



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

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Abstract. Long-term (2003-2015) satellite-derived sea-ice extent, sea surface temperature (SST), and lower tropospheric methane (CH₄) of the Barents and Kara Seas (BKS) were analyzed for statistically significant anomalies and trends for 10 focus areas and on a pixel basis that were related to currents and bathymetry. Large positive CH₄ anomalies were discovered around Franz Josef Land (FJL) and offshore west Novaya Zemlya in early fall. Far smaller CH₄ enhancement was around Svalbard, downstream of known seabed seepage.
Strongest SST increase was southeast Barents Sea in June due to strengthening of the warm Murman Current

(MC) and in the south Kara Sea in September, when the cold Percey Current weakens and the MC strengthens. These regions and around FJL exhibit the strongest CH₄ growth. Likely sources are CH₄ seepage from subsea permafrost and hydrates and the petroleum reservoirs underlying the central and east Barents Sea and the Kara Sea. The spatial pattern was poorly related to depth and better explained by shoaling. Peak CH₄ anomaly is several

20 months after peak *SST*, consistent with a several month delay between *SST* and seabed temperature. Continued MC 21 strengthening will increase heat transfer to the BKS, rendering the Barents Sea ice-free in about 15 years.

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23 Keywords: Arctic, methane, sea surface temperature, ice, Barents Sea, warming, currents, emissions

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- 25 Highlights:

• Warm, northwards flowing Murman Coastal Current penetrates further into the Barents and Kara seas

- 27 Currents transport positive SST and drive growing methane emissions
- Shoaling provides a mechanism that allows deep-water methane to reach the atmosphere
- Franz Josef Land and the west coast of Novaya Zemlya are important, un-inventoried and growing CH4 sources
- 30





31 **1. Introduction**

32 1.1 Changes in the Arctic Environment in the Anthropocene

Over recent decades, the Arctic has been experiencing amongst the fastest changes from global warming, termed Arctic amplification (Manabe and Stouffer, 1980) with the Arctic Ocean warming at nearly double the rate of the rest of the world's oceans (Hoegh-Guldberg and Bruno, 2010). Arctic amplification is strongly evident in the progressive reduction of sea ice cover over the Arctic Ocean associated with sea surface temperature increases (Comiso, 2012; Comiso et al., 2008; Graversen et al., 2008; Hoegh-Guldberg and Bruno, 2010; Overland and Wang, 2013; Screen and Simmonds, 2010; Stroeve et al., 2014). Data since 1948 show that Arctic Ocean and atmospheric temperatures and storm frequency increased as sea ice extent and volume decreased (NRC, 2014).

One of the most evident, recent manifestations of Arctic change is the observed progressive decrease in sea-ice extent, which underlies numerous ocean physical (NRC, 2014) and ecosystem feedbacks (Alexander et al., 2018). These include heat transfer, light availability in the water column, and momentum transfer (convective and wind mixing), as well as ocean heat and moisture exchange with the atmosphere. Sea ice changes affect weather and overall surface albedo (NRC, 2014).

45 The Arctic has global impacts by directly affecting weather including extremes at mid-latitudes (Cohen et al., 2014), by affecting global albedo and hence the earth's energy balance (NRC, 2014), and by contributing to the 46 47 budgets of methane (CH₄), a potent greenhouse gas with greater radiative impact on a 20 year time scale than carbon dioxide (IPCC, 2007; Fig. 2.21). Since pre-industrial times, CH₄ emissions have risen by a factor of 2.5 48 49 (Dlugokencky et al., 2011). Inventories have high uncertainty currently (IPCC, 2013), which is greater in future 50 projections (Saunois et al., 2016), with the Arctic contributing large uncertainty. Underlying the latter are the vast 51 Arctic CH₄, CH₄ hydrates, and organic material stores trapped under permafrost both onshore (Tarnocai et al., 2009) 52 and offshore (Archer et al., 2009). Rapid warming shallow of Arctic marginal seas will degrade the integrity of 53 submerged permafrost and its sequestered CH₄ (Shakhova et al., 2017). As a result, Arctic marine CH₄ emissions are 54 being driven by the processes that are underlie this marine warming including retreat of sea ice, heat transport by 55 currents, and increased solar energy inputs that arises in part from the higher albedo associated with sea ice retreat.

56 FIGURE 1 HERE

Arctic amplification has implications for seabed methane (CH₄) emissions – particularly for seabed CH₄ currently "sequestered" beneath subsea permafrost – terrestrial permafrost inundated by rising sea level after the Holocene. For example, extensive seabed CH₄ seepage is linked closely with destabilization of subsea permafrost in the East Siberian Arctic Sea (Shakhova et al., 2013) and has been estimated as comparable to emissions from Arctic Tundra (Shakhova et al., 2015). Warmer seabed temperatures will degrade subsea permafrost integrity further (Shakhova et al., 2017), enhancing emissions (Shakhova et al., 2015); however, time scales remain uncertain.

The marine Arctic also is affected by changes in the terrestrial Arctic, not just climate, but also fresh water and organic material inputs from rivers. Arctic soils contain 50% of the global subterranean carbon pool of which 88% is estimated sequestered in permafrost (Tarnocai et al., 2009). A quarter of this 1670 Pg (1 Pg= 10^{15} g) carbon pool may be mobilized into the Arctic Ocean and sub-marginal seas over the next century due to Arctic warming (Gruber et





al., 2004). The Arctic and sub-Arctic show strong terrestrial, high latitude, CH_4 anomalies for eastern Canada, Alaska, and Western Russia (**Fig. 1**). Still, the Barents Sea, where the most rapid ice loss has occurred (Onarheim

and Årthun, 2017) and the Kara Sea, show the strongest anomalies by far.

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71 **1.2. Study Motivation**

We hypothesize that both lower tropospheric CH_4 and primary productivity – two parameters that are amenable to remote sensing – correlate with changes in the overall water column temperature more directly than to changes in sea surface temperature (*SST*), another satellite remote sensing product. Satellite data are key as they allow repeat observations of multiple variables on synoptic scales. In this study, we investigate atmospheric CH_4 and *SST* trends to determine the relationship in the spatial distribution of Arctic Ocean tropospheric CH_4 with respect to areas that become ice-free seasonally and inter-annually.

The largest Arctic sea ice cover reductions over the last decade have occurred in the Barents Sea, which likely will be the first ice-free arctic sea. Sea ice reduction directly affects CH_4 flux to the atmosphere by no longer impeding gas transfer, but also may relate to other oceanographic changes that affect regional ocean-atmospheric CH_4 fluxes. Specifically, we propose that warming *SST* relates to warmer seabed temperatures and hence CH_4 emissions from subsea permafrost and hydrate destabilization. The relationship between CH_4 seepage and atmospheric CH_4 is indirect – seabed CH_4 must be transported across the water by bubbles, diffusion, vertical mixing, and advection on timescales faster than microbial oxidation timescales.

We investigate this question by analyzing the timing of sea ice retreat and identifying statistically significant *SST* and atmospheric CH₄ trends from satellite data for 2003-2015.

87 FIGURE 2 HERE

The potential for this approach was revealed in a scoping study of a small area (**Supp. Table S1, Box A2**) in the marginal ice zone where Barents Sea water flows into St. Anna Trough between Franz Josef Land and Novaya Zemlya (**Fig. 2b, star**). For these pixels, satellite observations 2003-2015 show a correlation between CH₄ and *SST* for one of two pixel populations (**Fig. 3**). Based on this result, we test our hypothesis in marginal ice zones where the relationship is predicted to be strongest. Given that the oceanography, including ice, varies dramatically across the Barents and Kara seas we select ten sub-areas to elucidate how trends differ across these marginal Arctic seas. Sub-areas were large enough to decrease noise and small enough not to reduce trends from averaging.

95 FIGURE 3 HERE

96 1.3 Global and Arctic Atmospheric Methane

The atmospheric CH_4 concentration has been rising in recent years (Nisbet et al., 2014) and depends on the balance between sources and sinks – primarily hydroxyl (OH). Several processes may explain this trend including increasing Arctic emissions, wetland emissions, fossil fuel emissions, and/or decreasing losses from OH (Ghosh et al., 2015; John et al., 2012; Nisbet et al., 2014; Turner et al., 2016) compensated by decreasing biomass burning (Desjardins et al., 2018). The current atmospheric lifetime of CH_4 is ~8.5-9 yr (Sonnemann and Grygalashvyly,





102 2014; Voulgarakis et al., 2013). Rigby et al. (2017) suggest a decline in OH likely contributed to CH_4 increases 103 since 2007. Many CH_4 source estimates have a large uncertainty (IPCC, 2013; Saunois et al., 2016) with future 104 emission estimates having still higher uncertainty (Prather et al., 2012).

