i> = 7	3.3 1.3–10.9 <i>n</i> = 11	10.1 <i>n</i> = 1	10.4 <i>n</i> = 1
^{222}Rn , dpm·m ⁻³	4.1×10^5 <i>n</i> = 2	5.7×10^3 $0.6\text{--}19.7 \times 10^3$ <i>n</i> = 291	not sampled not sampled not sampled	not sampled not sampled not sampled
[CH ₄], nM	3.5×10^4 $6.3 \times 10^2\text{--}1.5 \times 10^5$ <i>n</i> = 6	1.3×10^2 $4.7 \times 10^1\text{--}2.7 \times 10^2$ <i>n</i> = 34	not sampled not sampled	4.3×10^2 <i>n</i> = 1
		2012		
^{224}Ra , dpm·m ⁻³	63.2 7.2–186.2 <i>n</i> = 13	6.9 2.2–14.9 <i>n</i> = 18	6.4 5.4–7.4 <i>n</i> = 2	0.8 <i>n</i> = 1
^{222}Rn , dpm·m ⁻³	4.9×10^5 $4.8\text{--}5.1 \times 10^5$ <i>n</i> = 4	2.9×10^3 $0.3\text{--}7.3 \times 10^3$ <i>n</i> = 388*	17.9×10^3 $9.0\text{--}22.4 \times 10^3$ <i>n</i> = 125*	13.9×10^3 $7.5\text{--}17.6 \times 10^3$ <i>n</i> = 29
[CH ₄], nM	8.1×10^3 $1.2 \times 10^1\text{--}4.6 \times 10^4$ <i>n</i> = 16	1.4×10^2 $2.2 \times 10^1\text{--}8.2 \times 10^2$ <i>n</i> = 50	1.1×10^2 $9.5 \times 10^1\text{--}1.2 \times 10^2$ <i>n</i> = 3	5.3×10^1 <i>n</i> = 1

*Rn values include measurements from both survey and time series records.

throughout the world (22). Methane concentrations, as well as ^{224}Ra and ^{222}Rn activities, in water collected from the active layer are at least an order of magnitude higher than those observed in Toolik Lake water (Table 1 and Tables S1 and S2). Measurements of ^{224}Ra and ^{222}Rn in Toolik Lake indicate that water high in Ra and Rn (from the active layer) is discharging into the lake. This is consistent with a water balance estimate that noted an unidentified flow of water to the lake contributing $3.64 \times 10^4 \text{ m}^3 \cdot \text{d}^{-1}$ (23). Water input calculations using Rn mass balance ($2.1 \pm 1.3 \times 10^4 \text{ m}^3 \cdot \text{d}^{-1}$ and $1.6 \pm 0.6 \times 10^4 \text{ m}^3 \cdot \text{d}^{-1}$, in 2011 and 2012, respectively) suggest similar input rates, and Ra modeling also produces results of similar magnitude (Table 2 and SI Text). Estimates of methane fluxes to Toolik Lake determined by multiplying the average active layer methane concentrations ($3.5 \times 10^4 \text{ nM}$ and $8.1 \times 10^3 \text{ nM}$ in 2011 and 2012, respectively) by the Rn based water discharge rate estimates range from 2.9 to $0.5 \text{ g} \cdot \text{m}^{-2} \cdot \text{y}^{-1}$ in 2011 and 2012 respectively (Table 2 and SI Text). The range in flux reflects differences in average methane concentrations in the active layer as well as differences in water discharge from the active layer between the years. These rates are comparable in magnitude to reported methane emissions from Toolik Lake to the atmosphere [$\sim 2 \text{ g} \cdot \text{m}^{-2} \cdot \text{y}^{-1}$ (24)] (Table S3).

