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Key Points:
- Twenty-nine years of CH_4 observations at Barrow, AK, suggest emissions stagnant despite warming
- Seasonal CH_4 emissions at Barrow continue late into December
- Projected enhancements in Arctic CH_4 emissions will have small global impact

Supporting Information:
- Supporting Information S1

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No significant increase in long-term CH_4 emissions on North Slope of Alaska despite significant increase in air temperature

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Abstract Continuous measurements of atmospheric methane (CH_4) mole fractions measured by NOAA's Global Greenhouse Gas Reference Network in Barrow, AK (BRW), show strong enhancements above background values when winds come from the land sector from July to December from 1986 to 2015, indicating that emissions from arctic tundra continue through autumn and into early winter. Twenty-nine years of measurements show little change in seasonal mean land sector CH_4 enhancements, despite an increase in annual mean temperatures of 1.2 ± 0.8°C/decade (2σ). The record does reveal small increases in CH_4 enhancements in November and December after 2010 due to increased late-season emissions. The lack of significant long-term trends suggests that more complex biogeochemical processes are counteracting the observed short-term (monthly) temperature sensitivity of 5.0 ± 3.6 ppb CH_4/°C. Our results suggest that even the observed short-term temperature sensitivity from the Arctic will have little impact on the global atmospheric CH_4 budget in the long term if future trajectories evolve with the same temperature sensitivity.

1. Introduction

The last two decades have witnessed significant changes to the Arctic climate in the form of surface and below-ground warming [*Cohen et al.*, 2014; *Romanovsky et al.*, 2010] and massive decreases in sea ice [*Maslowski et al.*, 2012]. These large physical changes in the Arctic are driving changes in biogeochemical processes that may result in either positive or negative feedback in Arctic climate [*Parmentier et al.*, 2013]. In particular, there has been significant speculation about how Arctic CH_4 emissions to the atmosphere might change as a result of anaerobic degradation of massive amounts of labile organic carbon that exist in the permafrost underlying large portions of the Arctic [*Cohen et al.*, 2014; *Schaefer et al.*, 2014; *Schneider von Deimling et al.*, 2012; *Schuur et al.*, 2015; *Schuur et al.*, 2013]. In total, the near surface (0–3 m depth) contains 1035 billion tons (gigatons) of carbon (GtC) [*Hugelius et al.*, 2014], 2.5 times the amount of anthropogenic carbon (as CO_2) that has been released to the atmosphere since the industrial revolution [*Sabine et al.*, 2004]. Because CH_4 has almost 28 times the radiative impact of CO_2 on a 100 year timescale [*Myhre et al.*, 2013], it is critical that we understand the fate of this large carbon reservoir.

The idea that this carbon reservoir has the potential to be released as CH_4 has catalyzed significant research focused on a better understanding of the processes that are likely to take place as organic carbon sequestered in permafrost is released and metabolized. Two key questions arise from this research: (1) What fraction of the mobilized soil organic carbon will be released as CH_4? and (2) What is the sensitivity of CH_4 emissions to temperature change? These two questions are intertwined. They cannot be easily separated because an increase in temperature will also lead to a change in many of the variables (such as soil water content and aboveground biomass) that influence anaerobic CH_4 production. Process studies suggest that both water table depth and soil temperature are significant drivers for CH_4 emissions [*McEwing et al.*, 2015; *Natali et al.*, 2015; *Olefeldt et al.*, 2013; *Sturtevant et al.*, 2012]. Modeling studies that attempt to upscale process-level CH_4 emissions estimates to regional and global scales conclude that with the observed changes in temperature during the last two decades, CH_4 emissions should also have increased significantly [*Koven et al.*, 2013; *Zhu et al.*, 2014; *Riley et al.*, 2011]. On the other hand, atmospheric observations of CH_4 gradients derived from the Global Greenhouse Gas Reference Network (http://www.esrl.noaa.gov/gmd/ccgg/, GGGRN) have not

indicated any significant increase in CH_4 emissions from either inverse analysis [*Bergamaschi et al*., 2013; *Bruhwiler et al*., 2014] or analysis of trends in the interpolar difference [*Dlugokencky et al*., 2009].