105 Uncertainty is particularly acute for Arctic sources. Global atmospheric CH₄ concentrations are highest in the 106 Arctic, increasing poleward (Xiong et al., 2016). Supporting this enhancement are strong Arctic CH₄ sources, 107 including seabed emissions, terrestrial riverine runoff (Shakhova et al., 2013), and atmospheric transport from 108 terrestrial sources (industrial, permafrost, wetlands, fires, etc.). Given that Arctic OH concentrations are less than at 109 lower latitudes (Liang et al., 2017), winter Arctic CH₄ lifetime is longer than in summer. Arctic OH exhibits a 110 seasonal variability that imposes an approximately 10 ppb seasonality in CH₄ concentrations (Thonat et al., 2017). Future uncertainty in Arctic sources is magnified by the strong and continued projected Arctic warming (Graversen 111 112 et al., 2008).

Arctic seabed CH_4 sources include thermogenic (geological) seepage (Shakhova et al., 2013), biogenic CH_4 113 production (James et al., 2016; Reeburgh, 2007) and submerged permafrost, originally from biogenic and/or 114 115 thermogenic sources (Shakhova et al., 2013). Seabed emissions largely are bubbles or dissolved gas; however, 116 microbial oxidation in near seabed sediments (the microbial filter) limits the importance of dissolved seabed CH₄ 117 fluxes (Reeburgh, 2007). Bubble seepage directly transports CH₄ up into the water column and potentially the sea 118 surface after losses through dissolution. Bubble seepage also indirectly transports fluid with dissolved CH_4 (Leifer and Patro, 2002). The fate of dissolved seep CH₄ depends strongly on its dissolution depth (Leifer and Patro, 2002) 119 120 with microbial oxidation expected to remove dissolved CH₄ below the Winter Wave Mixed Layer (WWML) 121 (Rehder et al., 1999), whereas dissolved CH₄ in the WWML mostly escapes to the atmosphere. The fraction of seep 122 CH₄ that dissolves below versus within the WWML depends strongly on seabed depth, volume flux (Leifer and 123 Patro, 2002), plume synergies that include the upwelling flow (Leifer et al., 2009), and bubble surface properties including the presence of impurities on the bubble surface (Leifer and Patro, 2002). Frequent Arctic storms both 124 deepen the WWML significantly and efficiently sparge dissolved CH₄ to the atmosphere (Shakhova et al., 2013). 125 126 Field studies and numerical modeling have documented that even for deepsea seepage (to ~1 km) seep bubbles can transport some of the CH₄ to the upper water column and potentially sea surface due to plume and deepsea bubble 127 processes (MacDonald, 2011; Rehder et al., 2009; Solomon et al., 2009; Warzinski et al., 2014). 128

129 1.4. Arctic Sea Surface Temperature

130 Arctic sea surface warming arises from several factors, including the distributions of cloud cover, sea ice, ocean 131 color, upper-ocean stratification, and heat transport between the world's oceans. Sea ice significantly reduces 132 surface albedo in the Arctic summer; thus, its absence increases absorption of solar insolation by Arctic Ocean 133 waters. This may lead to a feedback wherein reduced Arctic Ocean ice cover drives greater warming and further decreasing ice. For example, anomalously warm Barents Sea SST (>+2° C in 2015 relative to 1982-2010) may be 134 135 associated with low sea-ice cover in this region and exposure of surface waters to direct solar heating (Timmermans, 136 2016). Ocean color affects the vertical profile of absorption of solar energy with near-surface heating more likely lost to the atmosphere than deeper heating. Visible solar energy is absorbed in near surface waters by both 137





phytoplankton and other suspended and dissolved organic matter (DOM) that originates in riverine discharges
(Stedmon et al., 2011). The colored fraction of DOM (CDOM) reduces available light across the water column,
suppressing photosynthesis and increasing stratification (Granskog et al., 2007).

141 Stratification plays an important role in the Barents Sea energy budget. Barents Sea water column structure is 142 modulated by winter cooling of surface waters and their convective mixing as well as brine ejection of fresh water 143 during ice formation. Winter vertical mixing extends to the seabed over large portions of the shallow (200-300 m) 144 Barents Sea. In spring, the warming of surface waters and freshwater from melting ice support water column 145 stability and stratification in the central and southern Barents Sea (Loeng, 1991). Stratification isolates deeper waters 146 from the atmosphere and prevents vertical mixing of dissolved CH₄, trapping it in deeper water (Leifer et al., 2015) 147 while also isolating it from heat exchange with the atmosphere. Coastal waters off Norway and the Murman coast 148 remain stratified year round due to terrestrial freshwater inputs. Offshore, stratification strengthens in the spring as 149 surface waters warm and ice melts (Loeng, 1991).

150 Currents drive heat exchange between the Arctic Ocean and the Atlantic and Pacific Oceans. These exchanges 151 are major drivers of Arctic Ocean spatial thermal heterogeneity with additional inputs in coastal waters from riverine 152 outflow. Atlantic currents are the major contributor of oceanic heat to the Arctic climate system on annual (Lien et 153 al., 2013) and seasonal time scales (Lien et al., 2017). These currents include the West Spitsbergen Current 154 (Piechura and Walczowski, 2009), the Bear Island Channel Current (BICC), the Murman Current (MC), and the Norwegian Atlantic Current (NAC) (see Fig. 4a), which transport Atlantic Ocean heat to the Arctic through the 155 Barents Sea. The south fork of the NAC is entrained into the Norwegian Coastal Current (NCC), which is 90% 156 157 Atlantic water and 10% river discharge (Skagseth et al., 2008). Variability appears related to the North Atlantic 158 Oscillation (NAO) with higher Barents Sea temperatures during the NAO's positive phase (Dickson et al., 2000). 159 This transport has caused significant warming and ice retreat in this area of the Arctic Ocean (Smedsrud et al., 2013). Winds modulate the volume flow of Atlantic water into the Barents Sea - stronger in winter and weaker is 160 summer (Stiansen et al., 2009; Fig, 2.3.4). Ice processes further complicate the re-distribution of heat for surface 161 162 Arctic Ocean waters - ice coverage insulates the water from atmospheric cooling (heat transfer to the atmosphere), better preserving its heat and thereby furthering heat transport into the Arctic (Lien et al., 2017; Lien et al., 2013). 163 164 Sea temperature lags atmospheric temperatures by 2-3 months, peaking for the Kola area (offshore Murman, Russia) between 0 and 200 m in September-October, whereas air temperature peaks in July (Stiansen et al., 2009, Figs. 165 166 2.3.3. 2.3.8).

Positive feedbacks underlie Arctic amplification. For example, sea-ice reduction increases albedo and greater heat absorption in upper water layers (IPCC, 2013). There also are more complicated feedbacks. The progression of warmer water into the Barents Sea drives local winds that decrease wind-advection of sea ice, with decreased sea ice coverage *increasing* heat loss from the atmosphere (Lien et al., 2017).

171 1.5. Airborne and Satellite observations of Tropospheric Methane

Although the Arctic covers a vast territory, our knowledge of Arctic processes is highly limited both in spatial





Satellite sensors have advantages for Arctic observations including quick revisit times and synoptic coverage (Leifer
 et al., 2012) and can fill the significant existing temporal and spatial gaps between the few airborne field datasets.

176 1.5.1. Airborne Measurements

177 A few airborne campaigns have been conducted to measure Arctic atmospheric CH_4 since 2005. Given the 178 highly extensive spatial scales of the Arctic, these campaigns provide only a few summer snapshots of a highly 179 variable domain.

180 CH₄ concentration profiles over the Arctic Ocean were measured on five flights during the HIAPER Pole-to-181 Pole Observations (HIPPO) campaign (Kort et al., 2012; Wofsy, 2011) and produced evidence of sea surface CH₄ 182 emissions from the northern Chukchi and Beaufort Seas in most profiles, up to 82° N. Enhanced concentrations near 183 the surface of the Ocean were common over fractured floating ice in sample profiles collected on 2 Nov. 2009, 21 184 Nov. 2009, and 15 Apr. 2010. On 13 Jan. 2009 and 26 Mar. 2010, when the seasonally highest level of sea-ice 185 coverage occurred, CH₄ emissions were weak or non-existent. Some of the observational variability was correlated 186 with carbon monoxide (CO), indicating terrestrial origin.

187 The Carbon in Arctic Reservoirs Vulnerability Experiment (CARVE) program sought to quantify Alaskan CO2 and CH₄ fluxes between the atmosphere and surface terrestrial ecosystems. Intensive aircraft campaigns with 188 ground-based observations were conducted during summer from 2012-2015 (Chang et al., 2014). No open ocean 189 190 measurements were made. Additional Alaskan airborne data were collected summer 2015 (Jun.-Sept.) by the 191 Atmospheric Radiation Measurements V on the North Slope of Alaska (ARM-ACME) project (38 flights, 140 192 science flight hours), with vertical profile spirals from 150 m to 3 km over Prudhoe Bay, Oliktok Point, Barrow, 193 Atqasuk, Ivotuk, and Toolik Lake. Continuous data on CO2, CH4, CO, and nitrous oxide, N2O, were collected 194 (Biraud, 2016).