Good correlation between ^{222}Rn and methane in the lake water (Fig. 2) and the enrichment of both gases close to the lake shoreline, where the active layer interfaces with the lake and where water from the active layer may enter the lake, are consistent with the transport of methane to the lake from this source (Fig. 3). We did not observe bottom water enrichments in methane or ^{222}Rn in water depth profile sampling conducted during our campaigns (in 2011, surface water ^{222}Rn was $9.2 \times 10^3 \text{ dpm} \cdot \text{m}^{-3}$ with methane concentrations of 93 nM, while bottom water ^{222}Rn was $9.0 \times 10^3 \text{ dpm} \cdot \text{m}^{-3}$ with methane concentrations of 87 nM; in 2012, water ^{222}Rn was $5.8 \times 10^3 \text{ dpm} \cdot \text{m}^{-3}$ with methane concentrations of 132 nM, while bottom water ^{222}Rn was $3.1 \times 10^3 \text{ dpm} \cdot \text{m}^{-3}$ with methane concentrations of 73 nM), indicating that sediments in Toolik Lake are not a major source of these gases, at least during the sampling period; Ra was also not enriched at depth (Table S1). Indeed, Cornwell and Kipphut

(25) found very low organic matter sedimentation rates and low dissolved oxygen consumption rates within Toolik Lake sediments, suggesting that the methane in Toolik Lake may be from a different origin. Similar spatial distribution patterns of methane, showing highest concentrations close to the lakeshore, have been seen in other Arctic lakes (9, 26, 27). While other explanations for this spatial distribution are possible (e.g., active slumping of organic-rich soil along the shoreline or wave-induced methane release), this is also consistent with the allochthonous input from the active layer as we propose here, which may contribute to the total lake methane inventory. This methane has the potential to be emitted to the atmosphere from the lake surface.

Methane input rates to the lake from the active layer depend on the SGD flux and on methane concentrations in the active layer water. The former is a function of the hydrogeology (aquifer recharge, porosity, permeability, and hydraulic head) of the different lake watersheds, and the latter is a function of methane production in the active layer. Elevated methane concentrations occur naturally in anoxic conditions via biogenic production in organic-rich soils. In the Arctic, organic soil carbon storage in near-surface permafrost can be substantial and can

Table 2. Water and methane discharge from the active layer determined using Table 1 ^{222}Rn values

Method	Active layer water discharge, $\text{m}^3 \cdot \text{d}^{-1}$	Associated methane flux, $\text{g} \cdot \text{m}^{-2} \cdot \text{y}^{-1}$ (1)
Whalen and Cornwell, 1985 water budget "undefined flow"	3.64×10^4	NA
2011 ^{222}Rn mass balance	$2.1 \pm 1.3 \times 10^4$ *	2.9
2012 ^{222}Rn mass balance	$1.6 \pm 0.6 \times 10^4$ **	0.5

NA, not available.

*The estimates and attributed uncertainties are based on average ^{222}Rn concentration from three moorings deployed during each of the sampling campaigns around the perimeter of the lake and thus representative of the spatial heterogeneity in groundwater discharge to Toolik Lake, not on the method analytical precision.

