



# Satellite ice extent, sea surface temperature, and atmospheric methane trends in the Barents and Kara Seas

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**Abstract.** Over a decade (2003-2015) of satellite data of sea-ice extent, sea surface temperature (*SST*), and methane ( $\text{CH}_4$ ) concentrations in lower troposphere over 10 focus areas within the Barents and Kara Seas (BKS) were analyzed for anomalies and trends relative to the Barents Sea. Large positive  $\text{CH}_4$  anomalies were discovered around Franz Josef Land (FJL) and offshore west Novaya Zemlya in early fall. Far smaller  $\text{CH}_4$  enhancement was found around Svalbard, downstream and north of known seabed seepage. *SST* increased in all focus areas at rates from 0.0018 to 0.15 °C yr<sup>-1</sup>,  $\text{CH}_4$  growth spanned 3.06 to 3.49 ppb yr<sup>-1</sup>.

The strongest *SST* increase was observed each year in the southeast Barents Sea in June due to strengthening of the warm Murman Current (MC), and in the south Kara Sea in September. The southeast Barents Sea, the south Kara Sea and coastal areas around FJL exhibited the strongest  $\text{CH}_4$  growth over the observation period. Likely sources are  $\text{CH}_4$  seepage from subsea permafrost and hydrate thawing and the petroleum reservoirs underlying the central and east Barents Sea and the Kara Sea. The spatial pattern was poorly related to seabed depth. However, the increase in  $\text{CH}_4$  emissions over time may be explained by a process of shoaling of strengthening warm ocean currents that would also advect the  $\text{CH}_4$  to areas where seasonal deepening of the surface ocean mixed layer depth leads to ventilation of these water masses. Continued strengthening of the MC will further increase heat transfer to the BKS, with the Barents Sea ice-free in ~15 years. We thus expect marine  $\text{CH}_4$  flux to the atmosphere from this region to continue increasing.

**Keywords:** Arctic, methane, sea surface temperature, ice, Barents and Kara Seas, warming, currents, emissions

## Highlights:

- Warm, eastwards-flowing Murman Coastal Current penetrates further into the Barents and Kara Seas
- Currents transport heat, driving increasing methane emissions
- Ocean current shoaling enhances shallow methane transport and allows deep methane transport to the atmosphere.
- Franz Josef Land and the west coast of Novaya Zemlya are important, unaccounted and growing  $\text{CH}_4$  sources
- Thermogenic methane from hydrates and submerged permafrost likely play an important role.





## 36 1. Introduction

### 37 1.1 Changes in the Arctic Environment in the Anthropocene

38 The Arctic has experienced the fastest warming on Earth over recent decades, termed Arctic amplification  
39 (Manabe and Stouffer, 1980) with the Arctic Ocean warming at nearly double the rate of the rest of the world's  
40 oceans (Hoegh-Guldberg and Bruno, 2010). Arctic amplification is strongly evident in the Arctic sea-ice reductions  
41 associated with increasing sea surface temperature (SST) (Comiso, 2012; Comiso et al., 2008; Graversen et al., 2008;  
42 Hoegh-Guldberg and Bruno, 2010; Overland and Wang, 2013; Screen and Simmonds, 2010; Stroeve et al., 2014).

43 Multiple positive feedbacks underlie Arctic amplification; such as decreased sea-ice cover increasing solar  
44 insolation absorption, thereby decreasing sea ice further, which also increases humidity and thus downwelling  
45 infrared radiation (Screen and Simmonds, 2010). These feedbacks can be complex. Poleward humidity transport has  
46 been identified as leading to greater downwelling longwave radiation and resultant ice loss in the Pacific Arctic,  
47 which increased humidity and downwelling longwave radiation (Lee et al., 2017). The progression of warmer water  
48 into the Barents Sea drives local winds that decrease wind-advection of sea ice, with decreased sea-ice *increasing*  
49 heat loss by cooling from the atmosphere (Lien et al., 2017). The progressive decrease in Arctic-ice extent underlies  
50 numerous oceanic physical (NRC, 2014) and ecosystem feedbacks (Alexander et al., 2018). Retreating ice affects  
51 heat transfer, light availability in the water column, momentum transfer (convective and wind mixing), and ocean  
52 heat and moisture exchange with the atmosphere. Thus, beyond albedo, sea-ice changes affect weather (NRC, 2014).  
53 Data since 1948 show that Arctic Ocean and atmospheric temperatures and storm frequency increased as sea-ice  
54 extent and volume decreased (NRC, 2014).

55 Arctic amplification has implications for seabed methane (CH<sub>4</sub>) emissions – particularly that which currently is  
56 “sequestered” beneath subsea permafrost – terrestrial permafrost inundated by rising sea level after the Holocene.  
57 For example, extensive seabed CH<sub>4</sub> seepage is linked closely with destabilization of subsea permafrost in the East  
58 Siberian Sea (Shakhova et al., 2013) with emissions estimated as comparable to those from Arctic Tundra  
59 (Shakhova et al., 2015). Warmer seabed temperatures degrade subsea permafrost integrity (Shakhova et al., 2017),  
60 enhancing emissions (Shakhova et al., 2015); however, timescales remain uncertain. Subsea permafrost is likely  
61 extensive in the Kara Sea, and possibly southeast Barents Sea (Osterkamp, 2010). Another feedback occurs from sea  
62 ice reduction, which increases CH<sub>4</sub> flux to the atmosphere by no longer impeding gas transfer.

63

64 FIGURE 1 HERE – *Arctic methane map*

65

66 The marine Arctic is affected by changes in the terrestrial Arctic, both climate and fresh water and organic  
67 material inputs from rivers. Arctic soils contain 50% of the global subterranean carbon pool of which 88% is  
68 estimated sequestered in permafrost (Tarnocai et al., 2009). A quarter of this 1670 Pg (1 Pg=10<sup>15</sup>g) carbon pool may  
69 be mobilized into the Arctic Ocean and sub-marginal seas over the next century due to Arctic warming (Gruber et  
70 al., 2004). The Arctic and sub-Arctic show strong terrestrial, high latitude, positive CH<sub>4</sub> anomalies for eastern  
71 Canada, Alaska, and Western Russia (Fig. 1). Still, the Barents Sea, where the most rapid winter ice loss has





72 occurred and which likely will be the first year-round ice-free arctic sea (Onarheim and Årthun, 2017) and the Kara  
73 Sea, show the strongest CH<sub>4</sub> anomalies by far.

## 74 1.2. Study Motivation

75 We hypothesize that increases in water column temperature drive subsea permafrost and hydrate destabilization  
76 that result in seabed CH<sub>4</sub> emissions, which manifests as increases in lower tropospheric CH<sub>4</sub> over the Barents and  
77 Kara Seas. The relationship between seabed CH<sub>4</sub> and atmospheric CH<sub>4</sub> is indirect – seabed CH<sub>4</sub> must be transported  
78 through the water column on timescales faster than microbial oxidation timescales. CH<sub>4</sub> transport is by bubbles,  
79 diffusion, vertical mixing. We propose that current heat transport is driving increasing seabed emissions. Currents  
80 are the major contributor of oceanic heat to the Barents Sea on annual (Lien et al., 2013) and seasonal time-scales  
81 (Lien et al., 2017) and towards sea ice loss (Årthun et al., 2012).

82 Our study investigates this hypothesis using *SST* and lower atmospheric CH<sub>4</sub> for which satellite data 2003-2015  
83 were analyzed for statistically significant trends in the Barents and Kara Seas relative to the Basin trends. Seabed  
84 temperatures cannot be observed by satellite, thus we test our hypothesis using *SST* as a proxy, albeit one affected  
85 both by currents and other processes including meteorology and solar insolation/long-wave downwelling radiation  
86 (i.e., cloudiness and GHGs including CH<sub>4</sub>). Still, *SST* variations from these atmospheric and radiative processes only  
87 weakly affect the seabed due to stratification.

88 Satellite data are key as they allow repeat observations of multiple variables on synoptic spatial scales, thus we  
89 analyze satellite-derived-*SST*, CH<sub>4</sub>, and sea ice extent in cloud free pixels. Trends are analyzed with respect to data  
90 on currents and winds to understand how the spatial and temporal distribution of tropospheric CH<sub>4</sub> above the  
91 Barents and Kara Seas relates to heat transport into these seas. Specific focus is on areas that become ice-free  
92 seasonally and inter-annually. The analysis focuses on localized anomalies and trends (tens to hundreds of  
93 kilometers), relative to the overall Barents Sea and thus de-emphasizes processes such as poleward atmospheric  
94 moisture transport that affect *SST* on regional scales.

96 FIGURE 2 HERE – *Map of area and ice extent in January and September*

98 The approach's potential was revealed in a scoping study of a small area (**Supp. Table S1, Box A2**) in the  
99 marginal ice zone where Barents Sea water flows into the St. Anna Trough between Franz Josef Land and Novaya  
100 Zemlya (**Fig. 2b, star**). For these pixels, satellite *SST* and CH<sub>4</sub> (0-4 km) were correlated (CH<sub>4</sub> increased with  
101 increasing *SST*) for one of two pixel populations (**Fig. 3, Blue oval**). Given that the oceanography, including ice,  
102 varies dramatically across the Barents and Kara Seas we test our hypothesis on ten focus areas to elucidate how  
103 trends differ across the Barents Sea. Focus areas were large enough to decrease noise by pixel aggregation while  
104 small enough to avoid reducing trends by spatial averaging.

106 FIGURE 3 HERE – *SST vs CH<sub>4</sub> for scoping area*





### 108 1.3 Sea Surface Temperature (SST)

109 SST is the ocean skin layer temperature and depends on the balance between downwelling and upwelling  
110 radiation (modified by clouds and aerosols), and heat transfer between the underlying ocean and overlying  
111 atmosphere and evaporative cooling (Frankignoul, 1985). The upper layer of the ocean is well mixed (the WML) by  
112 turbulence and windwaves. Thus, persistent (multiple days) SST anomalies generally reflect (to some degree)  
113 temperature anomalies of the WML but not water below the WML, which is insulated by stratification. Screen and  
114 Simmonds (2010) found that the strongest Arctic warming was in the near-surface atmospheric layer and was most  
115 strongly related to the retreat of sea ice. Scales are important – atmospheric heat transport in the marine atmosphere  
116 tends to be on large spatial scales, whereas currents affect SST on smaller scales both near the seabed and near the  
117 sea surface.

118 Solar insolation changes immediately affect SST, primarily from cloud cover changes. This effect is removed on  
119 daily timescales by cloud filtering. On longer timescales, changes in persistent cloudiness can cumulatively alter  
120 upper ocean temperatures (and SST). Increased cloudiness decreases incoming short wavelength radiation (cooling)  
121 while increasing long-wave radiation (warming) (Lee et al., 2017). However, these two effects largely counter each  
122 other with the balance further compensated by humidity and temperature profile changes (Schweiger et al., 2008).  
123 Given the canceling effects of persistent cloudiness and that significant changes in cloudiness are not observed  
124 outside areas of sea ice retreats, Screen and Simmonds (2010) conclude that “...changes in cloud cover have not  
125 contributed to recent [Arctic] warming.”

126 Currents also create persistent SST anomalies as do winds. During summer, the warm Barents Sea and Kara Sea  
127 currents that flow eastwards and northwards are met by northerly winds that transport cooler air from higher  
128 latitudes (Kolstad, 2008). This remains the case in fall for all of the Barents Sea except coastal Norway and Murman  
129 where winds track the currents and thus amplify warming (Supp. Fig. S3). The transition to north winds occurs  
130 offshore around the area of the Central Bank in the eastern Barents Sea. Fall winds further north are from the north  
131 and oppose current heating.

### 132 1.4 Global and Arctic Atmospheric Methane

133 Since pre-industrial times, emissions of the potent greenhouse gas, CH<sub>4</sub>, have risen by a factor of 2.5  
134 (Dlugokencky et al., 2011), with increases resuming after near stabilization in the late 1990s (Nisbet et al., 2014).  
135 On a 20-year timescale, CH<sub>4</sub> has greater radiative impact than carbon dioxide (IPCC, 2007; Fig. 2.21). Several  
136 processes may explain this trend including increasing emissions from the Arctic, wetlands, and fossil fuel, and/or  
137 decreasing losses from hydroxyl radical (OH) (Ghosh et al., 2015; John et al., 2012; Nisbet et al., 2014; Turner et  
138 al., 2016), which are proposed to compensate for decreasing biomass burning (Desjardins et al., 2018). OH  
139 concentration decreases with increasing latitude (Liang et al., 2017), enhancing Arctic winter CH<sub>4</sub> lifetime relative  
140 to lower latitudes. Arctic OH varies seasonally, imposing an ~10 ppb seasonality in CH<sub>4</sub> concentrations (Thonat et  
141 al., 2017).





142 Current CH<sub>4</sub> inventories have high uncertainty, with projections being even more uncertain (IPCC, 2013;  
143 Prather et al., 2012; Saunio et al., 2016). Arctic emissions contribute strongly to future CH<sub>4</sub> budget uncertainty due  
144 to Arctic amplification (Graversen et al., 2008) and uncertainty in the timescales of the release of the vast Arctic  
145 CH<sub>4</sub> deposits trapped as gas, CH<sub>4</sub> hydrates, and organic material under permafrost and sediments both onshore  
146 (Tarnocai et al., 2009) and offshore (Archer et al., 2009). For example, rapid warming of the shallow East Siberian  
147 Arctic marginal Sea has degraded submerged permafrost integrity, releasing sequestered CH<sub>4</sub> (Shakhova et al.,  
148 2017). Global CH<sub>4</sub> concentrations increase poleward and are highest in the Arctic (Xiong et al., 2016), driven in part  
149 by strong CH<sub>4</sub> sources, including seabed emissions, terrestrial riverine runoff (Shakhova et al., 2013), and Arctic  
150 terrestrial sources (industrial, permafrost, wetlands, fires, etc.).