195 West of Svalbard, an area of known widespread seabed CH₄ seepage aligned along a north-south fault parallel to the coast (Mau et al., 2017; Westbrook et al., 2008) was the focus of a field airborne campaign June-July 2014 196 197 (Myhre et al., 2016). Flights were conducted using the Facility for Airborne Atmospheric Measurements (FAAM) of the Natural Environment Research Council (NERC, UK). The campaign measured a suite of atmospheric trace gases 198 199 and was coordinated with oceanographic observations. Seabed CH₄ seepage led to significantly increased seawater 200 CH₄ concentrations. However, no significant atmospheric CH₄ enhancement was observed for the region above the seeps for data collected summer, 20 Jun. - 1 Aug. 2014 (Myhre et al., 2016), albeit under a period of mostly light 201 202 winds.

203 1.5.2. Satellite

Satellite observations provide long-term temporal context for campaign data, which are necessarily, limited in time, often limited in spatial coverage, and often only occur during a specific season when weather is acceptable for flight logistics. As such, satellite observations complement high spatial-resolution airborne and boat-based field observations, which are infrequent and sparse at best. Remote sensing measures column gas abundance and thus is independent of potential mismatches between the platform altitude and the altitude of enhanced CH₄. Airplanes may





not fly sufficiently low to collect data in the planetary boundary layer (PBL), which often is shallow in the marine
 Arctic (Aliabadi et al., 2016).

211 Satellite sensors leverage CH₄ spectral features at 1.67 and 2.32 µm in the Short Wave InfraRed (SWIR) (Clark et al., 2009) and around 7.28 µm in the thermal infrared (TIR) (Tratt et al., 2014). CH4 retrievals for SWIR sensors, 212 213 which use passive reflective solar radiance, are challenged in the Arctic by high cloud cover, low solar zenith angle, 214 and low reflectivity for ice, snow, and water (Leifer et al. 2013). TIR sensors measure emissivity radiance, and thus 215 comparatively shorter path lengths at high latitudes relative to SWIR sensors that measure reflected sunlight. Also, TIR sensors can retrieve CH₄ above low clouds, both daytime and nighttime – SWIR is only daytime and requires 216 217 cloud free skies. Thus, TIR sensors have significant Arctic advantages for marine CH4 retrievals compared to SWIR 218 sensors (Leifer et al. 2013). However, whereas SWIR sensors primarily respond to near-surface CH₄, TIR retrievals 219 generally have higher sensitivity to mid-tropospheric CH_4 than to near-surface CH_4 (Xiong et al., 2013).

Recent SWIR satellite CH₄ sensors include the recent Scanning Imaging Absorption SpectroMeter for Atmospheric CHartographY (SCIAMACHY-ESA: 2002-2012: 100-km resolution) mapping mission (Bovensmann et al., 1999) and the active, Greenhouse Orbiting Satellite (GOSAT-JAXA: 2009-: 9-km resolution) sampling mission (Kuze et al., 2009). The TROPOspheric Monitoring Instrument (TROPOMI-ESA: 2017– at 7-km resolution) mapping mission (Veefkind et al., 2012) returned first images in 2017. The scheduled geostationary CARBon cycle observatory (GEOCARB-NASA; at 4-km resolution) mapping mission will return hourly (daytime) revisit data for North America (Rayner et al., 2014) in the early 2020s.

In the TIR, the AIRS (Atmospheric InfraRed Sounder) mission onboard the Earth Observation Satellite, Aqua
satellite (Aumann et al., 2003b) and the EuMetsat IASI-1 (InfraRed Atmospheric Sounder Interferometer) mission,
on the MetOp-A platform (Clerbaux et al., 2009) provide long-term CH₄ observations. Accompanying IASI-1
(2007-) is IASI-2 (2013-) on the currently orbiting MetOp-B meteorological satellite. The IASI satellites follow sun
synchronous orbits. Additionally, three IASI New Generation instruments (Crevoisier et al., 2014) are planned for
launch in 2021, 2028, and 2035 (IASI-NG, 2017). AIRS CH₄ profiles are retrieved from the 7.8 µm TIR channel
(Aumann et al., 2003b).

234 The IASI instruments are cross-track-scanning Michelson interferometers that measure in 8461 channels at 0.5 cm^{-1} spectral resolution from three spectrometers spanning 645 to 2760 cm^{-1} . These spectrometers have a 2×2 array 235 of circular footprints with a nadir spatial resolution of 12 km that is 39 × 25 km at swath (2400 km) maximum 236 237 (Clerbaux et al., 2009). IASI-1 was launched into an 817 km-altitude polar orbit on 19 Oct. 2006, while IASI-2 was launched on 17 Sept. 2012. MetOp-A and MetOp-B cross the equator at approximately 09:30 and 21:30 local time, 238 239 separated by approximately half an orbit, resulting in twice daily, near-global coverage with 29 day. The on-flight 240 noise-equivalent delta temperature at 280K is estimated to be well below 0.1K in the spectral range of interest to CH₄ (Razavi et al., 2009). Like AIRS, IASI has a wide swath with a scan angle of ±48.3°. IASI CH₄ retrieval 241 242 algorithms are described by Xiong et al. (2013) and Gambacorta (2013).

AIRS is a grating diffraction nadir cross-track scanning spectrometer on the Aqua satellite (2002-) that is part of the Earth Observation System (Aumann et al., 2003a). AIRS was launched into a 705-km-altitude polar orbit on the EOS Aqua spacecraft on 4 May 2002. The satellite crosses the equator at approximately 01:30 and 13:30 local time,





producing near global coverage twice a day. Effective field of view after cloud clearing, as described by Susskind et al. (2006), is 45 km and the spectral resolution for CH_4 is 1.5 cm⁻¹. Version 6 of AIRS Levels 2 and 3 data are publicly available (AIRS, 2016) – see Xiong et al. (2010) for a description, evaluation, and validation of global CH_4 AIRS retrievals. Lower-troposphere (0-4 km altitude averaged) AIRS profiles (AIRS time series is longer than IASI) are analyzed herein.

251 Validation is critical to any remote sensing approach and has been addressed in a number of studies for the 252 lower and mid-upper Arctic troposphere. Xiong et al. (2010) compared aircraft data taken over Poker Flat, Alaska, and Surgut, Siberia with AIRS CH4 retrieved profiles and found agreement within 1.2% with mean 253 254 measured CH₄ concentration between 300-500 hPa; correlation coefficients were ~0.6-0.7. A significantly wider geographical coverage was achieved for IASI validation (Xiong et al., 2013) during a quasi pole-to-pole flight of the 255 National Science Foundation's Gulfstream V aircraft (Wofsy, 2011). A bias of nearly -1.74% was found for 374-256 477 hPa and -0.69% for 596-753 hPa. Yurganov et al. (2016) compared 5-year long IASI data for 0-4 km layer over 257 258 a sea area adjacent to the Zeppelin Observatory, Svalbard, Norway, at 474 m altitude, operated by the Norwegian Institute for Air Research (NILU). Monthly mean values and monthly trends were in good agreement, but daily 259 260 excursions did not correlate. Yurganov et al. (2016) explained the latter by the observatory's location being near the 261 top of the planetary boundary layer.

262 IASI has been used to study lower (<4 km) tropospheric CH₄. Yurganov et al. (2016) found low atmospheric CH_4 anomalies in summer for 2010-2015 with annual Arctic Ocean CH_4 fluxes estimated as being ~2/3 those from 263 the terrestrial Arctic. Positive CH₄ anomalies were observed along the coasts of Norway, Novaya Zemlya, and the 264 265 Svalbard archipelago primarily during November-January (Yurganov and Leifer, 2016a). A breakdown of the Arctic 266 oceanic summer thermal stratification by wind-induced mixing in autumn may underlie this seasonal trend. A 267 breakdown of stratification is proposed for CH₄ emissions to the atmosphere from the North Sea, which also is highly stratified in the summer and fall (Leifer et al., 2015). Additionally, Yurganov and Leifer (2016b) report 268 significant CH₄ increases during the 2015/2016 winter over the Sea of Okhotsk. 269

270 2. Method and Study Design

271 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 atmospheric CH₄ column. We concentrate on five area types: (1) Arctic water affected areas; (2) combined Arctic and Norwegian Atlantic Current affected areas; (3) Barents Sea Polar Front affected areas; (4) Murman Current affected areas; and (5) Norwegian Coastal Current and Murman Coastal Current affected areas. 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 then investigates these trends in relationship to oceanographic and meteorological processes and available, Barents and Kara Seas data 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 methane emissions.