In this study we leverage a 29 year record of quasi-continuous atmospheric CH_4 mole fraction observations made at the NOAA/Earth System Research Laboratory (ESRL) Observatory in Barrow, AK (BRW), to better understand how ecosystem emissions of CH_4 are responding to significant temperature changes at BRW and the surrounding region [*Wendler et al*., 2014]. We take advantage of the fact that BRW is not only an excellent site for monitoring the baseline CH_4 mole fraction for the pan-Arctic region, but also a site that has a large sensitivity to seasonal increases in CH_4 coming from Alaska's North Slope, which lies to the south of BRW (Figure S1 in the supporting information). The average difference between the northeasterly (seaward) baseline observations and the southern (landward) sector observations throughout the year enables us to examine the long-term changes in CH_4 emissions from the North Slope. This region of the Arctic tundra has experienced unprecedented air temperature changes from July to December (1.2°C/decade), more than double those of the pan-Arctic region during the past 29 years [*Wendler et al*., 2014]. The surface observations of CH_4 mole fraction provide an aggregate picture of how the processes controlling CH_4 emissions on the North Slope are responding to climate change both in the long term (decadal) and the short term (monthly).

2. Methods: Barrow Atmospheric CH_4 Mole Fraction Measurements

Barrow Observatory (71°19'N, 156°36'W) is located on tundra 8 km northeast of Barrow, Alaska (Figure S1). The dry mole fraction of CH_4 has been measured at this site since January 1986 from an inlet 16 m above ground level. From January 1986 to April 1996, CH_4 mole fraction measurements were made using a Carle Series 400 gas chromatograph (GC, EG&G, Tulsa, Oklahoma) with flame ionization detection (FID) [*Dlugokencky et al*., 1995]. From May 1996 to May 2012, an 6890 GC with FID was used. Repeatability of the analytical systems were ~3 ppb for the Carle GC and ~1.5 ppb for the HP (both assessed as 1σ of repeat injections of dry, natural air from a cylinder). In both cases, a single-point calibration extrapolated through zero was used to calibrate the response of the FID. Due to a lapse in funding, the GC system was shut down in June of 2012 and temporarily replaced by a cavity ring-down spectrometer (Picarro, 2401) from September of 2012 to June of 2013. In June 2013 an off-axis, integrated cavity output spectrometer (Los Gatos Research, model GGA-24EP) was installed. Its response is calibrated relative to a reference cylinder every 2 weeks with a suite of standards covering the nominal range 1590 to 2460 ppb. Drift is tracked by measurement of a reference (dry, natural air in a cylinder) every 65 min. Instrument performance is assessed with a well-calibrated surveillance cylinder every 23.5 h. Reproducibility of the measurements of the surveillance cylinder is ±0.25 ppb (1σ). All measurements are made on the World Meteorological Organization CH_4 X2004 scale [*Dlugokencky et al*., 2005].

All measurements for this study have been reduced to hourly means. To maximize the area of influence and minimize local influence from point sources, hourly averages were only considered when the wind speeds were greater than 3.0 m/s and variability (1σ) in the CH_4 mole fraction was less than 10 ppb. Based on the wind speed criterion, an air mass will travel ~10.4 km (360 s × 3 m/s) or greater during the hour-long average of each measurement (Figure S1).

In addition to CH_4 mole fraction measurements, wind and temperature measurements at BRW were used in this analysis. Wind direction and magnitude were measured with a Bendix Aerovane (Model UMQ-5) from the start of the methane measurement period (1986) to the summer of 1994 followed by a RM Young Wind Monitor Sensor (Model 5103) from 1994 to present. Temperature was measured at 2 m above ground from 1986 to 1994 using a YSI thermistor (Model 15133) and at 2 m and 10 m from 15 March 1994 to present using Logan Enterprise thermistors (4150 series). In this study we only use 2 m temperature measurements because the 10 m temperature measurements do not span the whole time series, but comparisons made during the 1994–2015 period suggest that similar trends can be calculated from both sensor heights.

Concurrent albedo and belowground temperature measurements were made at the BRW site (see detailed measurement methods in Text S2).