151 Arctic seabed CH<sub>4</sub> sources include thermogenic (geological) seepage (Shakhova et al., 2013), biogenic CH<sub>4</sub>  
152 production (James et al., 2016; Reeburgh, 2007) and submerged permafrost, originally from biogenic and/or  
153 thermogenic sources (Shakhova et al., 2013). Seabed emissions largely are bubbles or dissolved gas; however,  
154 microbial oxidation in near seabed sediments (the microbial filter) limits the importance of dissolved seabed CH<sub>4</sub>  
155 fluxes (Reeburgh, 2007). Bubble seepage transports CH<sub>4</sub> directly through the water column and potentially to the  
156 sea surface after losses through dissolution. Bubble seepage also indirectly transports fluid with dissolved CH<sub>4</sub>  
157 (Leifer and Patro, 2002). The fate of dissolved seep CH<sub>4</sub> depends strongly on its dissolution depth (Leifer and Patro,  
158 2002) with microbial oxidation expected to remove dissolved CH<sub>4</sub> below the Winter Wave Mixed Layer (WWML)  
159 (Rehder et al., 1999), whereas dissolved CH<sub>4</sub> in the WWML mostly escapes to the atmosphere. Microbial oxidation  
160 timescales are weeks in plumes to decades at ambient deepsea concentrations (Reeburgh, 2007). The fraction of seep  
161 CH<sub>4</sub> that dissolves below versus within the WWML depends strongly on seabed depth, volume flux (Leifer and  
162 Patro, 2002), plume synergies that include the upwelling flow (Leifer et al., 2009) and bubble surface properties  
163 including the presence of surface impurities (Leifer and Patro, 2002). Frequent Arctic storms deepen the WWML  
164 significantly and efficiently sparge dissolved CH<sub>4</sub> to the atmosphere (Shakhova et al., 2013). Field studies and  
165 numerical modeling show that bubbles can transport some of the seabed CH<sub>4</sub> to the upper water-column and  
166 potentially sea surface even for deep sea seepage (to ~1 km) due to plume and deep sea bubble processes  
167 (MacDonald, 2011; Rehder et al., 2009; Solomon et al., 2009; Warzinski et al., 2014).

## 168 1.5. Airborne and Satellite Observations of Arctic Tropospheric Methane

169 Although the Arctic covers a vast territory, our knowledge of Arctic processes is highly limited both in spatial  
170 and seasonal coverage. This is due to high cost, logistical challenges, and the harshness of Arctic weather. Satellite  
171 Arctic observations (since 1979) have advantages for Arctic observations including quick revisit times and global  
172 coverage (Leifer et al., 2012) and can fill the significant existing temporal and spatial gaps between the few airborne  
173 and field datasets.

174 Several airborne campaigns have measured Arctic atmospheric CH<sub>4</sub> since 2005: HIAPER Pole-to-Pole  
175 Observations (HIPPO), which was ocean-focused (Kort et al., 2012; Wofsy, 2011), Carbon in Arctic Reservoirs  
176 Vulnerability Experiment (CARVE), which was Alaska focused (Chang et al., 2014) and the Atmospheric Radiation  
177 Measurements V (ARM-ACME) on the Alaskan North Slope during summer and off Spitsbergen by the Facility for





Airborne Atmospheric Measurements (FAAM) for summer marine data (Myhre et al., 2016). Given the highly extensive spatial scales of the Arctic, these campaigns provide only a few summer snapshots of a highly variable domain. A review is presented in Supplemental Materials.

Satellite observations provide long-term temporal context for airborne campaign data, which are limited in time and often spatially, and often to the summer season when weather is acceptable. Remote sensing measures column gas abundance and thus is independent of potential mismatches between the platform altitude and the altitude of enhanced CH<sub>4</sub>. Airplanes may not fly sufficiently low to collect data in the Arctic marine planetary boundary layer (PBL), which often is shallow (Aliabadi et al., 2016).

Satellite CH<sub>4</sub> remote sensing uses spectral features at 1.67 and 2.32  $\mu\text{m}$  in the Short Wave InfraRed (SWIR) (Clark et al., 2009) and around 7.82  $\mu\text{m}$  in the thermal infrared (TIR) (Tratt et al., 2014). CH<sub>4</sub> retrievals for SWIR sensors, which use passive reflective solar radiance, are challenged in the Arctic by high cloud cover, low solar zenith angle, and low reflectivity for ice, snow, and water (Leifer et al. 2013). TIR sensors have significant advantages over SWIR sensors for Arctic marine CH<sub>4</sub> (Leifer et al. 2013). TIR sensors measure upwelling surface emitted radiation, which thus has comparatively shorter path lengths at high latitudes relative to SWIR sensors that measure reflected sunlight. TIR sensors can retrieve CH<sub>4</sub> above low clouds, both day and night, whereas SWIR can only observe during daytime and is extremely difficult in the presence of clouds. Additionally, SWIR sensors are insensitive to altitude. Thus, given that most of the CH<sub>4</sub> column abundance lies close to the surface, SWIR sensors respond strongest to the near-surface atmosphere, whereas TIR retrievals have higher sensitivity to mid-tropospheric CH<sub>4</sub> than to near-surface CH<sub>4</sub> (Xiong et al., 2013). Details on the (InfraRed Atmospheric Sounder Interferometer) (IASI) and Atmospheric InfraRed Sounder (AIRS) TIR instruments and validation are provided in **Supplemental Section S2**.

Based on IASI, Yurganov et al. (2016) found low atmospheric CH<sub>4</sub> anomalies in summer for 2010-2015 with annual Arctic Ocean CH<sub>4</sub> fluxes estimated at  $\sim 2/3$  terrestrial Arctic fluxes (to the north from 60° N). Positive CH<sub>4</sub> anomalies were observed along the coasts of Norway and the Novaya Zemlya and Svalbard archipelagos primarily November-January (Yurganov and Leifer, 2016a). A breakdown of the Arctic oceanic summer thermal stratification by wind-induced mixing in autumn may underlie this seasonal trend. Such an approach has been proposed for the North Sea due to a breakdown of stratification in the summer and fall (Leifer et al., 2015). Additionally, Yurganov and Leifer (2016b) report significant CH<sub>4</sub> increases during the 2015/2016 winter over the Sea of Okhotsk.

## 2. Method and Study Design

### 2.1. Overview

In this study, we characterize several processes by satellite observations aggregated on a monthly basis. Satellite data allow repeat regional observations spanning many years. Specifically, we investigated the relationship between ice-free months, sea surface temperature (*SST*), and the lower tropospheric CH<sub>4</sub> column. We concentrate on five focus areas affected by: (1) Arctic water; (2) combined Arctic and Norwegian Atlantic Current; (3) Barents Sea Polar Front; (4) Murman Current; and (5) the Murman Coastal Current and Novaya Zemlya Current.





Specifically, satellite products for the Barents and Kara Seas are quality reviewed and then analyzed to identify statistically significant trends on both a pixel basis and in focus areas relative to regional trends (**Section 2.2**). The analysis uses relative trends to reduce potential retrieval biases and uncertainty. The use of focus areas allows pixel aggregation to reduce the impact of a highly spatially heterogeneous signal and to reduce the effect of inter-annual spatial shifts, which could appear as local temporal variations.

The analysis investigates these trends in relationship to oceanography and meteorology and data for the Barents and Kara Seas relevant to heat transport to, within, and between the Barents and Kara Seas (**Section 2.3**). This analysis investigates the importance of different processes to improve our understanding of the fate of seabed CH<sub>4</sub> emissions.

## 2.2. Methodology

### 2.2.1 Satellite data

AIRS CH<sub>4</sub> data (version 6) are publicly available from NASA Goddard Space Flight Center (GSFC) since 2002 (AIRS Science Team/Joao Texeira, 2016). CH<sub>4</sub> data for 2003–2015 are retrieved by the NOAA Unique Combined Atmospheric Processing System (NUCAPS) algorithm, developed at NOAA/NESDIS in cooperation with Goddard Space Flight Center (GSFC). Data are analyzed for open ocean areas with high vertical thermal contrast, defined here as the temperature difference between the surface and altitude of 4 km (Yurganov and Leifer, 2016a; Yurganov et al., 2016). CH<sub>4</sub> data are re-projected to a 4-km azimuthal equal area projection. The CH<sub>4</sub> anomaly (CH<sub>4</sub><sup>'</sup>) is calculated by subtraction of the values computed within each of the 10 focus areas from the average of the whole Barents Sea for each year. As CH<sub>4</sub> shows high inter-annual variability, a three-year running average is applied. CH<sub>4</sub> retrievals are accurate over both ice and seawater.

Ocean SST are from the Moderate Resolution Imaging Spectroradiometer (MODIS) sensor on the Aqua satellite (NASA, 2015), obtained from the GSFC, Ocean Ecology Laboratory, Ocean Biology Processing Group (OEL-OBPG). The 4-km, Level 3 data are re-projected to a 4-km, equal azimuthal area projection. Satellite data products are cloud screened (Ackerman et al., 2010). The mapped products match the CH<sub>4</sub> data projection. Cloud filtering removes pixels with partial cloud coverage, which would change SST, from the dataset.

First, data are quality reviewed for sea ice coverage and cloud coverage filtered for coastlines, which are from the Global Self-consistent, Hierarchical, High-resolution Shoreline database (SEADAS, 2017). Shape files of sea-ice monthly extent are obtained from National Snow and Ice Data Center (Fetterer et al., 2017) and are based on monthly passive microwave radiometry with the Bootstrap algorithm (Comiso et al., 2008). Sea-ice fields are provided on a polar stereographic grid at 25-km resolution. The number of ice-free months is derived from the intersection of the monthly ice shape file for each year with the focus areas. The number of ice-free months each year is tallied by the following rules: if the intersection is less than 15%, it is counted as 0 months; if coverage is greater than 15% and less than 50% of the pixel, it is counted as 0.5 months. When coverage is greater than 50% in a single month the pixel is counted as ice covered for the month. Ice-covered (>50%) pixels are not used in the SST trend analysis and mean values.





## 2.2.2 Trend analysis

To estimate trends in the Barents Sea and adjacent areas, the monthly mean time series for each grid point in the images covering this region are calculated. Then, a first-order polynomial is calculated by linear regression analysis. Linear trends are analyzed using the Mann Kendall Test (Önöz and Bayazit, 2003) and Sen's linear trend analysis (Juahir et al., 2010; Sen, 1968).

Visual analysis of the trends and anomaly maps of the Barents Sea were used to determine the focus areas' locations. Trends for focus areas were calculated by averaging all valid (cloud cleared) pixels in each focus area for the same month for each year.

## 2.2.3 Focus Areas

The ten focus areas (**Fig. 4a**; **Table 1** for locations) are grouped into 5 oceanographic types. The north easterly focus areas (A1-A3) characterize the inflow of Arctic surface water through both gaps between the archipelagos of Svalbard and Franz Josef Land and between Franz Josef Land and Novaya Zemlya. Each of these focus areas exhibits different seasonal ice coverage. Another group of focus areas is west of Spitsbergen (A4-A6) and is influenced by the West Spitsbergen Current and water from the Barents Sea. A focus area near Bear Island (A7) is affected by the warm, north-flowing NAC and the cold, southwest-flowing Bear Island Current (BIC) and thereby the closest to the Barents Sea Polar Front region (Harris et al., 1998). Three focus areas (A8, A9, and A10) were selected that are influenced by the Murman Current and MCC with focus area (A9) situated in coastal waters offshore southwest Novaya Zemlya with strongly, seasonally-varying ice coverage.

For the analysis, three sets of focus areas are chosen, "Northwest of Barents" including the Greenland Sea and Fram Strait, west of Spitsbergen (A4-A6), "Northern Barents" in the marginal ice zone at the edge of the Arctic Basin (A1-A3) and "Southern Barents," which is strongly influenced by heat from the east fork of the NAC (A7-A10). Of these, A8 and A10 cover banks and A7 covers a shelf, near Svalbard Bank. These groupings are based on physical oceanography and exhibited similar trend patterns.

## 3.0 Results

### 3.1. Setting Overview

#### 3.1.1. Barents Sea Oceanography and Meteorology

Currents are very important to Barents Sea oceanography (**Fig. 4**), dominated by inflow of warmer North Atlantic water through the North Atlantic Current (NAC), which forks into outflows along western Svalbard and through the Saint Anna Trough into the Arctic Ocean (Loeng et al., 1997). Cold Arctic water also flows into the Barents Sea through the Saint Anna Trough as the Percey Current (also Percey Current, PC). River inputs and flows between the Barents and Kara Seas are also important.

The relatively shallow (230-m average depth) Barents Sea is an adjacent sea to the Arctic Basin with complex bathymetry and hydrography (Loeng, 1991). The Barents Sea is bounded to the south by northern Europe and to the north by two archipelagos, Svalbard and Franz Josef Land (FJL). To the east lies the large north-south oriented,





282 Novaya Zemlya archipelago, beyond which is the Kara Sea; to the west lies the Norwegian Sea. In winter the  
283 Barents Sea is partially ice-covered, while it is almost ice-free in the summer (**Fig. 4b**).