287 FIGURE 4 HERE

288 2.2. Methodology

289 Satellite data

290 AIRS CH₄ data (version 6) are publicly available from NASA Goddard Space Flight Center (GSFC) since 2002 291 (AIRS Science Team/Joao Texeira, 2016). CH₄ data for 2003-2015 are retrieved by the NOAA Unique Combined 292 Atmospheric Processing System (NUCAPS) algorithm, developed at NOAA/NESDIS in cooperation with Goddard 293 Space Flight Center (GSFC). Data are analyzed for open ocean areas with high vertical thermal contrast, defined 294 here as the temperature difference between the surface and altitude of 4 km (Yurganov and Leifer, 2016a; Yurganov et al., 2016). CH₄ data are re-projected to a 4-km azimuthal equal area projection. The CH₄ anomaly (CH₄) is 295 296 calculated by subtraction of the values computed within each of the 10 focus areas from the average of the whole 297 Barents Sea ocean climatology. As CH₄ shows high inter-annual variability, a three-year running average is applied.

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; Ackerman et al., 1998). The mapped products match the CH₄ data projection.

303 First, data are quality reviewed for sea ice coverage and cloud coverage filtered for coastlines, which are from 304 the Global Self-consistent, Hierarchical, High-resolution Shoreline database (SEADAS, 2017). Shape files of sea-ice 305 monthly extent are obtained from National Snow and Ice Data Center (Fetterer et al., 2017) and are based on 306 monthly passive microwave radiometry with the Bootstrap algorithm (Comiso et al., 2008). Sea-ice fields are 307 provided on a polar stereographic grid at 25-km resolution. The number of ice-free months is derived from the 308 intersection of the monthly ice shape file for each year with the focus areas. The number of ice-free months each 309 year is tallied by the following rules: if the intersection is less than 15%, it is counted as 0 months; if coverage is 310 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 311 single month the pixel is counted as ice covered for the month. Ice-covered (>50%) pixels are not used in the trend 312 or the climatology calculations for SST (CH₄ retrievals are accurate over both ice and seawater).

313 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 a linear regression





analysis. These linear trends are analyzed using the Mann Kendall Test (Önöz and Bayazit, 2003) and Sen's linear

317 trend analysis (Juahir et al., 2010; Sen, 1968).

318 Visual analysis of the trends and anomaly maps of the Barents Sea were used to determine the location of 10

focus areas. The average trends of the 10 focus boxes are calculated from the average of all valid pixels in each focus box each year for the same months.

321 2.3. Setting

322 Oceanography and Meteorology of the Barents and Kara Seas

Significant Arctic Ocean water derives from the North Atlantic, which becomes denser through cooling.
 Additional contributions are riverine and precipitation (which decrease water density) and Pacific water from the
 Bering Strait. Most of this water returns to the North Atlantic as part of the global thermohaline circulation (Aagaard
 and Carmack, 1989; Carmack and McLaughlin, 2011; Yamamoto-Kawai et al., 2008).

The relatively shallow (230-m average depth) Barents Sea (**Fig. 4**) is characterized by a deep Arctic shelf with complex bathymetry and hydrography (Loeng, 1991). The Barents Sea is bounded to the south by the northern coast of Europe, to the north by two archipelagos, Svalbard and Franz Josef Land (FJL), to the east by the large northsouth oriented island, Novaya Zemlya, and to the west by the Norwegian Sea. In winter, the Barents Sea is partially ice-covered, while it is almost ice-free in the summer (**Fig. 4b**).

332 Barents Sea physical oceanography is influenced strongly by inflows from the North Atlantic and the Arctic 333 Ocean. North Atlantic water inflows to the Norwegian Sea, forming the Norwegian Atlantic Current (NAC), one 334 track of which becomes the West Spitsbergen Current (WSC) in the Greenland Sea and the Fram Strait, before bearing eastwards to the north of Svalbard into the Arctic Ocean. The remainder flows into the Barents Sea through 335 the Barents Sea Opening. Whitehead and Salzig (2001) suggested (and demonstrated in the laboratory) that remote 336 337 forcing of the NAC through the Barents Sea lifts the current by several hundred meters to the sill of the Bear Island 338 Channel, forcing significant anticyclonic vorticity. This drives the retrograde Bear Island Channel Current (BICC) northeast along the slope of Svalbardbanken and the prograde MC along the slope of Tromsøflaket, eastward and 339 340 north to the east of Central Bank (Li and McClimans, 1998; Loeng, 1991). These meet east of Central and Grand 341 Banks. The resulting flow cools from contact with the atmosphere into a denser, modified Atlantic Water flow that 342 exits through the St. Anna Trough to the east of Franz Joseph Land (Gammelsrød et al., 2009). Cooling at the banks 343 also produces a dense westward underflow, depicted by the dashed line in Fig. 4a.

344 The Norwegian Coastal Current (NCC) follows the Norwegian and Murman coastlines and incorporates fresh 345 water runoff from northern Europe and Atlantic Water. The NCC is a major contributor of oceanic heat to much of 346 the southern and eastern Barents Sea and into the Kara Sea (Lien et al., 2013). The NCC cools significantly through 347 interaction with the atmosphere, whereas the western fork of the NAC (the WSC) submerges north of Svalbard 348 (location varying seasonally) under an isolating layer of colder and fresher water, better preserving its heat (Lien et 349 al., 2013). The name of the NCC changes to the Murman Coastal Current (MCC) as crosses into Russian waters. 350 Long-term (1905-) temperature data for the upper 200 m are available from a section off the Kola Peninsula (Fig. 4a, KS, black dashed line), which the MCC crosses (Boitsov et al., 2012). These data reveal long-term trends with 351





a cooler period from 1875-1930 and continuous warming of ~0.8°C since a minimum in 1970-1980 (Skagseth et al.,
2008). Skagseth et al. (2008) found good agreement in the Kola Section temperature trend with the Atlantic Multi-

decadal Oscillation (AMO) index.

The MCC continues eastward at the border of the White Sea until the western shores of Novaya Zemlya divert the flow northward. It then continues into the Arctic Ocean through the St. Anna Trough between Franz Josef Land and Novaya Zemlya (Loeng, 1991), which is the dominant outflow of the Barents Sea (Maslowski et al., 2004). A fork of the MCC flows eastward into the Kara Sea through narrow and very shallow (20-50 m) straits (see **Supp. Fig. S2** for details of Kara Sea currents).

360 The Percey Current (PC) transports cold, low saline, Arctic surface water into the Barents Sea east of Svalbard (Supp. Fig. S1). The intersection of the Percey Current with the warmer, high saline waters of Atlantic origin in the 361 362 Barents Sea gives rise to the Barents Sea Polar Front (Oziel et al., 2016), whose location is controlled by seabed bathymetry, i.e., it is semi-stationary (Gawarkiewicz and Plueddemann, 1995). This front is part of a unique frontal 363 system due to its combination with the seasonally ice-covered zones in the northern, central, and eastern Barents Sea 364 365 (Vinje and Kvambekk, 1991). As a result, this front tends to exhibit enhanced phytoplankton blooms (Fer and 366 Drinkwater, 2014) and variability (Falk-Petersen et al., 2000). The PC merges with the East Spitsbergen Current 367 (ESC) and flows up the west Spitsbergen coast, inshore of the WSC, as the Spitsbergen Coastal Current (SCC), 368 leading to the Barents Front looping around Spitsbergen (Svendsen et al., 2002).

Winds in the eastern Barents Sea generally circulate counterclockwise (cyclonically), from the north along 369 Novaya Zemlya in winter and spring and weakly from the south in summer and fall (Gammelsrød et al., 2009). This 370 371 leads to calm over the Central Bank, easterlies to the north of Franz Josef Land and from the north to the west of 372 Svalbard (Kolstad, 2008; Moore, 2013). The Barents Sea is stormy - over 125 days per year have winds above 15 m s^{-1} , which are mostly from the south (Kolstad, 2008). In spring, winds are similar, but displaced to the south. In the 373 summer, winds blow to the south over most of the Barents Sea, except from the west off Svalbard, with average 374 summer winds of ~6 m s⁻¹ (Kolstad, 2008). In the fall, winds are similar to the summer, but stronger (~8-10 m s⁻¹) 375 376 and strongly northerly between Svalbard and Greenland. Decadal-averaged air temperatures on Bear Island have been rising (~2.5°C) since 1980 (Boitsov et al., 2012), about four times larger than the global atmospheric trend over 377 the same period of ~0.6°C (http://eca.knmi.nl/). 378

379 Kara Sea hydrography is controlled by the freshwater outflow of the Ob and Yenisei Rivers (Fig. 2b; Supp. Fig. S2), which contribute 350 and 650 km³ yr⁻¹, respectively (Stedmon et al., 2011), about double that of the 380 Mississippi. Sediment from these estuaries lead to the northeast Kara Sea being mostly shallow (< 50 m). The 381 382 western Kara Sea is deep (mostly >100 m), descending to below 500 m in the Novaya Zemlya Trough (Polyak et al., 383 2002). River outflows are primarily (>75%) between May and September. As a result, east Kara Sea surface waters can be brackish. The inflow of modified Atlantic water from the Barents Sea supplies the deeper water in the trough. 384 385 Cooler surface water from the MCC, local runoff, and ice from the north flow into the NZCC, some of which returns to the Barents Sea through the Kara Strait. The trough is dense and fully saline. Cooler surface water from the Arctic 386 387 Ocean flows south in the narrow and weak Novaya Zemlya Coastal Current (NZCC) and exits through the Kara 388 Straits. Additionally, there is a strong, submerged, cool southward flow of Arctic Ocean water along the Novaya





Zemlya Trough. Warmer water enters from the Barents Sea flows east through the Kara Strait and joins a slope current to the north. Much of this water mixes with the southern and returns to the Barents Sea through the northern Kara Strait (McClimans et al., 1999; McClimans et al., 2000). Overall, currents through the Kara Sea are largely northwards, driven by river outflow. The remainder of the north current flow splits to the west and joins the cold southward NZCC. 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).

