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Source characterization and temporal variation of methane seepage from thermokarst lakes on the Alaska North Slope in response to Arctic climate change

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CURRENT STATE OF INFORMATION OR TECHNOLOGY

Background

Atmospheric methane is a potent greenhouse gas, contributing directly to increasing CO_2 concentrations once oxidized (Badr et al., 1992; Kheshgi et al., 1999). Yet, profound knowledge gaps remain regarding nearly all aspects of past and present methane emissions from terrestrial arctic sites and the potential increases in methane flux under the Intergovernmental Panel on Climate Change scenarios (IPCC, 2001; IPCC, 2007) or even more conservative warming scenarios. Predicted increases in methane flux, connected in large part to expected changes in the thickness of permafrost, will in turn affect atmospheric methane concentrations (Walter et al., 2007; Zhuang et al., 2006).

Related to the overarching issue of present and future arctic methane emissions is the more focused problem of the susceptibility of arctic methane hydrate deposits to global climate change. Much of the published research on methane hydrates purports to be partially motivated by the potential that global warming could lead to dissociation of a portion of the global hydrate reservoir, which may in turn exacerbate global warming if/once the methane reaches the atmosphere. Permafrost gas hydrates, here defined to include gas hydrates both within and beneath terrestrial (not arctic shelf) permafrost, are thought to be particularly susceptible to climate forcing since the overlying thermal buffer of ocean water is absent. At the same time, the absence of ocean water, where much of the methane emitted at the seafloor may dissolve or be oxidized, also means that dissociation of permafrost hydrates leads to more direct and presumably more efficient methane transfer to the atmosphere. To date, no study has proved that dissociating methane hydrates are contributing to arctic methane emissions, nor have sufficient field data been acquired to assess the potential for such a contribution in the future. This issue is central to the evaluation of the role of methane hydrates in climate change.

Terrestrial methane seeps occur at all latitudes, although the seep mechanisms and methane sources vary locally and regionally (e.g., based on soil organic content, microbial processes, existence of deep hydrocarbon system, and the availability of migration conduits) and by latitude (i.e., reflecting largely climatic factors). To date, only a limited number of studies in Alaska and Russia have revealed that the contemporary methane system is characterized by a variety of seep types, ranging from ebullition sites within thermokarst lakes to gas seeps (Walter et al., 2008; Walter et al., 2006) at river edges or even within the tundra. Thermokarst lakes, which are freshwater lakes that form in topographic depressions as permafrost thaws, are particularly important to arctic methane flux and indeed global methane budgets. A recent study estimated that, taken alone, thermokarst lakes north of 45^oN (includes those in mountainous regions at lower latitudes) emit 24.2 ± 10.5 Tg CH_4 yr^{-1} directly to the atmosphere (Walter et al., 2007), with thermokarst lakes having the highest flux (Walter et al., 2007). While this methane flux is less than that from lower latitudes, it represents flux from only a single type of arctic seep and is about two times the flux from the oceans (Reeburgh, 2007). As opposed to recent trends in some lower-latitude discontinuous permafrost areas, where lakes may be disappearing due to permafrost thaw and climate change (Smith et al., 2004), emissions from high-latitude lakes are expected to increase throughout this century, particularly in regions of continuous, icy, organic-rich permafrost like the Alaskan North Slope. In these areas, the number and size of thermokarst lakes have been increasing during the past 30 years (Smith et al., 2005; Walter et al., 2006). Thermokarst lakes typically develop roots or "thaw bulbs" of water-saturated, organic-rich material that hosts an active microbial community producing large amounts of methane released primarily through ebullition (Walter et al., 2008; Zimov et al., 1997). Where not frozen, geologic structures (e.g., faults) or permeable strata may also provide conduits for migration of other methane from deep hydrocarbon sources, coalbeds, and possibly dissociating gas hydrate. A critical feature of some large thermokarst lakes is the development of thaw bulbs that exceed tens of meters and sometimes thicken to more than 100 m. Such thick thaw bulbs radically perturb near-surface thermal gradients (Schwamborn et al., 2000) and hydrologic regimes and can potentially disrupt the top of the hydrate stability zone. This makes thermokarst lakes the best terrestrial laboratory for encountering terrestrial methane emissions that may have a hydrate-derived component. High latitude regions, including the ~230 km^2 area of the Alaskan North Slope, are uniquely poised to respond more rapidly to future climate change than most other land areas.

Current Approaches and Their Limitations

A major challenge for predicting the future impact of climate change on methane emissions in the Arctic is the paucity of contemporary data to establish baseline conditions.