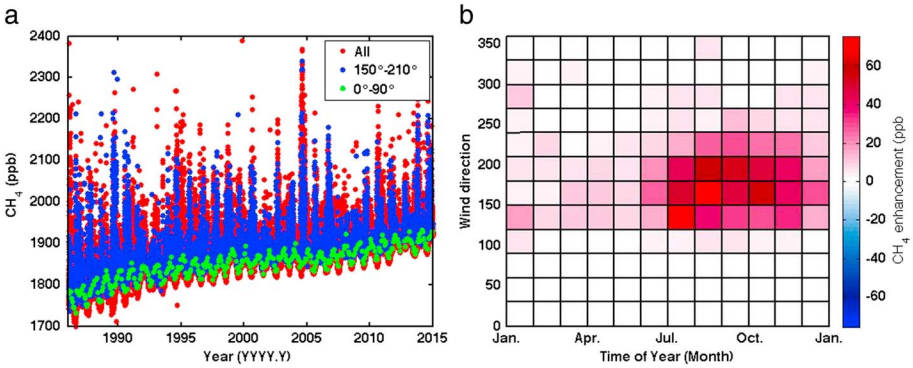

Figure 1. Barrow, Alaska (71°19′N, 156°36′W), CH_4 mole fraction. (a) Hourly CH_4 mole fraction measurements made from 1986 to 2015. Green points show mean monthly averages when winds were coming from 0° to 90°, and blue points show all measurements made when winds were coming from the land sector (150°–210°). Red points show measurements from other sectors. (b) Enhancement of CH_4 mole fraction relative to the clean air sector (0°–90°) binned by month and direction when winds are >3.0 m/s and when the standard deviation of the hourly average CH_4 is less than 10 ppb at BRW.

3. Results

3.1. North Slope Directional Sector Analysis

A time series plot of all the CH_4 measurements from BRW exhibits a strong seasonal cycle in the baseline concentration, which is best identified by selecting the monthly mean of all measurements made when winds are between 0° and 90° (Figure 1a). *Dlugokencky et al*. [1995] originally defined this "clean air sector" as between 20° and 100°; our analysis suggests that the sector between 0° and 90° has lower variability. To further examine the relationship between CH_4 measured in the clear air sector and all other sectors, we have subtracted the clean air sector measurements binned by month from the monthly mean of the CH_4 mole fraction in 12 evenly spaced directional sectors around BRW (Figure 1b). From this wind sector analysis a clear enhancement of CH_4 can be seen starting in July and lasting until late December when winds are coming from 120° to 240° (Figure 1b). Given the potential influence from the town of Barrow, AK (bearing 240°, Figure S1) and oil and gas operations along the coastal region (bearing 120°), we have selected the sector between 150° and 210°, where the seasonal enhancement is the strongest, as the focus of our study to determine if the land sector enhancements are increasing over time (Figure S1).

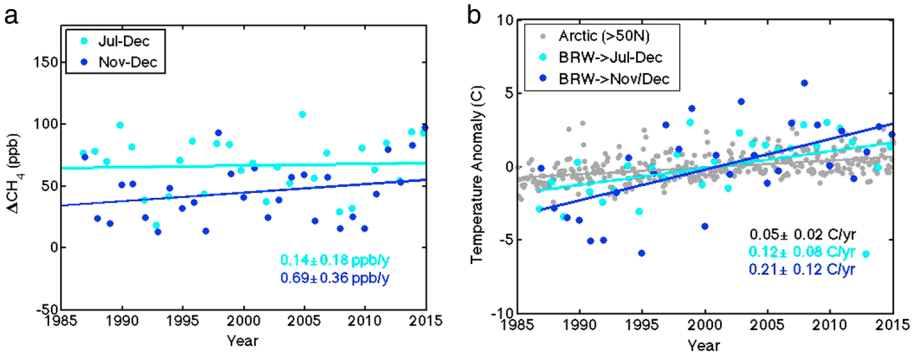

Figure 2. Trends in CH_4 enhancements and surface temperature from the North Slope 1986–2014. (a) Land sector CH_4 enhancements, July–December (cyan) and November–December (blue) during >3 m/s wind events coming between 150° and 210°. (b) Air temperature anomalies, relative to the time series mean, at BRW and the entire Arctic. Grey points are monthly anomalies in Arctic (>50°N) temperature from the GISS Surface Temperature Analysis (GISTEMP, http://www.esrl.noaa.gov/psd/data/gridded/data.gistemp.html). Cyan and blue points show surface air temperature anomalies at 2 m made at the BRW tower for 1986–2014, July–December and November–December, respectively. Trends are calculated from 1986 to 2015. Slope uncertainties (2σ) are described in Text S3.