395 Ten focus areas (Fig. 4a; Table 1 for locations) were selected based on the location of Barents and Kara Seas 396 currents and ice formation dynamics to investigate the effect of the significant differences between winter and 397 summer ice coverage. These focus areas are grouped into 5 oceanographic types. The most northerly focus areas (A1-A3) characterize the inflow of Arctic surface water through both gaps between the archipelagos of Svalbard and 398 399 Franz Josef Land and between Franz Josef Land and Novaya Zemlya. Each of these focus areas exhibits different 400 seasonal ice coverage. Another group of focus areas are west of Svalbard (A4-A6) and are influenced by the West 401 Spitsbergen Current and water from the Barents Sea. These areas also are affected by Arctic Ocean ice outflow 402 along Greenland. A focus area near Bear Island (A7) is affected by the warm, north flowing NAC and the cold, 403 southwest-flowing Percey Current and is located in a subduction area. Two focus areas (A8 and A10) were selected 404 that are influenced by the Murman Current and Arctic water from the Percey Current in the Barents Sea Polar Front 405 region (Harris et al., 1998). Finally, one focus area (A9) is situated in the coastal waters to the west of Novaya Zemlya and is influenced by the MCC with strongly, seasonally varying ice coverage. 406

407 3.0 Results

408 3.1. Barents Sea In situ

In situ CH₄ measurements were made by cavity enhanced absorption spectroscopy (Los Gatos Research Inc., Mountainview, CA). Both transits followed a very similar trajectory (**Supp. Fig. S3**) and found very strong, localized, CH₄ anomalies - see **Supp. Fig. S3** for full dataset. These anomalies were far from shore, 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. Material Sect. 2** for more details.

414 FIGURE 5 HERE

There was an abrupt drop in CH_4 around 72°N for the outwards transit, which increased again around 75°N. This depressed CH_4 portion of the transit corresponded fairly closely with where the vessel left the warm Murman Coastal Current (**Supp. Fig. S4a**). The strongest anomaly, to 2000 ppb, was observed on the southwards transit where the MCC rises over the sill into the Santa Anna Trough (78.7°N), close to the focus area shown in **Fig. 3**.

The two transits were separated by about a month with the September transit higher by \sim 30 ppb than in August, consistent with strong seasonal CH₄ trends. There were other significant differences. Whereas there were many narrow, implying local, CH₄ anomalies during the southwards transit, there were far more than during the northwards transit and the peak at 78.7°N was not repeated, indicating variability in the emissions.





423 The difference between these transits highlights the challenges of interpreting such snapshot data, supported by 424 the comparison with IASI pixels that were proximal and within several days (**Supp. Fig. S4**). Agreement for the

425 northwards transit was reasonably good (generally within 10 ppb), and generally poor for the southwards transit.

426 3.2. Focused Study Area Annual Trends

Trends in aggregated pixel "focus areas" are compared for three Barents Sea regions, "Northwest of Barents" including the Greenland Sea and Fram Strait, west of Svalbard (A4-A6), "Northern Barents" in the marginal ice zone at the edge of the Arctic Ocean (A1-A3) and "Southern Barents," which is strongly under the heat influence of the east fork of the NAC (A7-A10). Of these, A7, A8, and A9 also cover banks – offshore areas of elevated seabed topography. Grouping of focus areas with similar trend patterns into these three Barents Sea regions was based both on physical oceanography and the detailed character of these trends, described below.

433 FIGURE 6 HERE

Focus areas with the strongest decreasing ice cover trends are in the marginal ice zone of the northern Barents Sea (south and east of Franz Josef Land), at the southern margin of the Arctic Ocean (**Fig. 6a**, **A1-A3**). Trends for these three study areas are very similar (after classifying 2006 and 2014 for focus area A3 (Svalbard 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**).

439 The similarity in ice coverage trends for area A3 (along the cold Percey Current) with areas A1 and A2 (along 440 the Murman Current's warm, northward leg) suggests not only increasing northward heat transfer, but also 441 weakening southward cold-water advection. Area A4 (northwest of Svalbard) also shows decreasing ice coverage 442 towards more frequent year-round ice-free status and lies at the Arctic Ocean 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 443 444 Barents Sea (A1-A3). The Central Bank of the Barents Sea (Fig. 6c, A10) last saw an ice-covered month in 2005, 445 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. 446

447Overall, all focus areas are trending towards year-round ice-free, with the entire Barents Sea likely to be year448round ice free by ~2030 based on an extrapolation of trends in Northern Barents Sea focus areas, A1-A3.

449 FIGURE 7 HERE

450 SST increases in all focus areas (Fig. 7) albeit at rates spanning a wide range, from 0.0018 to 0.15 $^{\circ}$ C yr⁻¹ (see 451 Table 1). In the Northern Barents Sea, the strongest warming trend is for area A1, east of Franz Josef Land. This is 452 located in a marginal ice zone, in the path of the warm MC. Area A3 shows the weakest warming trend lies along 453 the cold Percey Current. These trends also are consistent with a strengthening of warm currents and weakening of 454 cold currents inferred from the changes in ice coverage. For the Northwest of Barents focus areas (Fig. 7b, A4-A6), 455 the strongest warming is at the northernmost focus area, A4, whereas the weakest trend is for the southernmost focus 456 area (Fig. 7, A6). This also is consistent with a strengthened northwards penetration of the warm NAC forming the 457 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 and the warm BIC (A7). Areas A10 and A8, and to a lesser extent A9 all show 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 east of Franz Josef Land (areas A1 and A2). All the boxes that exhibit this oscillation lie along the Murman Current, whose origin is in the NAC.

465 FIGURE 8 HERE

466 A positive CH₄ trend is observed across the Barents and Kara Seas from June through September with some 467 regions exhibiting far stronger trends than average (Supp. Fig. S5). Areas of faster CH_4 increase include near Franz 468 Josef Land (Fig. 8a, A1, A2), the shallower waters offshore W. Svalbard (Fig. 8b, A4), and offshore Novaya 469 Zemlya (Fig. 8c, A9). These areas of increasing CH_4 correspond to areas of consistent warming for 2003-2015 (Fig. 470 7a, A1, A2) and consistent warming since ~2004/2005 for southwest offshore Novaya Zemlya and the Central Bank 471 of the Barents Sea (Fig. 7c, A8-A10). All these focus areas lie along the northwards flow of the Murman Current 472 and the Murman Coastal Current. In contrast, focus areas along the Percey Current show a slowly decreasing CH₄ 473 relative to the trend for the entire Barents Sea (Fig. 8, A3, A7), despite an (albeit weakly) increasing SST.

The strongest CH_4 growth is south of Franz Josef Land (**Table 1 A2**, 3.49 ppb yr⁻¹), followed by offshore 474 northwest Svalbard (Table 1 A4, 3.37 ppb yr⁻¹ - 2003-2015, 3.6 ppb yr⁻¹ 2005-2015). This positive trend is sustained 475 over the analysis period. This area off the Fram Strait has natural CH₄ seepage associated with hydrate 476 477 destabilization (Westbrook et al., 2008). This increase is annualized, and thus does not result from shifts in the 478 timing of seasonal warming. Note, the CH₄ slopes for areas A4-A10 all are larger when calculated from the 2005 479 minimum, but not for A1-A3 (Table 1). The former lie along the NAC and its eastern current fork, the Murman 480 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. 481

482 TABLE 1 HERE

483 The largest variability in *SST* and CH_4 was in the focus area to the north of Murman in the MC (**Table 1, A8**; 484 **Fig. 8**) and likely arises from variations in the strength and location of the MC, which bifurcates around this focus 485 area. Importantly, this area is located above a small, unnamed bank to the south of the Central Bank (**Fig. 2b**).

486 FIGURE 9 HERE

487 3.3. Barents and Kara Sea Climatology

Atlantic heat input is very important to the energy budget of the Arctic Ocean and Barents Sea and is driven by the two forks of the NAC (**Fig. 4a**) (Lien et al., 2013). This importance is evident in the Barents Sea *SST* climatology (**Fig. 9**). Warmer water flows eastward along the northern Norwegian and Murman coasts and along the eastern coastline of Svalbard (BIC), and north along the western Svalbard coast. Water becomes cooler as it penetrates eastward, and as it reaches the ice edge. 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





494 waters of Novaya Zemlya, warmer water extends further north than elsewhere. The warm signature disappears in the

area where the NAC submerges, near the northern shores of Novaya Zemlya (Fig. 4a).