Methane flux and seep pathways: While methane emissions from terrestrial sites can be studied using a variety of established approaches (Walter et al., 2007; Walter et al., 2006), these methods have not been widely applied at seeps sites on the Alaska North Slope. Also lacking are constraints on the pathways for methane migration and emission, which may include faults that tap into biogenic methane in thaw bulbs, deeper thermogenic methane, or possibly fluids and gas derived from dissociating hydrate.

Methane sources: The sources of arctic methane are known only to first order. Characterizing the sources of methane and associated gases is important for determining the nature of present-day emissions (i.e., how much is derived from microbial processes, hydrate dissociation, and migration of coalbed/hydrocarbon system gases) and predicting how the flux of gas from individual thermokarst lakes will respond to arctic climate change. For example, lakes dominated by a flux of shallow microbial gas produced within the thaw bulb will be affected by warming because soil erosion caused by permafrost thawing will continue or accelerate the transport of labile organic matter to the lakebed sediments, where it is biologically converted to methane. In contrast, thermokarst lakes fed by deep gas sources could interact with the gas hydrate reservoir and be affected by warming because downward propagation of heat could lead to gas hydrate dissociation, which is likely to increase the vertical gas flux.

Methane sinks: The sinks for arctic methane and the biological systems involved in methane consumption are poorly understood. Most critically, the role of aerobic methane oxidation in mitigating the flux of methane from lake systems has never been quantified. Despite the large net flux of methane from thermokarst lakes, there is no information on the extent to which oxidation in the sediments and water column of thermokarst lakes regulates the net flux. Nor have organisms responsible for methane oxidation in arctic Alaska been identified. Identification of methane oxidizers is critical, since different methane oxidizing organisms respond differently to and have different tolerance to environmental change (Einola et al., 2007; King, 1997; Mor et al., 2006).

Spatiotemporal variability in the context of climate change: Predictions about the future of methane production from terrestrial seeps in the Arctic will require measurements of contemporary flux on a range of timescales and investigation of the geologic record of methane emissions. No known group has systematically undertaken routine flux measurements from arctic terrestrial seeps, although spot sampling and observations provide provocative indications for large (10-fold) abrupt and/or seasonal/inter-annual changes in flux (pers. obsv. K. Hinkel, unpublished data, K. Walter). In terms of past methane emissions, the instrumental record of methane emissions is short (IPCC, 2007), and longer-term records (thousands of years) of methane production generally relate to the global atmosphere (O'hara, 2008; Sowers, 2006), not the temporal variability of methane production from specific seep sites in the Arctic. However, recent analysis of basal ages of thermokarst lakes in Alaska and Siberia suggests that the flare up of thermokarst lakes during deglaciation caused up to 88% of the increase in northern hemisphere sources to atmospheric CH_4 concentration (Walter et al., 2007). Using geologic approaches, site-specific, long-term records (greater than the instrumental record) of the past duration and variability of methane production at current seep sites should be reconstructed. Such records will provide data to understand variability at specific seep sites in the larger context of pan-arctic climate change records.

DEVELOPMENT STRATEGY

The University of Alaska (UAF) Team will target these knowledge gaps through the application of state-of-the-art field measurements and analytical techniques to better characterize methane flux, methane sources and sinks, and spatial and temporal variability of methane seepage from thermokarst lakes in the Arctic, with the goal of assessing the past and current state and future sensitivity of the terrestrial arctic methane system to climate change processes.

Assessing changes in the size or intensity of terrestrial arctic methane seeps as both a possible cause and a probable effect of future climate change requires a baseline understanding of (a) the contemporary factors controlling the spatial distribution and temporal frequency of arctic methane emissions; (b) the key methane sources; and (c) the associated methane flux.

This project's solution to the scientific problems outlined above is to conduct a study of methane sources, sinks, and climate-linked records over geologic time scales using thermokarst lake study sites at two permafrost/gas hydrate areas on the Alaska North Slope and a Fairbanks-area control site (permafrost only) characterized by widespread methane seepage. The approach will integrate field-based measurements and direct sampling/surveying, extensive laboratory analyses, and synthesis activities designed to draw on the unique strengths and backgrounds of our multidisciplinary PI group, which includes expertise on limnology, biogeochemistry, organic chemistry, microbiology, geophysics, hydrology, arctic permafrost dynamics, and gas hydrate systems.