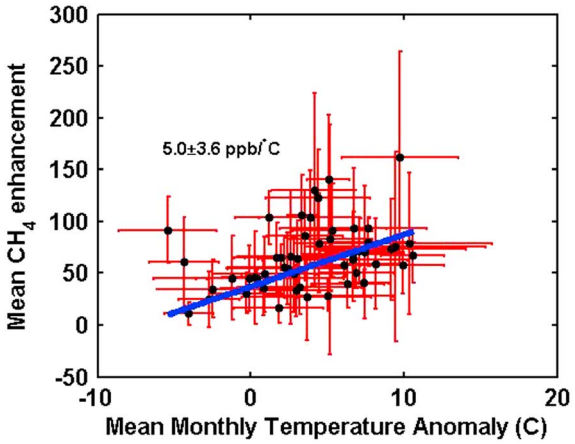

Figure 3. Average temperature deviation from monthly means versus CH_4 enhancements when air is coming from the south (150° and 210°) at Barrow during August and September. Line fit uses total weighted least squares fit to account for *x* and *y* errors in each point [*Krystek and Anton*, 2007] with uncertainties representing 1σ standard deviations in monthly mean temperatures and CH_4 enhancements. The uncertainty in the slope (d*C*/d*T*) is 2σ and is described in Text S4.

3.2. Trends in Summer and Fall CH_4 Enhancement

Given the likely connection between increased temperatures and microbial production of CH_4, we have analyzed trends in land sector CH_4 enhancements and temperature in multiple time periods during the year. There is no significant long-term trend in land sector CH_4 enhancement during the summer months (July and August) and even into early fall (September and October); however, there is a significant (2 sigma (σ)) trend in late fall (November and December, 0.69 ± 0.36 ppb CH_4/yr, 2σ, Figure 2a, Text S3). This late-fall trend in the 29 year record is mostly due to an almost 25 ppb increase in average CH_4 enhancements and the indication of a prolonged period of emissions starting in 2010 (Figure S4). This late-fall trend in CH_4 enhancements is not large enough to make the aggregate July–December trend in CH_4 enhancements significant (0.14 ± 0.18 ppb CH_4/yr, 2σ, Figure 2a, Text S3).

The lack of a long-term trend in CH_4 enhancements except in the late fall is particularly noteworthy when the large, long-term trend in average surface air temperatures for both the combined summer and fall seasons and the late fall (0.12 ± 0.08°C/yr and 0.21 ± 0.12°C/yr, respectively, Figure 2b) is considered. This late-fall trend in surface air temperatures at BRW is 4 times greater than the land-based temperature trend found over the whole Arctic during the same months (0.05 ± 0.02°C/yr, GISTEMP, http://www.esrl.noaa.gov/psd/data/gridded/data.gistemp.html).

3.3. Short-Term Temperature Sensitivity of North Slope CH_4 Emissions

Multiple analyses were performed to investigate the short-term relationship and possible correlation between surface air temperature and CH_4 enhancements coming from the land sector (150° to 210°). The only significant relationship between BRW surface air temperature and CH_4 enhancements was found by comparing the mean monthly temperature anomalies with the mean CH_4 anomalies during periods when winds were coming from the land sector during August and September. Like the CH_4 anomalies, the mean monthly temperature anomalies were calculated by subtracting mean monthly background temperature (when winds are coming from the 0° to 90° sector) from the mean monthly temperature when winds were coming from the land sector. By subtracting the background monthly mean temperature, the long-term (>monthly) changes in temperature are eliminated and only the deviations from the monthly mean temperatures of an air mass coming from the land sector are measured. Hence, the relationship derived for August and September (5.0 ± 3.6 ppb CH_4/°C, Figure 3) is defined as the short-term temperature sensitivity. No significant relationship was found for enhancements after September.

4. Discussion

A wind sector analysis of the CH_4 enhancements measured at BRW shows that the land sector enhancements have not changed significantly between 1986 and 2015 except in the late fall where the average of the last 5 years has changed in magnitude significantly. The long-term record of measurements at BRW provide an important baseline for understanding both the seasonal cycle of CH_4 emissions in the Arctic tundra region and an indication of how CH_4 emissions have changed in this region over the last 29 years.

4.1. The Representativeness of BRW Clean Air and Land Sector

To better understand the significance of the land sector CH_4 enhancements observed at BRW, it is important to contextualize the two different sectors with other available data to illustrate the extent and magnitude of the signal being generated from the North Slope and the importance of our trend analysis.