496 FIGURE 10 HERE

In June, the warm water extent corresponds fairly well with the median ice edge location, which trends along the location of the cold Percey Current. Southeast of Svalbard, the Percy 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. S5**). For north Spitsbergen in the Svalbard archipelago (**Supp. Fig. S1**) 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 along southwest Spitsbergen underneath colder surface waters, likely below strong summer stratification. The WSC warm water is further offshore the west coast of Svalbard in June than in September (**Supp. Fig. S1**), i.e., the Barents Front shifts shoreward in summer.

By 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. 10b**). However, Barents Sea warming does not follow the ice edge between Svalbard and Franz Josef Land, corresponding instead to the location of the Percy Current. Still, whereas warm water is more extensive in south and east Barents Sea in September than August, in the northwest, cold water associated with the Percey Current has expanded from August to September (**Supp. Fig. S5**).

515 The now mostly ice-free Kara Sea in September exhibits coastal warming, particularly to the east, where there 516 also is heat input from the Ob and Yenisei Rivers (east of the Yamal Peninsula). This area exhibits evident warming 517 despite partial ice coverage of the Gulf of Ob in June and likely is driven by warmer riverine water inputs.

518 CH_4 concentrations show a clear latitudinal trend that increases towards the north. This latitudinal gradient is 519 weak in June and strong in September. Strong localized variations also occur in different Barents Sea regions. CH_4 520 concentrations along the Murman Current and in the (ice-covered) Kara Sea largely are below the latitudinal mean 521 in June, whereas west of Svalbard and in the north-central Barents Sea they are above average.

In June, CH_4 is depressed strongly around Svalbard and around Franz Josef Land and Novaya Zemlya. For Svalbard, this corresponds to the SCC that hugs the shore. By September, CH_4 concentrations are notably different with significantly higher CH_4 and a distinctly different spatial distribution. Most notable is the shift from depressed to strongly enhanced CH_4 in the region to the west of Novaya Zemlya and around the Franz Josef Land archipelago. Strong CH_4 enhancement also occurs in the outflow plumes of the Ob and Yenisei Rivers in the Kara Sea, around the Taymyr Peninsula. Around Svalbard, CH_4 has risen to near latitudinal mean levels in September, except for offshore north Spitsbergen and Nordaustlandet, where sea ice remains.





529 3.3. Barents and Kara Seas Trends

Across the Barents Sea, a number of different focus areas with distinct *SST* and CH₄ trends are identified (Fig.
7). These manifest significant spatial heterogeneity at the pixel scale and at the focus-area size scale. We analyze
trends in aggregated-pixel "focus areas" located in key regions where *SST* change is the strongest (Sec. 3.3; Supp.
Fig. S6 for July and August trends).

534 June SST warming trends are fairly different from September SST trends (Fig. 11). In June, warming occurs 535 much faster in the eastern Barents Sea, specifically, in waters affected by the Murman Coastal Current (MCC). This 536 suggests the magnitude of atmospheric cooling during transit from the Atlantic is decreasing. Warming occurs 537 primarily in shallow (generally less than 100-m deep) (Fig. 11b) waters that are generally well mixed. Sea ice is gone in this region by March-May, later in more northerly areas (Fig. 4b). Whereas there is no clear warming trend 538 in July and August; a strong warming trend appears in the Kara Sea by September (Supp. Fig. S5). This warming 539 540 trend occurs several months after the ice retreat - Kara Sea is ice-free in July (Supp. Fig. S5). This suggests 541 increasing MCC penetration into the Kara Sea. Loeng (1991) reported that MCC penetration into the Kara Sea was 542 uncommon in the middle of the 20th century.

543 FIGURE 11

544 More rapid warming occurs offshore of the western coast of Novaya Zemlya from June-September. This is 545 where the Murman Current (MC) transports water towards the St Anna Trough (the dominant Barents Sea outflow), 546 a region where shoaling is likely based on seabed topography (**Fig. 2b**) (Maslowski et al., 2004). The MC then flows 547 (and submerges under ice and Arctic surface water) along the east shores of Franz Josef Land. Accelerated warming 548 diminishes near the northern margin of the Kara Sea, where river outflow dominates the oceanography.

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

555 Overall Barents Sea atmospheric CH_4 is increasing (**Fig. 10C**), consistent with the global CH_4 trend (Nisbet et 556 al., 2014). However it is notable that some Arctic regions exhibit significantly more rapidly increasing CH_4 than the 557 global or Barents Sea trends. In June, CH_4 trends are largely similar in both ice-free and ice-covered areas. In near-558 coastal waters around Svalbard (except the east), in northern Norwegian fjords, and for the White Sea (Murmansk) 559 where CH_4 growth trends are elevated.

560 CH_4 trends in September, when ice coverage has retreated to the northern edge of the Barents and Kara Seas 561 (Fig. 10b), are very strongly enhanced in the East Barents Sea and the South Kara Sea. These areas coincided with 562 areas of enhanced *SST* warming and show CH_4 trends almost three times as high as the general Arctic trend. In 563 contrast, regions without enhanced warming, particularly waters affected by cold currents, exhibit the weakest CH_4 564 growth trend though slightly above the overall Barents Sea trend. In the Kara Strait between the Barents and Kara 565 Seas, CH_4 increases very strongly.





566 Enhanced CH₄ growth is not evident in either June or September to the north of Svalbard, despite strong *SST* 567 increases; however, significant increases are evident here in August. This follows significant CH₄ enhancement in 568 July to the southeast of Svalbard. This July-August shift follows the NAC.

569 4. Discussion

570 **4.1. Methane transport from the seabed to the atmosphere**

571 In this study, we hypothesized that lower tropospheric CH_4 correlates both with changes in the overall water 572 column temperature and with *SST* changes, both satellite remote sensing products. For this analysis, we also 573 considered the locations of currents and trends in these currents, seabed bathymetry, prevailing winds, and available 574 Barents Sea, water-column temperature data – primarily the long-term Kola Section data, which due to the 575 importance of the Murman Current was directly relevant.

576 The proposed source of the atmospheric CH₄ anomaly is seabed seepage from either thermogenic sources, i.e., 577 petroleum hydrocarbon reservoirs (Judd and Hovland, 2007), or degradation of submerged permafrost and hydrates 578 (Shakhova et al., 2017). Both permafrost and hydrate deposits can include both thermogenic and biogenic CH₄.

579 These emissions largely are as bubbles (Judd and Hovland, 2007) because the microbial filter generally removes 580 aqueous-enriched CH₄ fluid emissions from sediments to the water column (Reeburgh, 2003). As a bubble rises, it 581 loses CH4 to the water column by dissolution, transporting the remainder. Larger bubbles vertically transport a 582 greater fraction of their contents more efficiently than smaller bubbles (Leifer and Patro, 2002). Some portion of the 583 dissolved fraction is transported vertically by the bubble-driven upwelling flow (Leifer et al., 2009). The remaining 584 fraction may either diffuse to the atmosphere by turbulence or be oxidized microbially. In the deep sea, all the dissolved CH₄ likely is oxidized, where given the relevant depth scale is the winter wave mixed layer (WWML) 585 based on microbial oxidation timescales (Rehder et al., 1999). The Arctic WWML can extend to 100-200 m. In 586 587 shallow water (e.g., less than 20 m), most seep bubble CH4 reaches the sea surface, with the portion decreasing for 588 smaller bubbles or depth (Leifer and Patro, 2002). For example, Leifer et al. (2017) showed that for the Laptev Sea 589 that ~25% of seabed CH₄ from 70 m reaches the sea surface directly, consistent with sonar observations of bubble 590 plumes reaching the sea surface (Leifer et al., 2017). CH₄ that is deposited deeper in the wave mixed layer (WML) 591 likely diffuses to the atmosphere rapidly, although stratification powerfully suppresses this transport. Storms 592 breakdown this stratification (Leifer et al., 2015) sparging all the dissolved CH₄ to the atmosphere (Shakhova et al., 593 2013). Thus, CH₄ emissions that are deposited (by dissolution) into the WWML but below stratification may escape 594 many months later and distant from their seabed origin. In the process, the dissolved CH₄ drifts with currents, which if driven upslope, transports the CH_4 to shallower depths, potentially into the WML where it can escape to the 595 596 atmosphere, termed shoaling.

597 In practical terms, bubble transport means that seepage extends the depth of the WWML for CH_4 by 50-100 m, 598 i.e., 150-300 m, extending the depth that storms can sparge dissolve CH_4 to the atmosphere. This implies, CH_4 from 599 seabed seepage over a significant fraction of the Barents Sea (**Fig. 2b**) can reach the WWML, or can be transported 500 by currents into shallower waters (shoaling) into the WWML.