284

285 *FIGURE 4 HERE – Map of currents and focus areas in the Barents Sea*

286

287 The North Atlantic is a significant source of Arctic Basin water, whose density increases by cooling. Some of  
288 this water flows into the Barents Sea,  $\sim 2$  Sv ( $1 \text{ Sv} = 10^6 \text{ m}^3 \text{ s}^{-1}$ ), varying seasonally (Loeng et al., 1997) with most  
289 returning to the North Atlantic as part of the global thermohaline circulation (Aagaard and Carmack, 1989; Carmack  
290 and McLaughlin, 2011; Yamamoto-Kawai et al., 2008). McClimans and Nilsen (1993) used a laboratory simulation  
291 to duplicate most of the observed regional Barents Sea oceanographic features forced by the densities and volume  
292 fluxes of water from the Atlantic (the Norwegian Atlantic Current - NAC and the Norwegian Coastal Current -  
293 NCC) and Arctic Basin (Persey Current - PC) and Barents Sea hydrography. Key features were the general structure  
294 of fronts and major currents, etc., which were obtained without regional atmospheric forcing. This highlights the  
295 dominant importance of oceanography rather than meteorology to these features.

296 North Atlantic water flows through the Norwegian Sea, forming the NAC, one track of which becomes the  
297 West Spitsbergen Current (WSC), with the remainder flowing into the Barents Sea through the Barents Sea Opening  
298 as the North Cape Current (Piechura and Walczowski, 2009). The North Cape Current bifurcates into several forks  
299 mostly flowing to the east along the southern slope of the Barents Sea becoming the Murman Current (MC) near  
300 Murman.

301 The NAC is the major contributor of oceanic heat to the Barents Sea (Lien et al., 2017). Regional winds  
302 modulate the volume flow of Atlantic water into the Barents Sea—stronger in winter and weaker in summer (Stiansen  
303 et al., 2009; Fig. 2.3.4). Ice processes further complicate heat re-distribution for surface Arctic Ocean waters – ice  
304 insulates (better preserving its heat) the water from atmospheric radiative cooling. For example, the NAC's western  
305 fork (the WSC) submerges north of Spitsbergen (location varying seasonally) under an isolating layer of colder and  
306 fresher water furthering heat transport into the Arctic (Lien et al., 2017; Lien et al., 2013).

307 A south fork of the NAC is entrained into the NCC, which is 90% Atlantic water and 10% river discharge  
308 (Skagseth et al., 2008). The NCC is a major contributor of oceanic heat to much of the southern and eastern Barents  
309 Sea and into the Kara Sea (Lien et al., 2013). The NCC cools significantly through interaction with the atmosphere.  
310 Upon entering Russian waters, the NCC is renamed the Murman Coastal Current (MCC). Long-term (1905-)  
311 temperature data for the upper 200 m are available from a section off the Kola Peninsula (**Fig. 4a, Kola Section,**  
312 **black dashed line**), which the MCC crosses (Boitsov et al., 2012). These data reveal long-term trends with a cooler  
313 period from 1875-1930 and continuous warming of  $\sim 0.8^\circ\text{C}$  since a minimum in 1970-1980 (Skagseth et al., 2008).  
314 The Kola Section data (which is full water column) show good gross agreement with long-term (since 1850) Barents  
315 Sea ice-extent (Walsh et al., 2016) – the warm period from 1930-1965 corresponds to a significant reduction of  
316 spring sea-ice (from  $\sim 0.2$  to  $\sim 0.12$ ). The Kola Section data shows steady warming since 1970 that corresponds to a  
317 consistent general sea-ice extent decrease since 1980 in spring and since 1970 in fall. This highlights that important  
318 long timescale forcing sed by the MCand MCC affects sea ice extent, meteorology, and oceanography in the  
319 southern and eastern Barents Sea.





320 Although beyond this study's scope, changes in the NAC/MC flow through the Kola Section relate to larger  
321 oceanographic trends. Skagseth et al. (2008) found good agreement in the Kola Section temperature trend with the  
322 Atlantic Multi-decadal Oscillation (AMO) index. SST lags atmospheric temperatures by 2-3 months, peaking for the  
323 Kola area (offshore Murman, Russia) between 0 and 200 m in September-October, whereas air temperature peaks in  
324 July (Stiansen et al., 2009, Figs. 2.3.3, 2.3.8).

325 The MCC continues eastward along the northern edge of the White Sea, becoming the Novaya Zemlya Current  
326 (NZC) until diverted northwards by Novaya Zemlya. It continues into the Arctic Basin through the Saint Anna  
327 Trough (SAT) between Franz Josef Land and Novaya Zemlya (Loeng, 1991), which is the dominant outflow of the  
328 Barents Sea (Maslowski et al., 2004). A fork of the MCC flows eastward into the Kara Sea through the narrow and  
329 shallow (20-50 m) Kara Strait (**Supp. Fig. S1** shows detailed Kara Sea currents).

330 A fork of the North Cape Current flows north through the Bear Island Channel towards the Hopen Deep (Loeng  
331 et al., 1997), underneath the cold, south-flowing Bear Island Current (BIC). Whitehead and Salzig (2001) suggested  
332 (and demonstrated in the laboratory) that remote forcing of the NAC through the Barents Sea lifts the current by  
333 several hundred meters to the sill of the Bear Island Channel, forcing significant anticyclonic vorticity. This drives  
334 the retrograde Bear Island Channel Current (BICC, our connotation) northeast along the slope of Svalbard Bank and  
335 the prograde Murman Current (MC) along the slope of Tromsøflaket, eastward and north to the east of the Central  
336 and Great Banks (Li and McClimans, 1998; Loeng, 1991). Li and McClimans (1998) referred to the BICC as the  
337 "Warm Core Jet" to emphasize its physical significance at the Polar Front. These merge east of the Central and  
338 Great Banks. The resulting flow cools from contact with the atmosphere into a denser, modified Atlantic Water flow  
339 that exits through the Saint Anna Trough to the east of Franz Joseph Land (Gammelsrød et al., 2009). Cooling at  
340 these banks also produces a dense westward underflow, depicted by the dashed line in **Fig. 4a**.

341 The Percey Current (sometimes spelled Persey, PC), transports cold, low saline, Arctic surface water into the  
342 Barents Sea to the east of Spitsbergen, becoming the Bear Island Current (BIC) to the west of the Grand Bank  
343 (**Supp. Fig. S2**). The Percey Current meets warmer, higher salinity waters of Atlantic origin in the Barents Sea,  
344 giving rise to the Barents Sea Polar Front (Oziel et al., 2016), whose location is controlled by seabed bathymetry,  
345 i.e., it is semi-stationary (Gawarkiewicz and Plueddemann, 1995). This front is part of a unique frontal system due to  
346 its combination with the seasonally ice-covered zones in the northern, central, and eastern Barents Sea (Vinje and  
347 Kvambekk, 1991). Part of the Percey Current merges with the East Spitsbergen Current (ESC) to the west of the  
348 Svalbard Bank and then flows north along the west Spitsbergen coast, inshore of the WSC, as the Spitsbergen  
349 Coastal Current (SCC). This flow loops the Barents Sea Polar Front around Spitsbergen (Svendsen et al., 2002).

350 Stratification plays an important role in the Barents Sea energy budget. Barents Sea water-column structure is  
351 modulated by winter cooling of surface waters and their convective mixing as well as brine rejection of seawater  
352 during ice formation. Winter vertical mixing extends to the seabed or near to the seabed over large portions of the  
353 shallow (200-300 m) Barents Sea (de Boyer Montégut et al., 2004). In spring, the warming of surface waters and  
354 freshwater from melting ice support water column stability and strengthens stratification in the central and southern  
355 Barents Sea (Loeng, 1991). Stratification isolates deeper waters from the atmosphere, preventing heat exchange with





the atmosphere and the vertical mixing that traps dissolved CH<sub>4</sub> in deeper water (Leifer et al., 2015). Coastal waters off Norway and Murman remain stratified year-round due to terrestrial freshwater inputs (Loeng, 1991).

Eastern Barents Sea winds generally circulate counterclockwise (cyclonically), strongly to the north along Novaya Zemlya in winter and weakly to the south in summer and fall (Gammelsrød et al., 2009). This leads to calm winds over the Central Bank in fall and winter and generally weak easterlies near Franz Josef Land (fall to spring). Near Spitsbergen, winds are from the north year-round, weak in summer and strong in winter (Kolstad, 2008; Moore, 2013). The spring wind pattern is similar during winter, albeit displaced southwards and weaker. In summer, moderate winds (6 m s<sup>-1</sup> average) blow from the north over most of the Barents Sea. Fall winds are similar to the summer, but stronger (~8-10 m s<sup>-1</sup>) in the west (near Spitsbergen) and weaker in the east near Novaya Zemlya. Summer south Barents Sea winds are towards the north and later east near coastal Norway and Murman. The Barents Sea is stormy—winds are above 15 m s<sup>-1</sup> for over 125 days per year, mostly from the south (Kolstad, 2008). Thus, in the east Barents Sea, winter winds transport more southerly, potentially warmer air, and in the summer winds from the southwest can transport warmer air along Norway and from the west along the Murman coasts; however, most of the Barents Sea most of the year experiences cold northerly winds. Moreover, much of the winter eastern Barents Sea is ice covered, insulating the sea from the air.

Air temperatures on Bear Island have risen ~1.7°C since 1980 (Boitsov et al., 2012), about triple the global atmospheric trend over the same period of ~0.6°C (<http://eca.knmi.nl/>) and about double the overall Arctic average (Hoegh-Guldberg and Bruno, 2010). For reference, temperatures in Murman have risen far faster at 0.12°C yr<sup>-1</sup> and 0.11°C yr<sup>-1</sup> in June and September 2002-2017, respectively (Supp. Fig. S4). These differences reflect that Bear Island is embedded in the marine rather than the coastal atmosphere and is influenced by the cold Bear Island Current.

### 3.1.2. Kara Sea Oceanography and Meteorology

Kara Sea hydrography is controlled by the freshwater outflow of the Ob and Yenisei Rivers (Fig. 2b; Supp. Fig. S1 for finer details), which contribute 350 and 650 km<sup>3</sup> yr<sup>-1</sup>, respectively (Stedmon et al., 2011), approximately double that of the Mississippi River, primarily (>75%) between May and September. As a result, the eastern Kara Sea is brackish. Riverine sediment leads to the northeast Kara Sea being mostly shallow (< 50 m). The western Kara Sea is deep (mostly >100 m), descending to below 500 m in the Novaya Zemlya Trough (Polyak et al., 2002).

Deeper water in the trough is supplied by inflow of modified Atlantic water from the northern Barents Sea into the deep Novaya Zemlya Trough. Trough water is dense and fully saline. On the surface, inflows to the north Kara Sea come from the MCC, local runoff, and ice supplying the Novaya Zemlya Coastal Current (NZCC), with some flow returning to the Barents Sea through the Kara Strait. Warmer water enters the south Kara Sea from the Barents Sea as the MCC flows through the Kara Strait, joining a northward flowing slope current. Much of this water mixes with the southern flowing NZCC and returns to the Barents Sea through the Kara Strait (McClimans et al., 1999; McClimans et al., 2000, Section 11). River outflow drives the overall Kara Sea surface currents largely northwards. Prevailing winds are mostly from the southwest for the western Kara Sea and from the south to southwest in the





central Kara Sea (Kubryakov et al., 2016). Much of the Kara Sea (particularly north) is ice-covered in July and mostly ice-free by September.

### 3.2. Barents Sea *In situ* Observations

*In situ* CH<sub>4</sub> measurements were made by cavity enhanced absorption spectroscopy (Los Gatos Research Inc., Mountainview, CA). Both transits followed a very similar trajectory (Supp. Fig. S5b) that passed through focus areas A1 and A2. Very large, localized, CH<sub>4</sub> anomalies were observed, including in focus area A2 (Fig. 5; Supp. Fig. S5b shows the full dataset). These anomalies were far offshore, indicative of local (i.e., marine) not distant (i.e., terrestrial) sources. The only reasonable explanation is seep bubble plumes – vessel exhaust was ruled out – see Supp. Sect. S5 for more details.

FIGURE 5 HERE – *In situ* methane data

There was an abrupt decrease in CH<sub>4</sub> around 72°N for the outwards transit, which increased again around 75°N. This depressed CH<sub>4</sub> portion of the transit was near where the vessel left the warm Murman Coastal Current (Supp. Fig. S4b). The strongest anomaly, to 2000 ppb, was observed on the southwards transit where the MCC rises over the sill into the Saint Anna Trough (78.7°N), close to the focus area 8 (Fig. 5).

The two transits were separated by about a month with the September transit higher by ~30 ppb than in August, consistent with strong seasonal CH<sub>4</sub> changes. There were other significant differences. Whereas several narrow (and thus local) CH<sub>4</sub> anomalies were observed during the southwards transit, orders of magnitude more narrow anomalies were observed during the northwards transit. Also, the significant peak at 78.7°N only was observed during the southwards transit, indicating emissions variability.

The difference between these transits highlights the challenges of interpreting such snapshot data, supported by the comparison with IASI pixels that were proximal and within several days (Supp. Fig. S6). Agreement for the northwards transit was reasonably good (generally within 10 ppb), and generally poor for the southwards transit.

### 3.3. Focused Study Area Annual Trends

Focus areas with the strongest decreasing ice cover trends from 2003-2015 are in the marginal ice zone of the northern Barents Sea (south and southwest of Franz Josef Land) at the southern margin of the Arctic Basin (Fig. 6a, A1-A3). Trends for these three study areas are very similar (after classifying 2006 and 2014 for focus area A4 (Spitsbergen Northwest) as outliers. Note, focus areas A1-A3 show below-trend ice-free months in 2014 despite no significant 2014 SST deviation, supporting its classification as an outlier (Fig. 7a).