The clean air sector at BRW closely mimics the seasonal cycle of many sites including those at Alert, Canada (ALT) and Cold Bay, AK (CBA) (Figure S2) and supports the idea that this signal represents the large-scale Arctic seasonal cycle. The similarity between the CH_4 measured from the clean air sector at BRW and higher altitude flights made to the south at the Poker Flat, AK (PFA) site suggest that the clean air sector seasonal cycle is driven primarily by transport and the oxidation of methane from lower latitudes [*Sweeney et al.*, 2015]. It is also important to note that studies comparing the average background CH_4 above 53°N to those below 53°S show that the interpolar difference has actually decreased over the measurement record suggesting a slightly decreasing role of the Arctic in the global CH_4 budget [*Dlugokencky et al.*, 2003; *Worthy et al.*, 2015].

The representativeness of the land sector signal at BRW as a proxy for the North Slope can be demonstrated by the wide spread enhancements that were observed by the Carbon in Arctic Reservoirs Vulnerability Experiment (CARVE) [*Miller et al.*, 2012] (See Text S1 for background on measurements from CARVE). Multiple CARVE flights over the North Slope suggested a median CH_4 enhancement between 200 and 1500 m above sea level (asl) relative to measurements made in the free troposphere (>4500 m asl) of ~50 ppb in August and September of 2012 and 2013 (Figure S3). The bulk of measurements below 1500 m asl show enhancements of 0–150 ppb indicative of large areas of enhancements coming from natural emissions and not anthropogenic sources such as fossil fuel exploration or from the nearby town of Barrow, AK. Only a small fraction of the measurements had CH_4 enhancements >150 ppb, indicative of emissions from single-point sources with narrow plumes that are captured by only a few measurements. These flights provide a valuable piece of evidence that the large CH_4 enhancements (~65 ppb in August and September) coming from the land sector are not simply a local phenomenon but extending far to the south of Barrow, AK.

4.2. The Seasonal Cycle of North Slope CH_4

The results of this analysis confirm that the seasonal cycle of North Slope CH_4 emissions quickly ramps up in middle to late June, coincident with a rapid decrease in albedo signaling summertime snow melt and a rapid rise in the near surface (20 cm depth) soil temperatures (Figure S5). Although North Slope CH_4 enhancements peak in August and September, significant CH_4 enhancements persist until the end of December, when a rapid drop in near-surface soil temperature occurs as the active layer refreezes [*Hinkel et al.*, 2001]. This decrease in CH_4 emissions in December occurs months after albedo has increased to values indicating complete snow cover (typically middle to late September). The multiyear climatology of BRW CH_4 enhancements shows no evidence of short, intense regional-scale CH_4 pulses on the North Slope coinciding with either the spring thaw at the beginning of the growing season or with the active layer refreeze in the autumn. This result contrasts with the reports from smaller-scale studies using flux chambers that find fall Arctic CH_4 pulses [*Mastepanov et al.*, 2008; *Mastepanov et al.*, 2013]. The seasonal cycle at BRW is consistent with flux tower measurements around Barrow and to the south at Atqasuk and Ivotuk, AK [*Zona et al.*, 2016] where significant CH_4 fluxes are observed in November and December. These authors suggest that this late-fall flux is regulated by the "zero curtain" of active layer temperature, when soils are poised near 0°C but liquid water remains present. Only when soils finally freeze completely does production of CH_4 decline to the low values characteristic of deep winter.

4.3. Ecosystem Response to Temperature Change

With such large summertime enhancements in CH_4 coming from the land sector relative to the clean air sector at BRW we expect to see a proportionally large response to the change in surface air temperature measured at BRW relative to the rest of the Arctic over the last 29 years. Before the snow falls the magnitude of enhancements in CH_4 suggests a short-term (monthly) response to surface air temperature of 5.0 ± 3.6 ppb CH_4/°C (Figure 3). For reference, this linear change in temperature is equivalent to an exponential response to temperature known as Q_{10} of ~2.0 where $Q_{10} = ([\Delta CH_4]_{T2}/[\Delta CH_4]_{T1})^{10/(T2-T1)}$ such that a 20% change in the CH_4 enhancement (ΔCH_4) (e.g., $[\Delta CH_4]_{T2}/[\Delta CH_4]_{T1} = 1.2$) would require a ~3.0°C change in temperature. This temperature sensitivity characterizes the regional-scale North Slope ecosystem response to surface air temperature changes on a monthly average timescale and provides an important benchmark for ecosystem

models that predict the response of the ecosystem to surface air temperature change [e.g., *Riley et al*., 2011]. The fact that the correlation between air temperature and emissions of CH_4 is not significant after September is consistent with the onset of snow cover (Figure S5), which acts to insulate the soils from temperature anomalies in the overlying air in the short term. Unfortunately, this hypothesis cannot be tested due to a lack of long-term measurements of belowground temperature.