Measuring methane flux on multiple temporal scales and constraining migration paths**:** The UAF Team will use published protocols (Walter et al., 2008; Walter et al., 2006) to measure methane flux during visits to our field sites. Using the Fairbanks control site, we will also quantitatively determine whether early winter surveys of lake-ice bubbles can be reliably converted to annual whole-lake CH_4 emissions. Ebullition will be monitored at several Fairbanks-area lake sites for a year using underwater/under ice chambers equipped with automated bubble traps and fixed in the water column. At the North Slope thermokarst lake sites, methane flux will be measured using bubble traps deployed seasonally. The Qalluuraq Lake seep near Atqasuk will be studied in greater detail to quantify seasonal and inter-annual variability in methane emissions for comparison with data obtained at the Kilarney Lake Fairbanks-area control site. A full-scale geophysical imaging program in thermokarst lakes is beyond the scope of this project, but the UAF Team will carry out reconnaissance geophysical imaging for at least one of our sites to constrain thaw bulb thickness, loci of lakebed methane emissions, and possible pathways for fluid/gas migration within the thaw bulb and at the edges of thermokarst lakes. In light of the paucity of imaging data specifically targeted at understanding gas pathways and fluid/gas migration in these settings, almost any high-quality data we acquire would significantly increase potential understanding of the interplay among the chemistry, microbiology, hydrology, and geology in these complex thermokarst systems.

Gas sources and methanogenesis: There are numerous potential sources for the methane and other associated gases fluxing from thermokarst lakes. Potential deep-thermogenic sources are conventional natural gas from petroleum source rocks and coal-bed methane from humic source rocks. Potential microbial sources in thermokarst lakes are the carbonate reduction pathway and acetate fermentation pathway (Walter et al., 2008), which can occur in the relatively shallow thaw bulb sediments or deeper organic carbon reservoirs. To fully characterize the source and origin of gases emitted from thermokarst lakes, UAF will analyze the isotopic signatures (^{13}C, ^{14}C and D) of the C_1-C_5 hydrocarbons and CO_2 and composition of gas mixtures and investigate the biogeochemistry of coring-accessible microbial methane generation processes. Noble gas ratio analyses will also be conducted to investigate if it is possible to determine if hydrate derived gases are escaping the arctic permafrost through thermokarst lakes. Pore water will be collected from sediment cores and analyzed for chloride and other cations for evidence of deep-seated fluid origins. Concentrations and $\delta^{13}C$ signatures of dissolved inorganic carbon (DIC), dissolved organic carbon (DOC) and volatile organic acids (VFAs, includes acetate) will also obtained as part of the effort to obtain a complete mass balance of carbon cycling in the methanogenic zone of the thaw bulb.

<u>Methane oxidation in Alaskan thermokarst lakes:</u> The observation that the methane flux from thermokarst lakes is dominated by gas ebullition (Walter et al., 2007) is consist with observations in gas hydrate-bearing cold seeps in marine settings where gas discharge at discrete point sources can be six orders-of-magnitude higher than non-ebullition sites (Linke et al., 1994). However, the absence of a significant methane flux at 'background' sites does not imply that methane oxidation is not important in the overall methane budget of thermokarst lakes. In contrast, it suggests that aerobic methane oxidation could play a major role in mitigating the flux of methane from lake sediments where ebullition does not occur. In marine systems up to 28% of the methane consumed at the sediment water interface is due to aerobic methane oxidation (Sommer et al., 2006). Furthermore, oxidation of methane in the water column could also be an important term in the net flux of methane, as identified in the marine environment (Mau et al., 2007; Valentine et al., 2001). In this study, UAF will employ a comprehensive, multi-disciplinary approach for characterizing the rates of methane consumption, as well as and phylogeny of the microbes involved. The techniques to be employed include:

> *Characterization and quantification of aerobic methane oxidation:* Collection of pore water and sedimentary material from sediment cores for geochemical analysis of dissolved ions, gases and organic matter to identify where aerobic methane oxidation is occurring and characterize the process.
>
> *Characterization methane oxidizing microorganism using stable isotope probing and lipid biomarker analysis:* Stable isotope probing (SIP) will be used to identify the microbial populations active in methane oxidation at the field study sites. The analyses will benefit efforts to predict the impacts of climate change on methane oxidation in the Arctic. Lipid biomarker analysis provides a broad overview of the dominant microbial players actively cycling organic matter and their relative abundance. Coupled with 'traditional' compound specific stable isotope analysis of the non-polar lipid components and the stable isotope probing technique, an unprecedented level of information about the microbes actively involved in the aerobic oxidation of methane is possible. No known coupling of these techniques in a natural system has been previously undertaken.