FIGURE 6 HERE – *focus area ice-free trends*

The similarity in ice coverage trends for area A3 (along the cold Percey Current) with areas A1 and A2 (along the Murman Current's warm, northward leg) suggests not only increasing northward heat transfer, but also





weakening southward cold-water advection. Area A4 (northwest of Spitsbergen) also shows decreasing ice coverage towards more frequent year-round ice-free status and lies at the Arctic Basin boundary (Fig. 6b), albeit more under the influence of warmer NAC waters than those under the influence of the Murman Current in the north-central Barents Sea (A1-A3). The Central Bank of the Barents Sea (Fig. 6c, A10) last saw an ice-covered month in 2005, while a noisy trend of decreasing ice coverage is evident offshore coastal southwest Novaya Zemlya (Fig. 6c, B9), along the western fork of the Murman Coastal Current.

Overall, all focus areas are trending towards year-round ice-free, with the entire Barents Sea likely to be year round ice free by ~2030 based on an extrapolation of trends in Northern Barents Sea focus areas, A1-A3.

FIGURE 7 HERE – *focus area SST trends*

SST increases in all focus areas, albeit at rates spanning a wide range from 0.0018 to 0.15 °C yr<sup>-1</sup> (Fig. 7; Table 1). In the Northern Barents Sea, the strongest warming trend is for area A1, south of Franz Josef Land. This is located in a marginal ice zone in the path of the warm MC. Area A3 shows the weakest warming trend lies along the cold Percey Current. For the Northwest of Barents focus areas (Fig. 7b, A4-A6), the strongest warming is at the northernmost focus area, A4, whereas the weakest trend is for the southernmost focus area (Fig. 7, A6). This is consistent with a strengthened northwards penetration of the warm NAC and thus both the West Spitsbergen Current (WSC) and Bear Island Channel Current (BICC).

The strongest warming trend occurs southwest of Novaya Zemlya (Fig. 7c, A9) along the path of the northerly turn of the MCC, in shallow water. This trend is consistent with increased eastward MCC penetration east along the coast of Novaya Zemlya and into the Kara Sea. A very weak and highly variable SST warming trend is observed to the south of the Svalbard Bank at the intersection of the cold Percey Current with the warm NAC and BICC (A7). Areas A10 and A8, and to a lesser extent A9 all suggest a strong oscillation of ~8 years with peak values in 2005 – 2007, and a minimum around 2010. The same pattern also is observed to the south of Franz Josef Land (areas A1 and A2). All the boxes that exhibit this variability lie along the Murman Current, whose origin is in the NAC.

FIGURE 8 HERE – *Focus area methane trends*

A positive CH<sub>4</sub> trend is observed across the Barents and Kara Seas from June through September with some regions exhibiting far stronger trends than average (Supp. Fig. S7). Areas of faster CH<sub>4</sub> increase include near Franz Josef Land (Fig. 8a, A1, A2), the shallower waters offshore W. Spitsbergen (Fig. 8b, A4), and offshore Novaya Zemlya (Fig. 8c, A9). These areas of increasing CH<sub>4</sub> correspond to areas of consistent warming for 2003-2015 (Fig. 7a, A1, A2) and consistent warming since ~2004/2005 for southwest offshore Novaya Zemlya and the Central Bank of the Barents Sea (Fig. 7c, A8-A10). All these focus areas lie along the northwards flow of the Murman Current and the Murman Coastal Current. The Central Bank also gets heat inflow from the BICC “warm core jet.” Focus area A2 was crossed by the *in situ* transit and found CH<sub>4</sub> anomalies (Fig. 5c) best explained by CH<sub>4</sub> seepage. In





contrast, focus areas along the Percey Current show a slowly decreasing  $\text{CH}_4$  defined as relative to the entire Barents Sea trend (**Fig. 8, A3, A7**), despite an (albeit weakly) increasing *SST*.

The strongest  $\text{CH}_4$  growth is south of Franz Josef Land (**Table 1 A2**,  $3.49 \text{ ppb yr}^{-1}$ ), followed by offshore northwest Spitsbergen (**Table 1 A4**,  $3.37 \text{ ppb yr}^{-1}$  2003-2015,  $3.6 \text{ ppb yr}^{-1}$  2005-2015). This positive trend is sustained over the analysis period. The area off the Fram Strait has natural  $\text{CH}_4$  seepage associated with hydrate destabilization (Westbrook et al., 2008). This is an annual increase, and thus does not result from shifts in the timing of seasonal warming. Note, the  $\text{CH}_4$  slopes for areas A4-A10 all are larger when calculated from the 2005 minimum, but not for A1-A3 (**Table 1**). The former lie along the NAC and its eastern current fork, the Murman Current. Neither the Percey Current focus areas (A3, A7) nor other northern Barents Sea focus areas (A1, A2) show this effect depending on the reference time.

TABLE 1 HERE

The largest variability in *SST* and  $\text{CH}_4$  was in the focus area to the north of Murman in the Murman Current (MC) (**Table 1, A8; Fig. 8**) which could arise from variations in the strength of the MC – Skagseth et al. (2008) shows nearly 50% variability in the volume flux through the Barents Sea Opening flux on decadal time-scales. Additional variability occurs from meteorology (and resultant change in cloudiness and hence solar insolation/downwelling radiation), and shifts in the location of the MC, which bifurcates around the focus area.

In general,  $\text{CH}_4$  was at a low for most of the northwest Barents and southern Barents sites for the period 2004-2006 with an approximately 6-8 year cycle. Boitsov et al. (2012) shows variability in the seabed temperature for 2000-2009 suggesting a period of ~5-7 years, with the coolest seabed temperatures 2002-2005. This suggests a multiyear delay between changes in seabed temperatures and changes in  $\text{CH}_4$  emissions, likely related to timescales for heat transfer through overlying sediments.

### 3.4. Climatology of the Barents and Kara Seas

Atlantic heat input is very important to the energy budget of the Arctic Basin and Barents Sea and is driven by the two forks of the NAC (**Fig. 4a**) (Lien et al., 2013). In addition to meteorological forcing and radiative balance, currents significantly affect *SST*. This importance is evident in the Barents Sea *SST* climatology where warm *SST* follows the path of the warm currents (**Fig. 9; Supp. Fig. S3**). Warmer water flows eastward along the northern Norwegian and Murman coasts and offshore southeast of Spitsbergen along Svalbard Bank and then northward along the western Spitsbergen coast. In June, these flows correspond to “tendrils” of warmer water extending north to the east of the Central Bank and to the west of Novaya Zemlya and around Bear Island (**Supp. Fig. S3a**) and in September in the east Barents Sea (**Supp. Fig. S3b**). Water becomes cooler as it penetrates eastward, and as it reaches the (seasonally-varying) ice edge (**Supp. Fig. S3**). Across much of the Barents Sea there is a strong latitudinal *SST* gradient extending south from the ice edge, independent of the location of the eastern NAC branches. In the coastal waters of Novaya Zemlya, warmer water extends further north than elsewhere. The warm signature disappears in the area where the NAC submerges, near northwestern Novaya Zemlya (**Fig. 4a**).



FIGURE 9 HERE – *Maps of Mean values of SST and CH<sub>4</sub> for BKS*

In June, the edge of the cold (Arctic water) Percey Current/Bear Island Current corresponds well with the warm water's edge and also corresponds fairly well with the median ice edge location. Southeast of Spitsbergen, the Bear Island Current penetrates southward as a narrow extension of cold water ending south of Bear Island. Slightly cooler water is observed over the two banks in the central Barents Sea.

The shift to summer *SST* patterns occurs in July, increasing in August, and then beginning to decrease in September (**Supp. Fig. S6**). For Spitsbergen in the Svalbard archipelago (**Supp. Fig. S2**) the northerly cold Spitsbergen Coastal Current (SCC) inshore of the West Spitsbergen Current (WSC) breaks down. This suggests the SCC is entrained by the more energetic WSC (McClimans, 1994), flowing northwards underneath colder surface winds along southwest Spitsbergen, likely below strong summer stratification. The WSC flows farther offshore in June than in September, i.e., the Barents Front shifts shoreward in summer (**Supp. Fig. S3**).

September *SST* in the shallower eastern (coastal) Barents Sea has warmed to levels comparable to the warmer waters in the southwest Barents Sea where NAC heat input maintains elevated *SST*. Warmer *SST* also extends further offshore Norway and Murman. These seasonal *SST* changes match the sea ice's northwards retreat to Franz Josef Land (**Fig. 9b**) and shift of coastal winds to tailwinds over the currents (**Supp. Fig. S3**). However, Barents Sea warming does not follow the ice edge between Svalbard and Franz Josef Land, corresponding instead to the front of the cold Percey Current. From August to September, the warm water has begun retreating across the Barents Sea with cold water associated with the Percey Current (**Supp. Fig. S7**).

The now mostly ice-free Kara Sea in September exhibits coastal warming, particularly to the east, where there also is heat input from the Ob and Yenisei Rivers (east of the Yamal Peninsula). This area exhibits warming despite partial ice coverage of the Gulf of Ob in June and likely is driven by warmer riverine water inputs.

CH<sub>4</sub> concentrations show a clear latitudinal trend that increases towards the north. This latitudinal gradient is weak in June and strong in September. Strong localized variations also occur in different Barents Sea regions. CH<sub>4</sub> concentrations along the Murman Current and in the (ice-covered) Kara Sea largely are below the latitudinal mean in June, whereas west of Spitsbergen and in the north-central Barents Sea they are above average.

In June, CH<sub>4</sub> is depressed strongly around Svalbard and around Franz Josef Land and Novaya Zemlya. For Spitsbergen, this corresponds to the SCC that hugs the shore. By September, CH<sub>4</sub> concentrations are notably different with significantly higher CH<sub>4</sub> and a distinctly different spatial distribution. Most notable is the shift from depressed to strongly enhanced CH<sub>4</sub> in the region to the west of Novaya Zemlya, particularly the Novaya Zemlya Bank, and around the Franz Josef Land archipelago. Strong CH<sub>4</sub> enhancement also occurs in the outflow plumes of the Ob and Yenisei Rivers in the Kara Sea, around the Taymyr Peninsula. Around Svalbard, CH<sub>4</sub> has risen to near latitudinal mean levels in September, except for offshore north Spitsbergen and Nordaustlandet, where sea ice remains.





### 534 3.5. Barents and Kara Seas Trends

535 Across the Barents Sea, a number of different focus areas with distinct  $SST$  and  $CH_4$  trends are identified (**Fig.**  
 536 **7**). These manifest significant spatial heterogeneity at the pixel scale and at the focus-area size scale. We apply our  
 537 analysis to aggregated-pixel “focus areas” located in key regions where  $SST$  temporal and spatial changes are  
 538 strongest (**Sec. 3.3**; **Supp. Fig. S8** for July and August trends).

539 June  $SST$  warming trends ( $dSST/dt$ ) are fairly different from September  $SST$  trends (**Fig. 10**). In June, warming  
 540 occurs much faster in the eastern Barents Sea, specifically, in waters affected by the Murman Coastal Current  
 541 (MCC). Given that winds are from the north (**Supp. Fig. S3**) current-mediated heat transport opposes current  
 542 warming. This suggests the magnitude of atmospheric cooling during transit from the Atlantic is decreasing.  
 543 Warming occurs primarily in shallow (generally less than 100-m deep) (**Fig. 10b**) waters that are generally well  
 544 mixed. Sea ice is absent in this region by March-May, later in more northerly areas (**Fig. 4b**). Whereas there is no  
 545 clear warming trend in July and August; a strong warming appears in the Kara Sea by September (**Supp. Fig. S8**),  
 546 where winds also are cold northerlies. That this warming occurs several months after the ice retreat suggests that  
 547 insolation is less important after the ice melts – the Kara Sea is ice-free in July (**Supp. Fig. S7**). This is consistent  
 548 with increasing MCC penetration into the Kara Sea. Loeng (1991) reported that MCC penetration into the Kara Sea  
 549 was uncommon in the middle of the 20<sup>th</sup> century.

550

551 *FIGURE 10 – Maps of trend values of SST and CH<sub>4</sub> for BKS*

552

553 More rapid warming occurs offshore of the western coast of Novaya Zemlya from June-September. This is  
 554 where the Murman Current (MC) transports water towards the St Anna Trough (the dominant Barents Sea outflow),  
 555 a region where shoaling is likely based on seabed topography (**Fig. 2b**) (Maslowski et al., 2004). The MC then flows  
 556 (and submerges under ice and Arctic surface water) along the east shores of Franz Josef Land. Accelerated warming  
 557 diminishes near the northern margin of the Kara Sea, where river outflow dominates the oceanography.

558 Enhanced warming also occurs to the south and to the west-northwest of Svalbard in September, following  
 559 approximately the trend of the northerly fork of the NAC. In contrast, waters off east Svalbard, where the East  
 560 Spitsbergen Current (ESC) transports cold Arctic waters southwards, do not exhibit a significant warming trend in  
 561 September, although it does exhibit warming in July. This suggests changes in the seasonal penetration of the PC  
 562 into the Barents Sea, likely modulated by seasonal ice sheet retreat. There is no significant  $SST$  warming in June or  
 563 September to the north of Franz Josef Land with ice-coverage persisting through September.