601 The above discussion was for non-oily seepage. However, where seepage arises from a petroleum hydrocarbon 602 reservoir, bubbles likely are oily. Oil slows bubble rise (Leifer, 2010) but reduces dissolution dramatically, allowing 603 their survival to far higher in the water column than non-oily bubbles (Leifer and MacDonald, 2003). Oily bubbles 604 can reach the sea surface from the deep sea due also to processes associated with hydrates, for example MacDonald et al. (2010) tracked seabed seep bubbles by ROV from 1 km to the WML (upper 20 m), causing a significant 605 606 positive CH₄ anomaly in the surface waters. Given the presence of extensive proven and proposed petroleum 607 reservoirs across the Barents and Kara Seas (Rekacewicz, 2005), it is likely that some Barents Sea seepage is oily 608 bubbles, enhancing CH₄ transport to the sea surface.

Positive, localized, atmospheric CH_4 anomalies can reflect local seabed production and vertical transport to the atmosphere, or lateral near-seabed transport and shoaling that elevates the dissolved CH_4 into the WML and then air sea exchange into the atmosphere, or transport from a distant source. Detangling these processes leverages the strength of the continuous and synoptic view of satellite datasets compared to the high spatial and temporal resolution snapshot provided by field campaigns.

614 One unlikely source of CH₄ anomalies for the Barents and Kara Seas is atmospheric transport as there is neither 615 significant local industry, nor extensive wetlands/terrestrial permafrost nearby and downwind for the prevailing 616 winds. Note - synoptic systems can transport CH₄ from northern Europe or Russia to the Barents Sea area, but 617 synoptic system winds are not dominant (prevailing) and thus play a small role in a time-averaged dataset. Moreover, these terrestrial sources are distant, implying extensive anomaly size scale in the Barents Sea. 618 619 Additionally, they would decrease with distance from northern Europe. Instead, the anomalies are localized and 620 decrease towards Europe. Although the oil production and pipeline infrastructure on the Yamal Peninsula and near 621 Kolguyev Island (Supp. Fig. S7) likely are strong CH_4 sources, they are downwind of the prevailing winds for the 622 Barents and Kara Seas.

623 4.2. Local versus Transported and the Importance of Shoaling

624 In this study we explore the likelihood that atmospheric CH_4 anomalies result from local generation and vertical 625 transport to the sea surface, versus distant lateral transport by currents prior to vertical transport to the sea surface.

626 Areas of accelerated CH₄ increasing trends were closely related to the path of the Murman Current as it flows towards the Kara Strait rather than depth (Fig. 11). Both the rising seabed bathymetry and the presence of both 627 628 southwards and northwards currents through the Kara Strait imply strong mixing and thus transport to the 629 atmosphere. Along this path are significant offshore petroleum hydrocarbon reservoirs, which could be seeping CH_4 into the waters of the Murman Current. Further evidence for transport and shoaling is the spatial distribution of CH₄ 630 631 around Kolguyev Island (north of the White Sea), which increases faster on its western side than its eastern side, 632 even though the sea to the island's east is shallower. In fact the CH₄ spatial pattern correlates better with shadowing 633 in the island's lee from shoaling currents, rather than with seabed depth. Prevailing winds are from the south-634 southeast / north-northeast (Kubryakov et al., 2016), thus atmospheric transport cannot explain the pattern, which 635 would be consistent with easterly winds.





636 In the Kara Sea, the correlation of enhanced CH_4 with depth is poor, which is shallower to the north. Instead, 637 the location of enhanced September CH₄ closely matches the location of oil and gas reserves, e.g., Supp. Fig. S7 638 and Rekacewicz (2005) and also along the path of warm water transport by the Murman Coastal Current as it enters 639 the Kara Sea and then follows the coastline of the Kanin Peninsula. Although there are extensive oil and gas 640 production on the Yamal Peninsula, prevailing winds blow away from the Barents Sea. Note, the trend shows an 641 acceleration of CH₄ growth, implying increasing emissions, i.e., not steady-state seabed warming but accelerating 642 seabed warming. This increasing CH₄ growth is for September, not June, corresponding to when the water column is 643 warmest in the South Barents Sea (Stiansen et al., 2009). Also, the Barents Sea outflow through St Anna's Trough is 644 greater (about double) than June (Gammelsrød et al., 2009) when the growth in the CH₄ anomaly occurs (Fig. 11d). The importance of this transport also is apparent in the SST trend with the greatest warming occurring in June in the 645 646 southeast Barents Sea (offshore the Kanin Peninsula) near the Kara Strait. This region lies to the west of the areas of 647 accelerating CH₄ growth in September near the Kara Strait. In contrast, significant SST warming is not observed in 648 September in this easternmost region of the Barents Sea.

Two other areas of accelerating CH₄ growth lie in the north-central Barents Sea, north of Central Bank, and offshore northern Novaya Zemlya. These regions lie along the pathway of the Murman Current, which is an extension of the warm NAC and the largest northwards transport of Barents Sea water (Lien et al., 2013). 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 tens of meters (**Fig. 2B**). Additionally, this region of accelerating CH₄ growth corresponds spatially to potential (unproven) gas and oil reserves that extend across the St Anna Trough to Franz Josef Land (**Supp. Fig. S7**; (Rekacewicz, 2005)).

There are proven oil and gas fields to the south, along the path of the Murman Current, but south of the area of accelerating CH_4 . Again, there is good spatial correlation with these proposed reservoirs. Notably, the enhanced CH_4 around Franz Josef Land does not correlate with the potential reserves, but does correlate with depth and the flow of the Murman Current, consistent with shoaling. Although some of the accelerating CH_4 near Novaya Zemlya could arise from increasing local seabed emissions, seabed temperatures were below zero until 2009 (Boitsov et al., 2012), which would imply submerged permafrost and/or hydrate deposits here have not yet degraded significantly, supporting both a transport and a shoaling mechanism.

663 4.3. Sea Surface Temperature

The analysis shows that CH_4 growth from portions of the Barents and Kara Seas is accelerating faster than the Barents Sea mean and the latitudinal mean. To some level these correlate with accelerating *SST* warming, but the correlation is poor. One factor underlying this is the 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 oscillation in the *SST* trend in the Southern Barents Sea (areas A8, A9, and A10) has a very similar timescale to the seabed trends reported by Boitsov et al. (2012) but precedes it by ~2-4 years.

670 More rapid warming occurs offshore Novaya Zemlya moving northwards from June-September, where the 671 Murman Current transports water and the seabed topography is likely to cause shoaling. This suggests 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 was driving the trend, the greatest acceleration would be in the northern Kara Sea, which also is shallower.

676 There are a number of possible explanations for why SST is warming fastest in regions along the Murman 677 Current and NAC. One is sea-ice retreat; however, the warming occurs several months after the retreat of the sea ice. 678 Another is that currents are transporting warmer water; however, then warming would occur all along the pathway 679 of the current. Third is that stratification is becoming shallower, allowing more cooling to the atmosphere. This 680 would imply a weakening of storms and winds – which firstly is inconsistent with warmer SST, and secondly, there 681 is no indication that Barents Sea storminess is changing or progressing further northwards (Koyama et al., 2017). 682 Another possibility is that currents are strengthening. Stronger currents could relate to larger oceanographic trends. 683 Seabed September temperatures (Boitsov et al., 2012) do not suggest increased warmer seabed temperatures north of 684 Norway and Russia, but do suggest warmer seabed temperatures to the east and also along Novaya Zemlya -685 suggesting a greater importance of the MC. This is consistent with the model of McClimans et al. (2000) that 686 currents are pushing the marginal ice zone. The warming trend suggests a strengthening of the seasonal trend in the 687 Barents Sea outflow, which is greater in September than June (Gammelsrød et al., 2009).

688 The strongest warming trend is for the shallow water off northwest Svalbard (area A4) (Fig. 11b), which also exhibited the strongest acceleration of CH₄ growth for around Svalbard. In this area, seabed topography is nearly 689 level over an extensive shelf with depths in the range 250-400 m. Where the shelf falls off sharply, rising sea 690 691 temperatures will minimally induce hydrate destabilization. In contrast, where the shelf falls off very gently, small 692 temperature increases shift extensive areas of seabed from below to above the hydrate stability field. This area is 693 immediately to the north of the area where several researchers have identified extensive seabed seep CH₄ emissions 694 (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 Front. 695

696 4.4. Implications for Svalbard Area Emissions

697 There are few atmospheric and ocean CH₄ data for the Barents Sea and surrounding areas, the most prominent being associated with CH₄ seepage off Spitsbergen, Svalbard, located immediately south of focus area A4. Studies 698 699 to date have been in early summer; Mau et al. (2017); Myhre et al. (2016) who made measurements in the 700 atmosphere and water column; Westbrook et al. (2009) reported sonar observations of seep bubbles for August-September, and slightly elevated aqueous CH₄ in surface waters immediately above the bubble plumes. All 701 702 concluded transport to the atmosphere was not significant, attributed to trapping of dissolved CH4 below a sharp 703 pycnocline. It is important to note that with respect to the overall Barents Sea area CH₄ anomaly, the Svalbard area 704 is far less important than around Franz Josef Land, off the west coast of Novaya Zemlya, and the north-central 705 Barents Sea (Fig. 10).