Given the significant increase in surface land temperatures over the last 29 years (3.5 ± 2.3°C) and the short-term response of 5.0 +/− 3.6 ppb/C in CH_4 emissions, it is puzzling to see the muted change in the July–December emissions (4 ± 6 ppb CH_4, 29 yr × 0.14 ± 0.18 ppb CH_4/yr) over the entire measurement period indicating a long-term temperature response of only 1.1 ± 1.8 ppb of CH_4/°C. The small temperature response suggests that there are other processes at play in regulating the long-term CH_4 emissions in the North Slope besides those observed in the short term. *Tagesson et al*. [2013] show very little sensitivity in CH_4 emissions to surface air temperature changes in northeast Greenland from 1997 to 2010 and suggest that the lack of sensitivity might be due to the fact that the belowground temperature is not changing. Other variables such as water inundation and types of surface biomass have been shown to correlate with methane emissions [*McEwing et al*., 2015; *Natali et al*., 2015]. It is these variables and other region-specific controls like soil nutrients, energy balance, and organic carbon composition that may be countering the influence of surface temperature change. It is also possible that organic carbon has simply been mobilized as CO_2 instead of CH_4 [*Schuur et al*., 2015]. Recent field and remote sensing observations suggest increased ice wedge melting over large expanses of tundra permafrost leading to a decrease in water inundation [*Liljedahl et al*., 2016]. It is likely that this change in water inundation has counteracted CH_4 production. Our finding illustrates the need for year-round observations of atmospheric CO_2 and CH_4 alongside physical, geochemical, and biological observations such as belowground temperature and moisture, nutrient dynamics, oxidation state, and soil carbon, as well as aboveground biomass, throughout the Arctic to better understand what is controlling CH_4 emissions.

4.4. Implications for Future Emissions of CH_4

The short- and long-term surface air temperature sensitivity based on the 29 years of observed enhancements of CH_4 in air masses coming from the North Slope provides an important basis for estimating the CH_4 emission response to changing air temperatures in Arctic tundra. By 2080 autumn (and winter) temperatures in the Arctic are expected to change by an additional 3 to 6°C [*Snow, Water, Ice, and Permafrost in the Arctic*, 2011]. Based on the long-term temperature sensitivity estimate made in this study, increases in the average enhancements on the North Slope will be only between −2 and 17 ppb (3 to 6°C × 1.1 ± 1.8 ppb of CH_4/°C). Based on the short-term relationship calculated, the enhancements may be as large as 30 ppb. These two estimates translate to a −3 to 45% change in the mean (~65 ppb) CH_4 enhancement observed at BRW from July to December. Applying this enhancement to an Arctic-wide natural emissions rate estimate of 19 Tg/yr estimated during the 1990s and 2000s [*McGuire et al*., 2012] implies that tundra-based emissions might increase to as much as 28 Tg/yr by 2080. This amount represents a small increase (1.5%) relative to the global CH_4 emissions of 553 Tg/yr that have been estimated based on atmospheric inversions [*Kirschke et al*., 2013].

5. Conclusions

This study illustrates the value of long-term measurements for establishing baseline trends and confirms the importance of late-fall and early winter CH_4 emissions on the North Slope of Alaska [*Zona et al*., 2016]. We observe a recent increase in emissions in the late fall and early winter on the North Slope, but the mean seasonal enhancements for July through December do not show statistically significant increases despite the widespread increase in surface air temperatures. This study suggests that additional sites are needed to monitor changes in CH_4 emissions from the variety of ecosystems found throughout the Arctic region in addition to the continued above and belowground studies that monitor soil temperature profiles, changes in surface biomass, and soil moisture. These sites would be extremely valuable for understanding the processes driving CH_4 emissions and for better interpreting trends in methane emissions. The lack of sensitivity to large changes in air temperature at BRW suggests that CH_4 emissions are sensitive to variables or processes that are not currently well documented. Alternatively, emissions of CO_2 should be carefully considered as a pathway for mobilization of soil organic carbon as suggested by *Schuur et al*. [2015].

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