564 Overall Barents Sea atmospheric  $CH_4$  is increasing (**Fig. 9C**), consistent with the global  $CH_4$  trend (Nisbet et  
 565 al., 2014). However it is notable that some regions exhibit significantly more rapidly increasing  $CH_4$  than the global  
 566 or Barents Sea trends. In June,  $CH_4$  trends are largely similar in both ice-free and ice-covered areas. In near-coastal  
 567 waters around Svalbard (except the east), in northern Norwegian fjords, and for the White Sea (Murmansk) where  
 568  $CH_4$  growth is enhanced.

569 September  $CH_4$  trends ( $dCH_4/dt$ ), when ice coverage has retreated to the northern edge of the Barents and Kara  
 570 Seas (**Fig. 9b**), are strongly enhanced in the east Barents Sea and the south Kara Sea. These areas coincide with





571 areas of enhanced *SST* warming and show  $\text{CH}_4$  trends almost three times as high as the general Arctic trend.  
572 Moreover, they are under northerly winds and thus terrestrial sources cannot contribute (**Supp. Fig. S3**). In contrast,  
573 regions without enhanced warming, particularly waters affected by cold currents, exhibit the weakest  $\text{CH}_4$  growth;  
574 slightly above the rate of overall Barents Sea growth. Also,  $\text{CH}_4$  increases strongly in the Kara Strait between the  
575 Barents and Kara Seas.

576 Enhanced  $\text{CH}_4$  growth is not evident in June or September to the north of Spitsbergen, despite strong *SST*  
577 increases; however, significant increases are evident here in August. This follows significant  $\text{CH}_4$  enhancement in  
578 July to the southeast of Spitsbergen. This July–August shift follows the NAC.

#### 579 4. Discussion

580 In this study, we test the hypothesis that  $\text{CH}_4$  in the lower troposphere correlates with changes in overall water  
581 column temperature, which are reflected in *SST* trends. Specifically, the presence of increasing  $\text{CH}_4$  emissions where  
582 *SST* is increasing implies that the *SST* trend is not a surface skin effect, but increasing water column temperature.  
583 Both *SST* and  $\text{CH}_4$  are satellite remote sensing products

584 The proposed source of the atmospheric  $\text{CH}_4$  anomaly is seabed seepage (**Section 4.2**) from either thermogenic  
585 sources, i.e., petroleum hydrocarbon reservoirs (Judd and Hovland, 2007), or degradation of submerged permafrost  
586 and hydrates (Shakhova et al., 2017). Both permafrost and hydrate deposits can include both thermogenic and  
587 biogenic  $\text{CH}_4$ . Submerged permafrost likely is extensive in the east Kara Sea and potentially in the southeast Barents  
588 Sea (**Fig. 2**) (Osterkamp, 2010). Pockmark fields, typically associated with  $\text{CH}_4$  seepage, have been mapped in the  
589 central northern Barents Sea (Lammers et al., 1995; Solheim and Elverhøi, 1985) and southwest Barents Sea (Rise  
590 et al., 2015) and near Franz Josef Land (Sokolov et al., 2017), including water-column  $\text{CH}_4$  plumes.

591 For this analysis, we also considered the locations of currents and trends in these currents, seabed bathymetry,  
592 prevailing winds, and available Barents Sea, water-column temperature data—primarily the long-term Kola Section  
593 data, which due to the importance of the Murman Current is directly relevant (Boitsov et al., 2012), as well as  
594 meteorology data in coastal Murman (**Supp. Fig. S4**). We test the methane-shoaling hypothesis (**Section 4.3**) – that  
595 currents drive deep methane to shallower waters where it can transfer into the atmosphere distant from its seabed  
596 source.

#### 597 4.1. Seabed-atmosphere methane transport

598 There are a number of mechanisms that allow seabed  $\text{CH}_4$  emissions to reach the sea surface, both due to direct  
599 bubble-mediated transport and by turbulence (from bubble-dissolved  $\text{CH}_4$ ). Transport is bubble-mediated (Judd and  
600 Hovland, 2007), because the microbial filter generally reduces aqueous  $\text{CH}_4$  as it migrates through sediments into  
601 the water column (Reeburgh, 2003). As a bubble rises, it loses  $\text{CH}_4$  to the water column by dissolution, transporting  
602 the remainder. Larger bubbles lose less gas than smaller bubbles – rising higher in the water column (Leifer and  
603 Patro, 2002). In shallow water (e.g., less than 20 m), most seep bubble  $\text{CH}_4$  reaches the sea surface directly, with the  
604 fraction decreasing for smaller bubbles or deeper (Leifer and Patro, 2002). For example, Leifer et al. (2017) showed





that ~25% of seabed  $\text{CH}_4$  from 70 m reaches the Laptev Sea surface directly, consistent with sonar observations of seep bubble plumes reaching the sea surface (Leifer et al., 2017). Some of the dissolved fraction is transported vertically by the bubble-driven upwelling flow (Leifer et al., 2009), even for small plumes (Leifer, 2010).  $\text{CH}_4$  deposited within the WWML diffuses rapidly to the atmosphere, although seasonal stratification powerfully suppresses this transport. Storms breakdown this stratification (Leifer et al., 2015) sparging all the dissolved  $\text{CH}_4$  to the atmosphere (Shakhova et al., 2013). Seasonally, the collapse of the pycnocline from fall storms releases  $\text{CH}_4$  temporarily sequestered in the deep WWML (Nauw et al., 2015). Although some of the dissolved  $\text{CH}_4$  in the WWML will be oxidized microbially turbulence transport in stormy arctic seas is efficient. In practical terms, bubble transport means that seepage extends the effective WWML depth for  $\text{CH}_4$  by 50-100 m, i.e., 150-300 m, allowing wave turbulence and storms to sparge dissolved  $\text{CH}_4$  to the atmosphere over most of the Barents and Kara Seas (**Fig. 2b**).

Below the winter wave mixed layer (WWML), oceanic microbial oxidation timescales are shorter than transport timescales and all dissolved  $\text{CH}_4$  likely is oxidized (Rehder et al., 1999). Still,  $\text{CH}_4$  below the pycnocline may drift with currents that drive it upslope into the WML where it can escape to the atmosphere, possibly distant from its seabed origin - methane shoaling. Even for the worst case,  $\text{CH}_4$  microbial oxidation timescales in plumes are several weeks (Reeburgh, 2007), over which currents can transport  $\text{CH}_4$  order 100-1000 kilometers. Outside of plumes, oxidation timescales are much significantly longer—decades for deepsea background concentrations (Reeburgh, 2007). Methane shoaling also enhances transport of shallower dissolved  $\text{CH}_4$  to the atmosphere.

The above discussion was for non-oily seepage. However, where seepage arises from a petroleum hydrocarbon reservoir, bubbles likely are oily. Oil slows bubble rise (Leifer, 2010) and dramatically reduces dissolution, allowing their survival far higher in the water column than non-oily bubbles (Leifer and MacDonald, 2003). Oily bubbles can reach the sea surface from the deep sea – e.g., MacDonald et al. (2010) tracked seep bubbles by remote operated vehicle from 1 km depth to the WML and found a significant positive  $\text{CH}_4$  anomaly in surface waters. Given the presence of extensive proven and proposed petroleum reservoirs across the Barents and Kara Seas (Rekacewicz, 2005), some Barents Sea seepage is likely oily with enhanced  $\text{CH}_4$  transport to the sea surface. The *in situ* data (**Fig. 5**) showed localized strong atmospheric  $\text{CH}_4$  plumes above deep water that are best explained by oily bubbles. These plumes were above areas of confirmed oil and gas deposits within an extensive region of potential oil and gas deposits in the central and northern Barents Sea (**Supp. Fig. S9**). Thus, the *in situ* data suggest more extensive oil deposits than currently confirmed deposits. Observations of oil slicks would provide confirmation, but require calm winds.

One unlikely source of  $\text{CH}_4$  anomalies for the Barents and Kara Seas is atmospheric transport as there is neither significant local industry nor extensive wetlands/terrestrial permafrost nearby or upwind for the prevailing wind directions. Prevailing winds are from the north in June and September except for south and southeast Barents Sea where winds track the coast and NCC and MCC in September (**Supp. Fig. S3**). Note—synoptic systems can transport  $\text{CH}_4$  from northern Europe or Russia to the Barents Sea, but synoptic system winds are not dominant (prevailing) and thus play a small role in time-averaged datasets. Moreover, these terrestrial sources are distant, implying large size scale anomalies, which would decrease with distance from northern Europe. Instead, the anomalies are localized





and decrease towards Europe, and the *in situ* data (Fig. 5) show highly localized anomalies. The one area where September winds could transport terrestrial CH<sub>4</sub> into the marine atmosphere is from oil production and pipeline infrastructure from the Kanin and Yamal Peninsulas near Kolguyev Island (Supp. Fig. S9). However, extensive CH<sub>4</sub> plumes (Figs. 9 and 10) are not observed in coastal pixels, and dCH<sub>4</sub>/dt trends were not lower than those further offshore. This is consistent with the general trend of decreasing Russian CH<sub>4</sub> emissions from conventional oil production (Höglund-Isaksson, 2017).

#### 4.2. Hydrocarbon Reserves and Local Atmospheric Methane

Seabed seepage, often thermogenic (petroleum hydrocarbon), has been identified in all oceans and all petroleum-producing basins (Judd and Hovland, 2007) and likely plays a role in CH<sub>4</sub> anomalies in the Barents and Kara Seas.

In the Kara Sea, the correlation of enhanced CH<sub>4</sub> with depth is poor, which is shallower to the north. Instead, the location of enhanced September CH<sub>4</sub> closely matches the location of oil and gas reserves, e.g., Supp. Fig. S9; Rekacewicz (2005), and also the Murman Coastal Current's path of warm water as it follows the coastline of the Kanin Peninsula and then enters the Kara Sea. Although there is extensive oil and gas production on the Yamal Peninsula, prevailing winds blow away from the Barents Sea. Note, the trend shows enhanced CH<sub>4</sub> growth, implying increasing emissions, i.e., not steady-state seabed warming but increased seabed warming. This increasing CH<sub>4</sub> growth is for September, not June, corresponding to when the water column is warmest in the South Barents Sea (Stiansen et al., 2009). Also, the Barents Sea outflow through Saint Anna's Trough is greater in September (about double) than June (Gammelsrød et al., 2009) when the growth in the CH<sub>4</sub> anomaly occurs (Fig. 10d). The importance of this transport also is apparent in the SST trend with the greatest warming occurring in June in the southeast Barents Sea (offshore the Kanin Peninsula) near the Kara Strait. This region lies to the west of the areas of enhanced CH<sub>4</sub> growth in September near the Kara Strait. In contrast, significant SST warming is not observed in September in this easternmost region of the Barents Sea.

Two other areas of enhanced CH<sub>4</sub> growth lie in the north-central Barents Sea, north of Central Bank, and offshore northern Novaya Zemlya. These regions lie along the Murman Current and over the Central Bank – a region where the MC and the BICC “warm core jet” converge. Water flowing in this direction also is forced upwards – from 300–400 m to just 100 m as it crosses a sill into the St. Anna Trough with rising seabed towards the east and towards Novaya Zemlya with water depths of just tens of meters (Fig. 2B). Additionally, this region of increasing CH<sub>4</sub> growth corresponds spatially to the potential (i.e., unproven) gas and oil reserves that extend across the Saint Anna Trough to Franz Josef Land, e.g., Supp. Fig. S9; Rekacewicz (2005). There also are proven oil and gas fields to the south, also along the Murman Current's path, but south of the area of increasing CH<sub>4</sub> offshore northwest Novaya Zemlya. These hydrocarbon fields also correlate with increasing CH<sub>4</sub> trends offshore southwest Novaya Zemlya.





#### 675 4.3. Methane Shoaling Hypothesis

676 Where CH<sub>4</sub>-rich currents shoal, they vertically transport dissolved CH<sub>4</sub> into shallow waters where it can diffuse  
677 to the atmosphere. This process, termed *methane shoaling*, allows seabed CH<sub>4</sub> to reach the atmosphere distant from  
678 its seabed source, often beyond the reach of *in situ* studies. In this study, the global and continuous view of satellite  
679 data allowed investigation of the methane-shoaling hypothesis.

680 In general, if seabed CH<sub>4</sub> seepage is sufficiently deep and below the WWML and non-oily, its direct  
681 atmospheric contribution is small to none. Even plume microbial oxidation CH<sub>4</sub> rates of several weeks (Reeburgh,  
682 2007) allows significant horizontal transport by currents – note, timescales increase to decadal for typical deepsea  
683 concentrations (Rehder et al., 1999). Where currents force these waters upslope, CH<sub>4</sub>-rich waters are brought to  
684 shallower depths where mixing allows sea-air transport into the atmosphere. *Shoaling of methane-laden water*  
685 provides the best explanation for the localized, strong and growing, atmospheric CH<sub>4</sub> anomalies in the Barents and  
686 Kara Seas, specifically the Kara Straits and along the Novaya Zemlya coast near Central Bank and elsewhere.

687 Areas of enhanced CH<sub>4</sub> growth were closely related to the path of the Murman Coastal Current as it flows  
688 towards the Kara Strait rather than seabed depth (**Fig. 10**). Both the rising seabed bathymetry and the presence of  
689 both southwards and northwards currents through the Kara Strait imply strong vertical mixing and thus transport to  
690 the atmosphere. Along the path of the Murman Current are significant offshore petroleum hydrocarbon reservoirs  
691 that likely release seep CH<sub>4</sub> into the waters of the Murman Current.