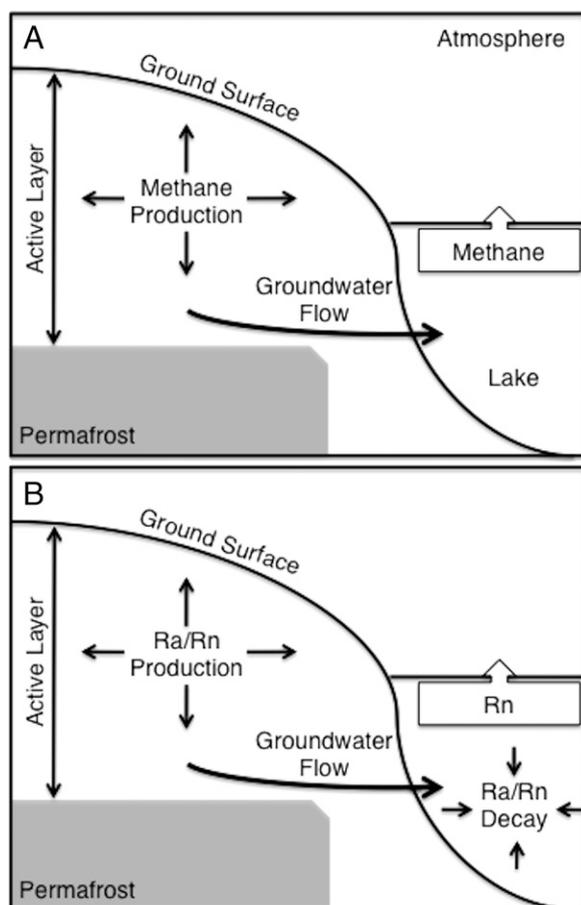


Fig. 1. (A) Schematic diagram of active layer water-associated methane flux to Arctic lakes and related climate feedbacks. Warming for more than a century has resulted in various degrees of permafrost thawing. As climate warms, permafrost stability is likely to decrease further. Methane produced in the active layer above the permafrost will be transported and released to receiving waters, eventually reaching the atmosphere and providing a positive feedback to global warming. (B) Schematic diagram of Ra and Rn enrichment in the active layer due to production in soil and peat, and their transport via groundwater to the lake where they radioactively decay and Rn-like methane is also released to the atmosphere.

support methane production in the active layer. Although limited, the methane isotope data ($\delta^{13}\text{C} = -58.9 \pm 3$) from active layer water samples ($n = 4$) taken around Toolik Lake suggest a biological source, consistent with lake isotope values (26).

Two interesting aspects of our data warrant further consideration; first, while ^{224}Ra and ^{222}Rn in groundwater samples collected at different locations throughout the lake watersheds show relatively consistent activities, methane concentrations in the active layer vary considerably, both spatially and temporally; and second, although all lake water ^{222}Rn and methane data are strongly correlated, the relationship is differentiated into two trends (Fig. 2), depending on sampling location and date. These observations suggest considerable spatial variability in methane production within the active layer. This is likely due to differences in organic matter content, water saturation, temperature, pH, and additional parameters that influence soil methanogenesis (3). Differences in water flow rate, which depends on watershed and soil characteristics, as well as the thickness of the active layer, may also affect the methane input through this conduit, as seen by the $\sim 25\%$ difference in SGD discharge between years (10).

We suspect that active layer water-associated methane input to Arctic lakes is a ubiquitous phenomenon. Our active layer methane input estimates from Toolik Lake are likely lower than in other lakes in permafrost regions due to the small amount of organic matter in the active layer around Lake Toolik compared with other lakes, the relatively large size of the lake (e.g., small shoreline length compared with lake surface area), and the somewhat limited groundwater flow due to the shallow active layer and continuous permafrost in the Northern Arctic region (more SGD is expected farther south in the noncontinuous permafrost regions). However, further investigation must be conducted to verify this hypothesis and identify and quantify the processes that control methane inputs from the active layer to Arctic lakes. Importantly, the controls on methane input to lakes from the active layer are fundamentally different from those impacting in situ methane production rates within the lakes, and the response of these disparate processes to climate and environmental change is also likely to be distinct (28). Future methane and other carbon emissions from the Arctic, as well as the ecological and economic impacts of these sources and their response to climate change remain topics of great interest (3, 29–34). The accuracy of atmospheric methane emissions predictions, and ultimately the extent of climate change that can be expected in the Arctic, depends on a holistic understanding of Arctic methane dynamics, including processes within the permafrost active layer.