<u>Establishing a long-term record of the variability in methane emissions in relation arctic climate change:</u> Evidence preserved in dated sediment cores taken from seep sites on the Alaska North Slope will be used to establish long-term records of past methane emissions. The hypothesis is that a direct relationship between the quantity of methane fluxing from the lakes and the isotopic signature of organic markers for aerobic methanotrophs exists and evidence of this can be found in the remains of chironomids (aquatic insect) larvae that consumed and assimilated the unique carbon isotope signature of the methanotrophic microbes. To test this hypothesis, UAF will compare the isotopic signature of methanotrophic organic markers and insect larvae from surface sediments of lakes with varying degrees of active methane discharge and methane oxidation. Reconstructions of past temperature will be based on oxygen isotopic analyses of fossils preserved in cores from the study sites.

The chronology of sediment deposition in the lake systems will be reconstructed from radiocarbon dating of macrofossil plant remains and compound specific algal biomarkers by accelerator mass spectrometry (AMS). Plant macrofossils provide an economic way of obtaining radiocarbon ages. However, complications related to the erosion of "pre-aged" soils into thermokarst lakes may have translocated fossil plant remains into the lake system, which could yield erroneously old ages for the associated sediments. To identify and circumvent this potential problem, we will also extract and concentrate lipid biomarkers from planktonic algae that built their biomass from dissolved CO_2 in equilibrium with the atmosphere, thereby providing a more accurate calendar age estimate for the sedimentary horizon within which they were buried. The calibration of the radiocarbon dates will allow direct chronological comparisons between cores taken from our different study sites as well as allow comparison to previously published records of climate change in the Arctic (Kaufman, 2004). Previous results from cores of sediment taken at sites north of the Brooks range in Alaska indicate that simply acquiring a 1 m core from our study sites is likely to yield a temporal record greater than 1000 years and up to several thousand years (Eisner, 1991; Eisner and Colinvaux, 1992; Eisner and Peterson, 1998; Oswald et al., 2003), which is a considerably longer than the instrumental record for arctic Alaska.

The flux of methane from thermokarst lake sediments is likely to support vigorous aerobic methane oxidation and production of ^{13}C-depleted methanotrophic microbial biomass in the water column and aerobic sediments. Burial and preservation of molecular fossils from the microbial biomass provides a long-term record of this activity (Hinrichs et al., 2003). By unraveling the history of sediment deposition in the lake basin, it is possible to reconstruct the history of methane flux for an individual lake. UAF will utilize the carbon isotope record from both methane oxidizing microbes that consume ^{13}C-depleted methane and insect larvae that consume the ^{13}C-depleted microbes as proxies for the methane flux.

POTENTIAL IMPACT OF THIS PROJECT

This project will be among the first to acquire direct, targeted data to assess whether permafrost gas hydrates are among the most susceptible to climate change. In the process of acquiring such data, methane fingerprinting techniques will be used that are at the cutting edge and extend these methods to the terrestrial hydrate system for the first time. Such developments will also advance the potential for methane source characterization in conventional methane systems and in the marine hydrate system and be of potential use to researchers on other DOE-NETL hydrates projects.

This research will produce publications in international peer reviewed journals. Data produced from the integrated approach to studying methane being produced from thermokarst lakes will result in:

1) quantification of methane emissions at seasonal to inter-annual scales in thermokarst lakes—both with and without hydrates—and conduct a robust study to determine the reliability of upscaling from flux measurements at discrete times and specific sites to longer timescales and larger areas.

2) determination of constrained gas/fluid migration pathways and the potential of the thaw bulb to interact with the gas hydrate stability zone through reconnaissance geophysical imaging and quantify relative contributions of shallow microbial and deeper gas sources (e.g., conventional hydrocarbon gas, coalbed methane, and possibly a gas hydrate source).

3) quantification and characterization of the processes associated with methane oxidation within the water column and lake bottom sediments, and

4) reconstruction of a millennial-scale record of methane emissions for the thermokarst lake sediment records and relate the history of methane flux to records of past arctic climate change.

This robust baseline data about contemporary methane flux from some of the most perturbed areas (thermokarst lakes and their thaw bulbs) of the Alaskan North Slope will be invaluable in decades to come as potential climate warming scenarios play out. At the same time, the methane flux data will help to further refine climate modeling and the contribution of atmospheric methane derived from arctic sources. Lastly, this project will increase the understanding of the relative roles of diffuse and discrete (seep) methane flux in terrestrial settings—a necessity for making progress on aspects of climate change and gas hydrate reservoir dynamics linked to climate change. In short, this project will have benefits in terms of broaching first-order scientific questions, developing new techniques and extending existing techniques to new environments, and building critical collaborations and knowledge sharing between marine and terrestrial methane seep and gas hydrate communities.

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