692 Further evidence that transport and methane shoaling is important is from the  $dCH_4/dt$  spatial distribution  
693 around Kolguyev Island (north of the White Sea), which is increasing faster on its western side than its eastern side,  
694 even though the seabed to the island's east is shallower. In fact the CH<sub>4</sub> spatial pattern correlates better with  
695 shadowing in the island's lee from shoaling currents, rather than with seabed depth. Prevailing winds are from the  
696 south-southeast (Kubryakov et al., 2016), thus atmospheric transport cannot explain the pattern.

697 Notably, the enhanced CH<sub>4</sub> concentrations around Franz Josef Land does not correlate with the potential  
698 reserves, but does correlate with depth and the flow of the Murman Current, consistent with methane shoaling.  
699 Although some of the enhanced CH<sub>4</sub> growth near Novaya Zemlya could arise from increasing local seabed  
700 emissions, seabed temperatures were below zero until 2009 (Boitsov et al., 2012), which would imply submerged  
701 hydrate deposits here have not yet degraded significantly, supporting the methane shoaling mechanism.

#### 702 4.4. Sea Surface Temperature

703 The analysis not only shows spatial anomalies that likely result from a combination of local sources - likely  
704 thermogenic seepage – and methane shoaling, but also shows CH<sub>4</sub> anomaly growth ( $dCH_4/dt$ ) that implies  
705 strengthening seabed sources if atmospheric conditions remain constant. Specifically,  $dCH_4/dt$  over *portions* of the  
706 Barents and Kara Seas is faster than the Barents Sea mean and the latitudinal mean. To some level these correlate  
707 with enhanced SST warming, but the correlation is poor. SST is the skin temperature and depends on radiative  
708 balance, atmospheric temperature (including transport and latent heat) and heat transfer from the bulk ocean.  
709 Another factor underlying this poor correlation is that there is a delay between SST warming and ocean-column





warming of several months (Stiansen et al., 2009). There also appears to be a several year response time; the ~6-8 year variability is suggestive of an oscillation in the *SST* trend in the Southern Barents Sea (areas A8, A9, and A10) and has a very similar timescale to the seabed trends reported by Boitsov et al. (2012), albeit preceding it by ~2-4 years.

More rapid *SST* warming occurs offshore Novaya Zemlya moving northwards from June-September, where the Murman Current transports water and the seabed topography is likely to cause shoaling. This suggests that warmer terrestrial weather is not driving Kara Sea changes as this would occur uniformly both in the south Kara Sea, which is influenced by the Barents Sea, and the northern Kara Sea, which is influenced by river outflow. Additionally, if increased riverine heat input were driving the trend, the greatest enhancement would be in the northern Kara Sea, which also is shallower.

There are a number of possible hypotheses for why *SST* is warming fastest in waters along the Murman Current and NAC. One is sea-ice retreat; however, the warming occurs several months after the retreat of the sea ice. Another is that the mixed layer is becoming shallower, allowing more rapid cooling to the atmosphere. This would imply a weakening of storms and winds – which firstly is inconsistent with warmer *SST*, and secondly, there is no indication that Barents Sea storminess is changing or progressing further northwards (Koyama et al., 2017). Cloudiness changes affect *SST*; however, cloud filtering at the pixel level removes this effect from the analysis, whereas persistent cloudiness changes largely cancel outside of areas of sea ice retreat (Schweiger et al., 2008).

Another hypothesis is that increasing ocean current heat transport is driving the *SST* warming. Although *SST* derives from several factors including heat transfer from the bulk ocean (i.e., currents), its co-spatial relationship to enhanced CH<sub>4</sub> anomaly is consistent with currents playing a major role both at the sea surface (*SST* anomaly trend) and at the seabed. This supports using *SST* as a surrogate for water column temperature. Greater heat transport could occur from strengthening currents, or warming currents, or a combination of both.

Seabed September temperatures (Boitsov et al., 2012) do not suggest increased warmer seabed temperatures north of Norway and Russia, but do suggest warmer seabed temperatures to the east and also along Novaya Zemlya – suggesting a greater importance of the MC. This is consistent with the model of McClimans et al. (2000) that currents are advecting ice, shifting the marginal ice zone location. The warming trend suggests a strengthening of the seasonal trend in the Barents Sea outflow, which is greater in September than June (Gammelsrød et al., 2009).

The most rapid warming is for the shallow water off northwest Svalbard (area A4) (**Fig. 10b**), which also exhibited the strongest CH<sub>4</sub> growth. In this area, seabed topography is nearly flat over an extensive shelf with depths in the range 250-400 m. Where the shelf falls off sharply, rising sea temperatures will minimally induce hydrate destabilization. In contrast, where the shelf falls off very gently, small temperature increases shift extensive areas of seabed from below to above the hydrate stability field. This area is immediately to the north of the area where several researchers have identified extensive seabed seep CH<sub>4</sub> emissions (Mau et al., 2017; Myhre et al., 2016; Westbrook et al., 2009). The most likely explanation is a strengthening of the West Spitsbergen Current, discussed below, and shifts in the Barents Sea Polar Front.





#### 745 4.5. Implications for Svalbard Area Methane Emissions

746 There are few atmospheric and ocean CH<sub>4</sub> data for the Barents Sea and surrounding areas, the most prominent  
747 being associated with CH<sub>4</sub> seepage off Spitsbergen, located immediately south of focus area A4. Studies to date  
748 have been in early summer; Mau et al. (2017); Myhre et al. (2016) who made measurements in the atmosphere and  
749 water column while Westbrook et al. (2009) reported sonar observations of seep bubbles for August-September, and  
750 slightly elevated aqueous CH<sub>4</sub> in surface waters immediately above the bubble plumes. All concluded transport to  
751 the atmosphere was not significant, attributed to trapping of dissolved CH<sub>4</sub> below a sharp pycnocline. It is important  
752 to note that with respect to the overall Barents Sea area CH<sub>4</sub> anomaly, the Svalbard area is far less important than  
753 around Franz Josef Land, off the west coast of Novaya Zemlya, and the north-central Barents Sea (**Fig. 9**).

754 Both *SST* and CH<sub>4</sub> in June (**Fig. 9**) and July (**Supp. Fig. S7**) for west Spitsbergen show that much of the area of  
755 active seepage was inshore of the Barents Sea Polar Front, and thus under the cooling Arctic waters of the  
756 Spitsbergen Coastal Current (SCC), supported by reported salinity data (Mau et al., 2017). Although *SST* remains  
757 suppressed off Spitsbergen in September, and extends further offshore, CH<sub>4</sub> concentrations no longer are depressed  
758 compared to Atlantic water further offshore, i.e., greater transport to the atmosphere. Such transport would not be  
759 expected downcurrent (north) of the bubble plumes observed by the early fall cruise reported in Westbrook et al.  
760 (2009).

761 Although the studies indicate these seeps do not contribute to summer atmospheric CH<sub>4</sub>, they did not consider  
762 methane shoaling, which would allow seabed CH<sub>4</sub> to reach the atmosphere far downstream. Interestingly, Mau et al.  
763 (2017; Fig. 3) show data that could be interpreted as methane shoaling with elevated aqueous CH<sub>4</sub> forced shallower  
764 by the north-flowing SCC, rising as it crosses onshore-offshore aligned subterranean ridges. Focus area A4 shows  
765 strong increase in CH<sub>4</sub> from 2005-2015 (the strongest of the focus areas (**Table 1**) and in increasing *SST* over this  
766 time period, consistent with shoaling. Larger enhancement of CH<sub>4</sub> growth is observed north of Spitsbergen in June  
767 (**Fig. 10c**), which is the most likely location for shoaling based on detailed Svalbard bathymetry and currents (**Supp.**  
768 **Fig. S2**). Specifically, this is where some of the warm West Spitsbergen Current mixes with the cold, Spitsbergen  
769 Coastal Current (SCC) that would be CH<sub>4</sub> enriched from seabed seepage, and then flows over relatively shallow  
770 seabed towards the Hinlopen Strait. Thus, there is evidence of increasing downstream CH<sub>4</sub> transport to the  
771 atmosphere downcurrent of seepage off West Spitsbergen after methane shoaling, albeit not significant to overall  
772 Barents Sea emissions.

773 There is evidence of acceleration in the CH<sub>4</sub> growth nearshore off West Spitsbergen in June, but not in  
774 September (**Fig. 11**) when CH<sub>4</sub> growth enhancement lies in the further offshore waters that are impacted by the  
775 warm WSC. Trends in *SST* also suggest a weakening of the Percey Current in June and more so in September. Given  
776 that from June to September the SCC extends further offshore, this suggests WSC control. Similarly, the WSC  
777 eastwards leg that crosses Nordaustlandet is driving a rapid increase in *SST* in September and likely relates to the  
778 increased CH<sub>4</sub> trend.



779 **4.6. Ice-Free Barents Sea**

780 The southern Barents Sea has been ice free since at least 1850 (Walsh et al., 2016). Meanwhile the northwest  
781 Barents Sea is near ice-free year round, whereas northeast Barents Sea (around Franz Josef Land and St. Anna  
782 Trough) remains ice-covered for about half the year (Fig. 6). The ice coverage trends suggest most of the Barents  
783 Sea will be ice free, year-round circa 2030. This is comparable to the 2023-2036 estimate of Onarheim and Årthun  
784 (2017; Fig. 3), which also notes that the current decreasing trend lies outside the oscillation envelope since 1850. Ice  
785 records since 1850 show fairly stable sea ice through 1980 in March (within  $\pm 20\%$ ), and 1970 in September (within  
786  $\pm 50\%$ ), decreasing to date (Walsh et al., 2016). For the Barents Sea, and other marginal Arctic seas most significant  
787 ice loss occurs in late summer (Onarheim et al., 2018).

788 This has implications for the Barents and Kara Seas ecosystems, and follows documented changes across the  
789 Arctic in satellite remote sensing of phytoplankton concentration (Arrigo et al., 2008; Arrigo and van Dijken, 2011;  
790 Kahru et al., 2011) and *in situ* studies (Grebmeier et al., 2006; Kędra et al., 2015). One example is a significant  
791 northwards shift ( $5^\circ$  over 20 yrs.) of phytoplankton blooms (Neukermans et al., 2018). Ice cover changes play a key  
792 role. For example, primary productivity increases in the northern Barents and Kara Seas (Fig. 2) are considered  
793 caused by decreased ice cover (Slagstad et al., 2015), which has driven changes in the higher trophic levels of the  
794 pelagic and benthic community (Kędra et al., 2015).

795 The Barents Sea is a marginal sea between the temperate Norwegian Sea and the Arctic Basin and thus is the  
796 conduit through which lower-latitude oceanic heat is transmitted to the Arctic Basin (Onarheim and Årthun, 2017).  
797 Given the significant role the Barents Sea plays in overall Arctic ice loss - fully 25% of the loss is attributed to the  
798 Barents Sea, which comprises 4% of the Arctic Ocean including marginal seas (Smedsrud et al., 2013), implications  
799 will be significant for weather at lower latitudes, and the marine ecosystem. Seemingly counter-intuitive, sea ice  
800 reduction increases the upwards surface heat flux as ice has an insulating effect. Thus ice-loss somewhat stabilizes  
801 Arctic Basin ice, particularly during winter (Onarheim and Årthun, 2017) and may even lead to growth of ice in the  
802 Arctic Basin and northern Greenland Sea. Still, the data herein are consistent with a progressive weakening of the  
803 Percey Current, which will continue to cause ice loss off east Svalbard and warming of these waters. This agrees  
804 with Alexander et al. (2004) who concluded that the (semi-stationary due to bathymetry) Barents Sea Polar Front  
805 has shifted due to domination of Atlantic over Arctic waters.

806 As noted, the progression of ice loss in the south and east Barents Sea along the pathway of the Murman  
807 Coastal Current has led to a progressive loss of ice in the south Kara Sea. Thus, the balance between the two  
808 processes – heat loss to the atmosphere from and progressive transport of heat by currents to the Kara Sea are clearly  
809 shifting towards warmer. The implications of decreasing ice cover in the shallow Kara Sea are significant with  
810 respect to  $\text{CH}_4$  emissions – the area is rich in hydrocarbon resources that currently are likely sequestered under  
811 submerged permafrost that will continue to degrade, while warming seabed temperatures will enhance microbial  
812 degradation of the vast organic material deposited over the millennia by the Ob and Yenisei Rivers. Thus, the  
813 already significant importance of Arctic  $\text{CH}_4$  anomaly from the Kara Sea will accelerate due to feedbacks from an  
814 ice-free Barents Sea.





#### 815 4.7. Future research

816 Oscillations with a 6-8 year timescale are suggested (e.g., **Fig. 8c**); however, the 13-year dataset is too short to  
817 investigate in detail. Extending the analysis to include more recent data (say through 2018) would span a full 2 1/2  
818 cycles and allow investigation of correlations with other driving oceanographic atmospheric cycles, such as the  
819 NAO. This would be particularly valuable given that recent data show that the most recent two years (2016-2017)  
820 are the most extreme in terms of Barents Sea ice coverage (Oziel et al. 2016) and CH<sub>4</sub> anomaly (**Supp. Fig. S10**;  
821 **Supp. Video**). Extending the analysis forward in time clearly would provide greater insights into the complex  
822 relationship between currents and CH<sub>4</sub> emissions. In this regard, the long-term commitment to IASI satellite  
823 instruments (MetOp-A/IASI-A launched in 2006 and remains in service, MetOp-C/IASI-C is scheduled to launch  
824 Nov 2018) will provide invaluable long-term satellite CH<sub>4</sub> data.