Materials and Methods

Field Sampling. Lake water, active layer water, and river water samples were collected in August 2011 and July 2012 following standard procedures for ^{224}Ra activities (35, 36), ^{222}Rn activities (37–39), and CH_4 concentration (40, 41). Surface lake water samples were collected from both the interior and perimeter areas of the lake. A deep water sample for Ra analysis was collected at ~ 14 m above the deepest point of the lake, a localized 25-m-deep basin surrounded by relatively shallow (< 3 m) areas, using a submersible pump. Active layer water samples were collected from the perimeter of the lake right at the lake shoreline by installing temporary well points or by digging pits. The depths of the well points and pits were limited by the depth of the active layer (< 0.5 m typically from the soil surface to the upper boundary of the permafrost). Water samples were also collected from the thaw bulb below the lake by means of the Toolik Lake Field Station camp water supply well (depth ~ 15 m below lake). Surface river water samples were collected from the primary and secondary inflows into Toolik Lake. Specific conductivity and temperature were measured with a handheld YSI 85 multiprobe.

Radon Activity. A survey coupling the measurement of ^{222}Rn and CH_4 was conducted in which instruments for the measurement of both gases were

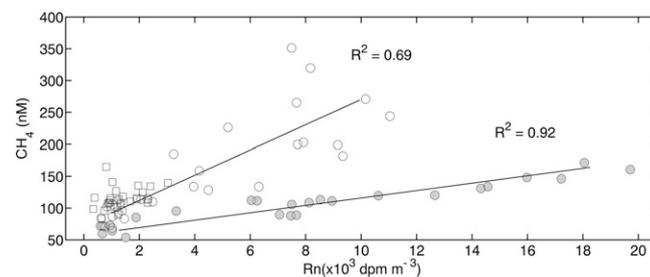


Fig. 2. Correlation between methane concentration ($[\text{CH}_4]$) (nanomolar) and ^{222}Rn activity ($10^3 \text{ dpm} \cdot \text{m}^{-3}$) concurrent measurements in Toolik Lake during the summers of 2011 and 2012. The data from 2011 (circles) fall on two trends, depending on location. Samples from the east and south sections of the lake fall on the tighter lower trend ($r^2 = 0.92$) with filled symbols; data from the west side of the lake fall on the upper steeper trend ($r^2 = 0.69$) with open symbols. All of the data from 2012 (squares) fall on the steeper trend with the 2011 west lake data. Positive relation is also seen between Ra and CH_4 (e.g., active layer water samples have high concentrations while lake water samples have low concentrations); however, due to the limited number of Ra data points, we did not derive a correlation.

atmospheric losses (based on wind speed and temperature data), the total ^{222}Rn input flux is estimated (38).

Radium Activity. Large volumes (300–500 L per sample) of lake water and river water were collected using either submersible pumps or buckets. Active layer water samples (volume 8–500 L per sample) were collected using submersible pumps, and thaw bulb water was collected from the camp water supply well. Active layer water samples were also collected in a few other lakes (e.g., a lake at the Barrow Environmental Observatory, 71.278425°N, 156.633399°W; Imikpuk Lake at Barrow, 71.331791°N, 156.661251°W; and in several small lakes near Toolik N1, NE2, S3, E1, and E5) and gave similar results (high Ra and high methane in active layer water; see *SI Text*); however, due to the limited data, water and methane fluxes were not calculated. Ra from the water samples was collected on MnO_2 -impregnated acrylic fiber [flow rate $<2\text{ L}\cdot\text{min}^{-1}$ (35)]. Samples were shipped to the University of California, Santa Cruz, for analysis on a Radium Delayed

Coincidence Counter (RaDeCC) for measurement of ^{224}Ra activities within 5 d (Alaska Air Cargo's GoldStreak Package Express). The fibers were analyzed on the RaDeCC again 3–5 wk after collection for correction of the ^{228}Th supported Ra. Standards were run on a monthly basis as part of the quality control for maintenance of the instrument. Analytical error was calculated using established procedures (36) and was typically $<10\%$.

Active layer water fluxes were also determined from ^{224}Ra mass balance. (See *SI Text* for comparison with Rn-derived fluxes and reported methane fluxes to the atmosphere from Toolik Lake.)

Methane Concentration. Samples for methane analysis were collected and prepared as described previously (40, 41).

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