825 Additionally, there is clearly need for these data to be incorporated into coupled atmospheric-oceanographic-ice  
826 models to understand in greater detail the processes underlying the changes, improving the ability to forecast trends  
827 in Arctic marine greenhouse gas emissions. Currently the strong and growing CH<sub>4</sub> anomaly from Novaya Zemlya  
828 and Franz Josef Land are the strongest in the Arctic, yet are not yet incorporated (or identified) in inversion models,  
829 e.g., Crevoisier et al. (2014). This identifies a key strength of satellite data, which can identify sources that are not  
830 part of an apriori (inversion model initialization of sources).

831 Finally, as part of this study, changes in chlorophyll were investigated with respect to changes in SST and CH<sub>4</sub>.  
832 These relationships need to be evaluated in future research to tie the dramatic changes in the ecosystem and physical  
833 oceanography of the Barents and Kara Seas, leveraging the strengths of satellite data.

#### 834 5. Conclusion

835 In this study, the global, repeat nature of satellite data was used to investigate the relationship between currents,  
836 and trends in sea surface temperature, ice extent, and methane (CH<sub>4</sub>) anomaly for the Barents and Kara Seas for  
837 2003-2015. Large positive CH<sub>4</sub> anomalies were discovered around Franz Josef Land and offshore west Novaya  
838 Zemlya in September, in areas where downstream current shoaling, with far smaller CH<sub>4</sub> enhancement around  
839 Svalbard, again, strongest where currents likely shoal downcurrent of seabed seepage. This highlights a major  
840 strength of satellite data: Identification of sources that are not part of an apriori used to initialize inversion models.

841 The strongest SST growth was southeast Barents Sea in June where strengthening of the warm Murman Current  
842 (an extension of the Norwegian Atlantic Current) could explain the trend, and in the south Kara Sea in September,  
843 whereas the cold southwards-flowing Percey Current weakened. These regions also exhibit the strongest CH<sub>4</sub>  
844 growth enhancement as well as around Franz Josef Land. Likely sources are CH<sub>4</sub> seepage from extensive oil and gas  
845 reservoirs underlying the central and east Barents Sea and Kara Sea; however, the spatial pattern was poorly  
846 correlated with depth and best correlated by strengthened currents that shoal.

847 Trends in the Barents Sea and Kara Seas will lead to an ice free Barents Sea free in about 15 years, while  
848 driving seabed warming and enhanced CH<sub>4</sub> emissions, particularly from areas where currents drive methane  
849 shoaling.





850

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1182 **TABLES**1183 **Table 1.** Slopes of  $SST$  ( $^{\circ}\text{C yr}^{-1}$ ),  $\text{CH}_4$  (ppb  $\text{yr}^{-1}$ ), and  $\text{CH}_4'$  (ppb  $\text{yr}^{-1}$ ) for focus boxes. <sup>a</sup>

1184	Box	$SST$	$\text{CH}_4$	$\text{CH}_4$	$\text{CH}_4'$ (Barents) <sup>b</sup>	$\text{CH}_4'$ (Arctic) <sup>c</sup>
1185		2003-2015	2003-2015	2005-2015	2003-2015	2003-2015
1186	A1	0.102	3.35	3.26	0.179	0.0750
1187	A2	0.0319	3.49	3.38	0.267	0.213
1188	A3	0.00178	3.19	3.17	-0.0185	0.00574
1189	A4	0.0867	3.37	3.60	0.310	0.391
1190	A5	0.0279	3.10	3.22	0.0105	0.0319
1191	A6	0.00259	3.07	3.24	-0.0123	0.0548
1192	A7	0.0323	3.06	3.27	-0.0460	-0.119
1193	A8	0.0552	3.11	3.35	0.0642	-0.0544
1194	A9	0.145	3.20	3.44	0.103	0.109
1195	A10	0.0527	3.32	3.51	0.122	0.0613

1196 <sup>a</sup>  $SST$  – Sea Surface Temperature,  $\text{CH}_4'$  – methane anomaly.1197 <sup>b</sup>  $\text{CH}_4'$  relative to the Barents Sea1198 <sup>c</sup>  $\text{CH}_4'$  relative to the Arctic Ocean





# FIGURE CAPTIONS

**Figure 1 a)** Arctic and sub-arctic methane ( $\text{CH}_4$ ),  $0.5^\circ$  gridded, 0–4 km altitude, 2016, from the Infrared Atmospheric Sounding Interferometer (IASI); mountainous regions blanked. Data were filtered as in Yurganov and Leifer (2016a). Data key on panel.

**Figure 2 a)** Map of the Arctic Basin, showing study area (Blue Square) and average January and September 2003–2015 ice extent. **b)** Bathymetry of the study area (87.468 N, 1.219E; 72.056N, 0.173E; 63.008N, 48.05E; 69.707N, 82.793E) from Jakobsson et al. (2012). Dashed black line shows approximate Barents Sea boundaries. Dashed white line shows edge of submerged permafrost from Osterkamp (2010). Star shows scoping study pixels location.

**Figure 3.** Comparison of the sea surface temperature ( $SST$ ) and methane ( $\text{CH}_4$ ) for 2003–2015 for pixels between Franz Josef Land and Novaya Zemlya (**Fig. 2b, Star, Supp. Table 1, Box A2**). Red diamonds show  $SST$  and  $\text{CH}_4$  averages within the study area. Blue and green ovals highlight pixels with different  $\text{CH}_4$  trends for  $SST$  (all  $\text{CH}_4$ ), and ( $\text{CH}_4 > 1925$  ppb), respectively.

**Figure 4. a)** Currents for Barents and nearby seas, bathymetry features, and focus-area locations. Green, red, and blue arrows are coastal, warm Atlantic origin, and cold polar currents, respectively. Broken lines illustrate current subduction. Bathymetry from Jakobsson et al. (2012). **b)** Monthly ice extent for 2015. Focus study boxes (numbered); coordinates listed in **Supp. Table S1**. Barents Sea currents adapted from Stiansen et al. (2006); see **Supp. Fig. S2** for greater detail for Svalbard area; for Kara Sea area from Polyak et al. (2002); see **Supp. Fig. S2** for greater detail. For Barents Sea Opening area from Bøe et al. (2015).

**Figure 5. a)** Surface *in situ* methane ( $\text{CH}_4$ ) during northward Barents Sea transect on the *R/V Akademik Federov* for 21 Aug. 2013. Also shown is the 300-m depth contour and edges of the Murman Coastal Current, from PINRO ([http://www.pinro.ru/labs/hid/kolsec1\\_e.htm](http://www.pinro.ru/labs/hid/kolsec1_e.htm)). Data key on figure. **b)**  $\text{CH}_4$  profiles during northerly and southerly transits, labeled.

**Figure 6.** Ice-free months from 2003 to 2015 for focus boxes for **a)** Northern Barents (A1–A3), **b)** Northwest of Barents (A4–A6), and **c)** Southern Barents (A7–A10). Box names on panels. See **Fig. 4a** and **Supp. Table S1** for locations.

**Figure 7.** Sea surface temperature ( $SST$ ) time series for 2003 to 2015 for focus box areas **a)** Northern Barents (A1–A3), **b)** Northwest of Barents (A4–A6), and **c)** Southern Barents (A7–A10). Annual values are average of all months, generally May–October, which are ice-free. Box names on panel a. Data key on figure.

**Figure 8.** Focus study area methane ( $\text{CH}_4$ ) for 2003 to 2015 for **a)** Northern Barents study boxes, **b)** Northwest of Barents study boxes, and **c)** Barents Sea focus study boxes. Annual data and 3 year, rolling-average data shown. Anomaly is relative to entire Barents Sea. Data key on figure.

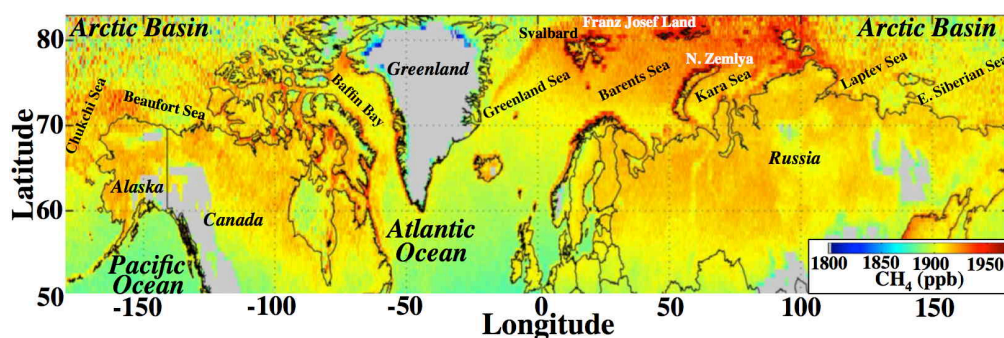
**Figure 9.** Mean values for 2003 to 2015 of sea surface temperature ( $SST$ ) for **a)** June and **b)** September. Mean methane ( $\text{CH}_4$ ) concentration for **c)** June and **d)** September. Median ice edge for same period is shown. Years with reduced ice extent contribute to values of  $SST$  north of this ice edge. Data key on figure.

**Figure 10.** Linear trends for 2003 to 2015 of sea surface temperature ( $dSST/dt$ ) for **a)** June and **b)** September. Methane concentration trend ( $d\text{CH}_4/dt$ ) for **c)** June and **d)** September. ND – not detectable – failed statistical test. Blue, black dashed lines shows 100 and 50 m contour, respectively. Data key on figure.



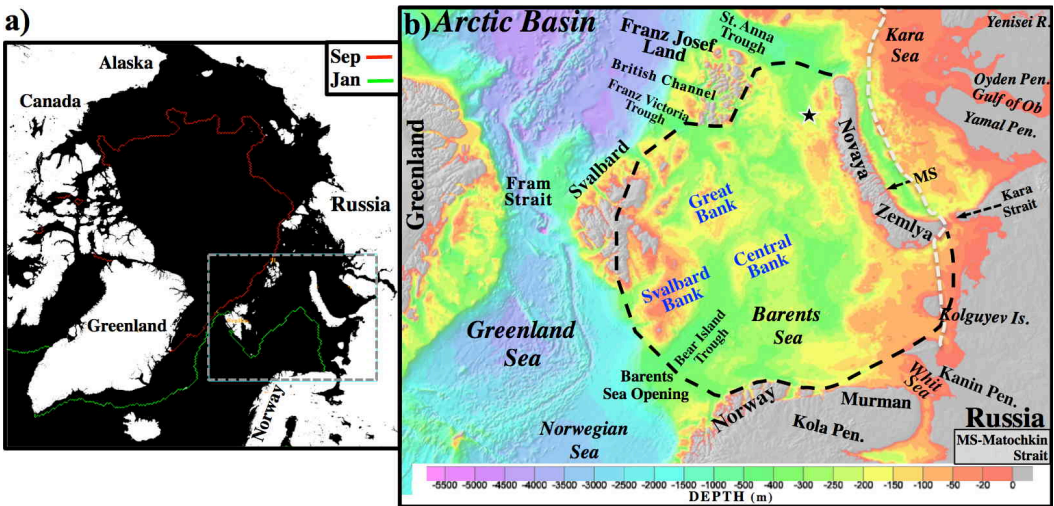


# FIGURES



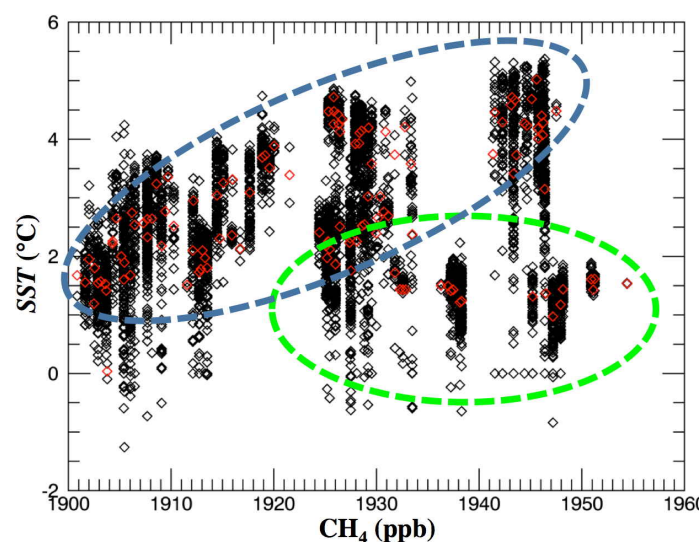
**Figure 1** Arctic and sub-arctic methane ( $\text{CH}_4$ ),  $0.5^\circ$  gridded, 0-4 km altitude, 2016, from Infrared Atmospheric Sounding Interferometer (IASI); mountainous regions blanked. Data were filtered as in Yurganov and Leifer (2016a). Data key on panel. For polar stereographic view see **Supp. Fig. S8** and Supplemental Movie showing entire time series.





**Figure 2 a)** Arctic map, showing study area (Blue Square) and average January and September 2003-2015 ice extent. **b)** Bathymetry of the study area (87.468 N, 1.219E; 72.056N, 0.173E; 63.008N, 48.05E; 69.707N, 82.793E) from Jakobsson et al. (2012). Dashed black line shows approximate Barents Sea boundaries. Dashed white line shows edge of submerged permafrost from Osterkamp (2010). Star shows scoping study pixels location. Depth data key on panel.





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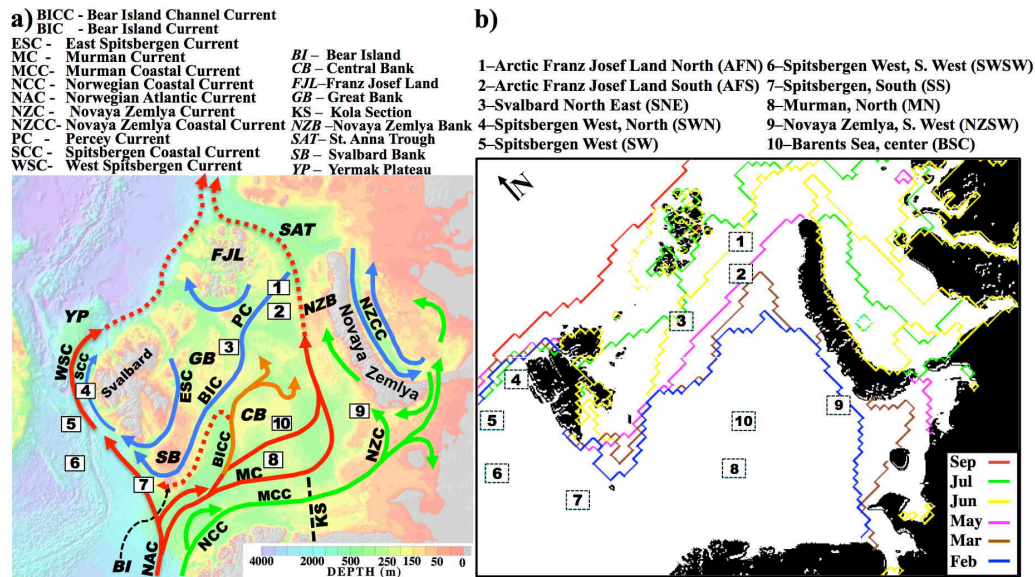
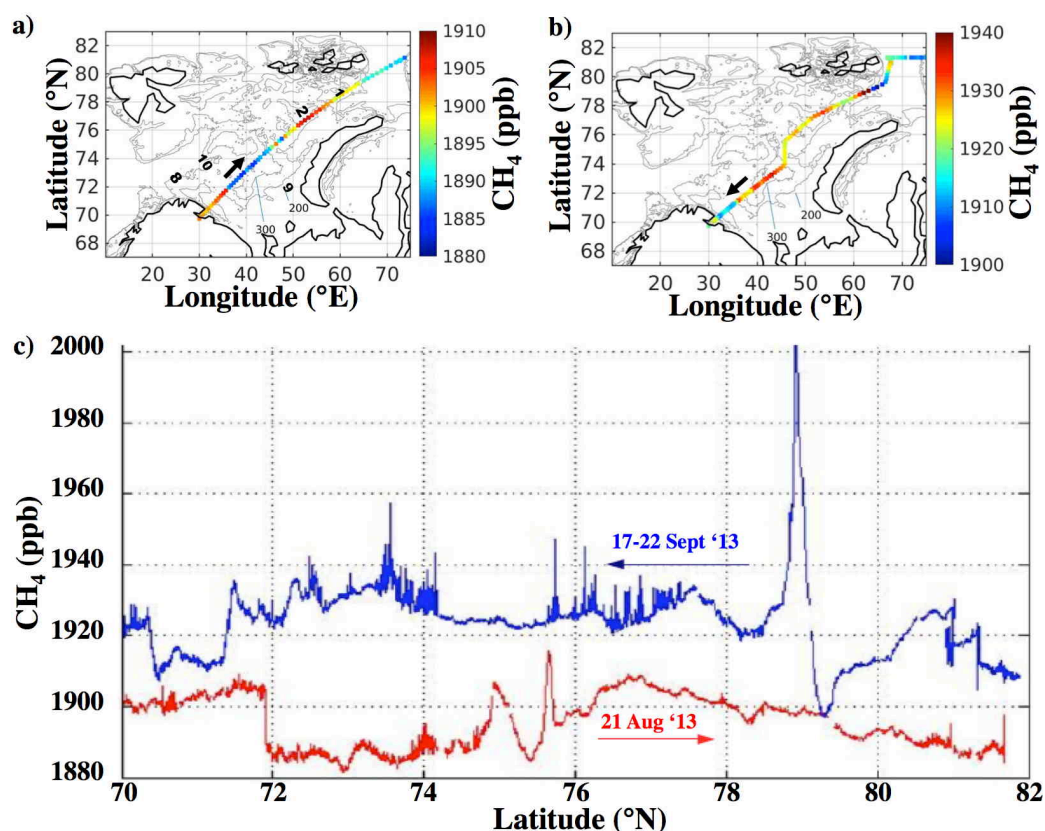


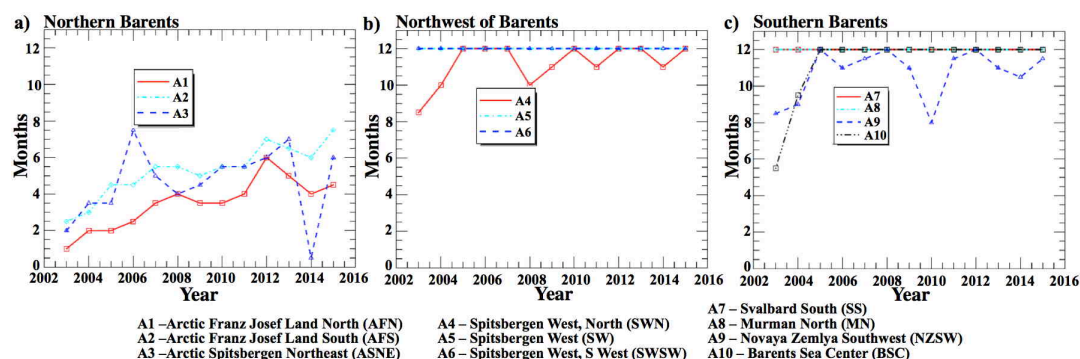
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**Figure 5.** Surface *in situ* methane (CH<sub>4</sub>) on the *R/V Akademik Fyodorov* for Barents Sea **a)** northwards transect for 21 Aug. 2013. Focus areas along pathway shown. **b)** Southwards transect for 17-22 Sept. 2013. Also shown is the 300-m depth contour and edges of the Murman Coastal Current, from PINRO ([http://www.pinro.ru/labs/hid/kolsec1\\_e.htm](http://www.pinro.ru/labs/hid/kolsec1_e.htm)). Data key on figure. **c)** CH<sub>4</sub> profiles during northerly and southerly transits, labeled.



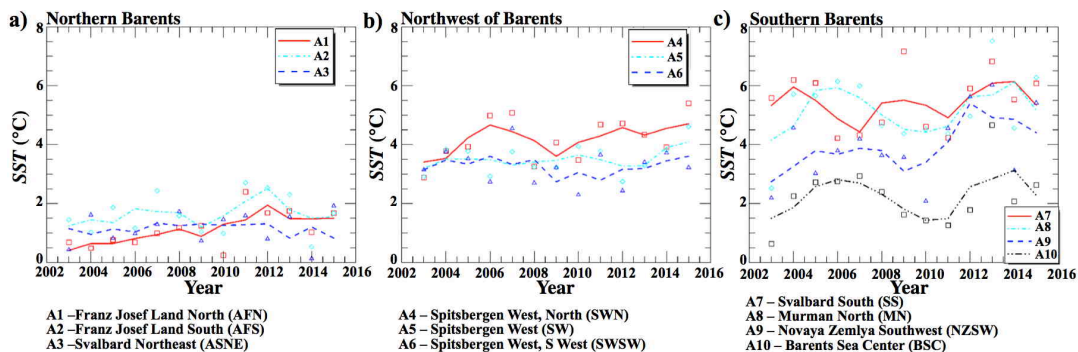


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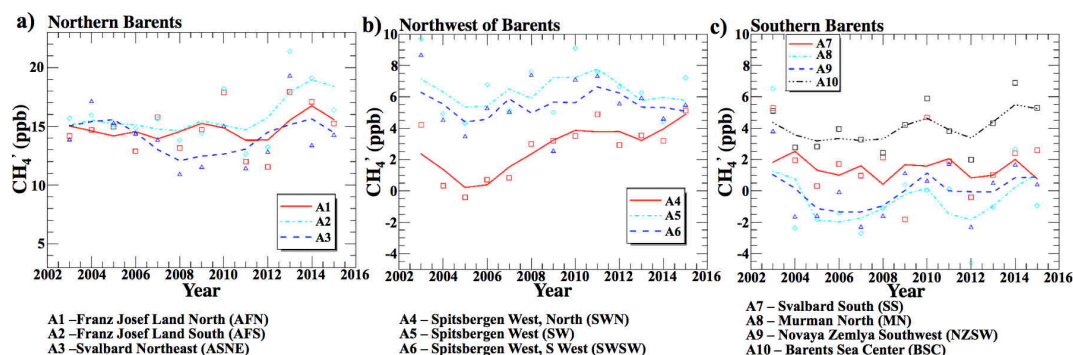
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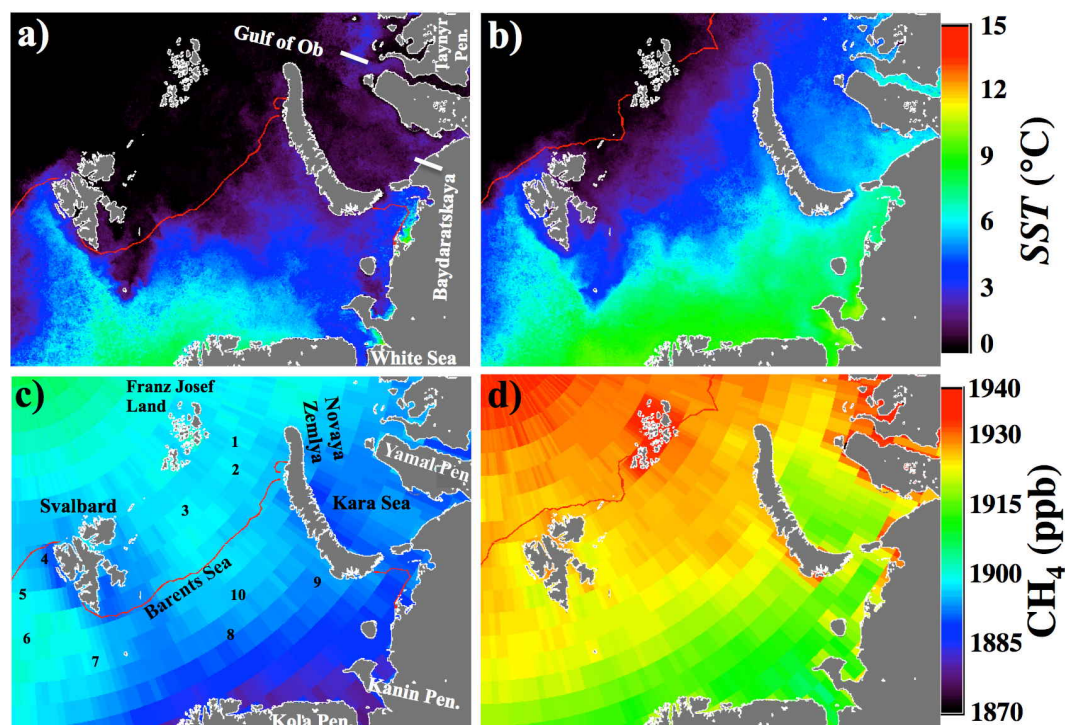


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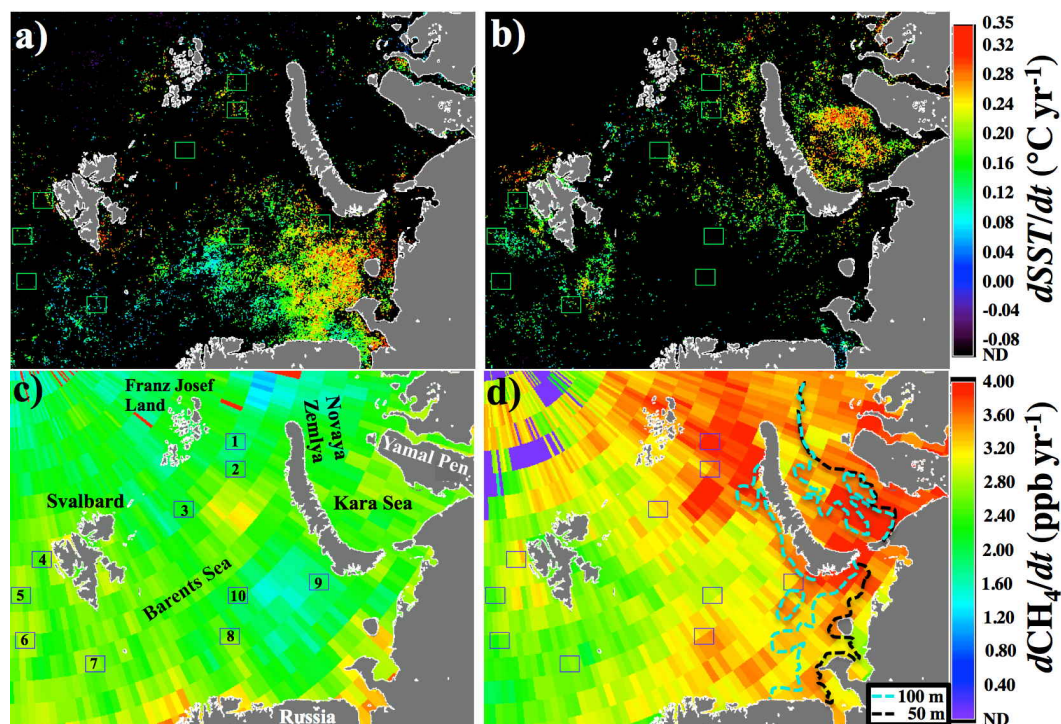
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