Both *SST* and CH_4 in June (**Fig. 10**) and July (**Supp. Fig. S5**) for west Svalbard show that much of the area of active seepage was inshore of the Barents Sea Front, and thus under the cooling Arctic waters of the Spitsbergen





Coastal Current (SCC), supported by reported salinity data (Mau et al., 2017). Although the *SST* remains suppressed off Spitsbergen in September, and extends further offshore, CH_4 concentrations no longer are depressed compared to Atlantic water further offshore, i.e., greater transport to the atmosphere. Such transport would not be expected downcurrent (north) of the bubble plumes observed by the early fall cruise reported in Westbrook et al. (2009).

712 Although data indicate these seeps do not contribute to summer atmospheric CH₄, this study suggests that the 713 emitted CH₄ likely is reaching the atmosphere far downstream where currents shoal. Interestingly, Mau et al. (2017; 714 Fig. 3) show data that could be interpreted as shoaling with elevated aqueous CH_4 forced shallower by the north-715 flowing SCC, rising as it crosses onshore-offshore aligned subterranean ridges. Focus area A4 shows strong increase 716 in CH_4 from 2005-2015 (the strongest of the focus areas (**Table 1**) and in increasing SST over this time period, consistent with shoaling. Larger acceleration of CH_4 growth is observed north of Spitsbergen in June (Fig. 11c), 717 718 which is the most likely location for shoaling based on detailed Svalbard bathymetry and currents (Supp. Fig. S2). Specifically, this is where some of the warm West Spitsbergen Current mixes with the cold, Spitsbergen Coastal 719 720 Current (SCC) that would be CH₄ enriched from seabed seepage, and then flows over relatively shallow seabed 721 towards the Hinlopen Strait. To summarize, although there is evidence of increasing downcurrent CH4 transport to 722 the atmosphere downcurrent of seepage off West Svalbard after shoaling, it is not significant with respect to overall 723 Barents Sea emissions.

There is evidence of acceleration in the CH_4 growth nearshore off western Svalbard in June, but not in September (**Fig. 11**) when CH_4 growth acceleration lies in the further offshore waters that are impacted by the warm WSC. Trends in *SST* also suggest a weakening of the Percey Current in June and more so in September. Given that from June to September the SCC extends further offshore, this suggests WSC control. Similarly, the WSC eastwards leg that crosses Nordaustlandet is driving a rapid increase in *SST* in September and likely relates to the increased CH_4 trend.

730 4.5. Ice-Free Barents Sea

The ice coverage trend shows that northeast Barents and southern Barents Sea already are ice-free or near icefree year round, whereas northwest Barents Sea (around Franz Josef Land and St. Anna Trough) remains icecovered for about half the year. The ice coverage trends (**Fig. 6**) suggest most of the Barents Sea will be ice free, year-round in another decade and a half, circa 2030. This is comparable to the 2023-2036 estimate of Onarheim and Årthun (2017; Fig. 3), which also notes that the current decreasing trend lies outside the oscillation envelope since 1850.

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





744 The Barents Sea is a marginal sea between the temperate Norwegian Sea and the Arctic Ocean and thus is the 745 conduit through which lower-latitude oceanic heat is transmitted to the Arctic Ocean (Onarheim and Årthun, 2017). 746 Given the significant role the Barents Sea plays in overall Arctic ice loss - fully 25% of the loss is attributed to the 747 Barents Sea, which comprises 4% of the Arctic Ocean and marginal seas (Smedsrud et al., 2013), implications will be significant for weather including at lower latitudes, and the marine ecosystem. Seemingly counter-intuitively, the 748 749 reduction of sea ice increases the upwards surface heat flux as ice has an insulating effect. Thus ice-loss somewhat 750 stabilizes Arctic Ocean ice, particularly during winter (Onarheim and Årthun, 2017) and may even lead to growth of 751 ice in the Arctic and Northern Greenland Seas. Still, the data herein showed a progressive weakening of the Percey 752 Current, which will continue to cause ice loss off east Svalbard and warming of these waters. This agrees with 753 Alexander et al. (2004) who concluded that the (semi-stationary due to bathymetry) Polar Front has shifted to 754 domination of Atlantic water over Arctic waters.

755 As already seen, though, the progression of ice loss in the south and east Barents Sea along the pathway of the 756 Murman Coastal Current has led to a progressive loss of ice in the south Kara Sea. Thus, the balance between the 757 two processes - heat loss to the atmosphere from and progressive transport of heat by currents to the Kara Sea are 758 clearly shifting towards warmer. The implications of decreasing ice coverage in the shallow Kara Sea are significant 759 with respect to CH₄ emissions – the area is rich in hydrocarbon resources that currently are sequestered (albeit data 760 show already poorly) under submerged permafrost that will continue to degrade, while warming seabed temperatures will enhance microbial degradation of the vast organic material deposited over the millennia by the Ob 761 and Yenisei Rivers. Thus, the already significant importance of Arctic CH₄ anomaly from the Kara Sea will 762 763 accelerate due to feedbacks from an ice-free Barents Sea.

764 4.6. Future research

765 Long-term oscillations with a timescale of 6-8 years were identified (e.g., Fig. 9); however, the length of the 766 dataset (13 years) is too short to investigate this in detail. Extending the analysis to include more recent data (say through 2020) would span a full 2 1/2 cycles and allow investigation of correlations with other driving 767 768 oceanographic atmospheric cycles, such as the NAO. This would be particularly valuable given that recent data 769 show that the most recent two years are the most extreme in terms of Barents Sea ice coverage Oziel et al. (2016) 770 and CH₄ anomaly (Supp. Fig. S8; Supp. Video). Extending the analysis forward in time clearly would provide 771 greater insights into the complex relationship between currents and CH₄ emissions. In this regard, the long CH₄ time 772 series planned to be collected by the IASI satellite series through the 2030s (Onarheim and Årthun, 2017) will be 773 invaluable.

Additionally, there is clearly need for these data to be incorporated into coupled atmospheric-oceanographic-ice models to understand in greater detail the processes underlying the changes, improving the ability to forecast trends in Arctic marine greenhouse gas emissions. Currently the strong and growing CH_4 anomaly from Novaya Zemlya and Franz Josef Land are the strongest in the Arctic, yet are not yet incorporated (or identified) in inversion models, e.g., Crevoisier et al. (2014). This identifies a key strength of satellite data, which can identify sources that are not part of an apriori.





- Finally, as part of this study, changes in chlorophyll were investigated with respect to changes in SST and CH₄.
- 781 These relationships need to be evaluated in future research to tie the dramatic changes in the Barents and Kara Sea
- recosystem to physical changes in oceanography, leveraging the strengths of satellite data.

783 5. Conclusion

In this study, the synoptic, repeat nature of satellite data was used to investigate the relationship between currents, and trends in sea surface temperature, ice extent, and methane (CH_4) anomaly for the Barents and Kara Seas for 2003-2015. Large positive CH_4 anomalies were discovered around Franz Josef Land archipelago and offshore west Novaya Zemlya in September, in areas where currents shoal, with far smaller CH_4 enhancement around Svalbard, again, strongest where currents likely shoal, downcurrent of seabed seepage. This highlights a major strength of satellite data: Identification of sources that are not part of an apriori in inversion models.

The strongest *SST* increase was southeast Barents Sea in June due to strengthening of the warm Murman Currents (an extension of the Norwegian Atlantic Current) and in the south Kara Sea in September, whereas the cold Percey Current weakened. These two regions also exhibit the strongest CH_4 growth acceleration as well as around Franz Josef Land. Likely sources are CH_4 seepage from extensive oil and gas reservoirs underlying the central and east Barents Sea and Kara Sea; however, the spatial pattern was poorly correlated with depth and best correlated with strengthening current that shoal.

796If current trends continue, then heat flows to the Barents Sea and Kara Sea by strengthening currents will lead to797an ice free Barents Sea free in about 15 years, while driving seabed warming and enhanced CH_4 emissions,798particularly from areas where currents drive shoaling.

799

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1242	TABLI	ES									
1243	Table 1 . Slopes of <i>SST</i> (°C yr ⁻¹), CH ₄ (ppb yr ⁻¹), and CH ₄ (ppb yr ⁻¹) for focus boxes. ^{<i>a</i>}										
1244	Box SS2	Г	CH_4	CH_4	CH4' (B	arents) ^b	CH ₄ ' (Ai	rctic) ^c			
1245	2003-20		15	2003-2015		2005-20	15	2003-2015		2003-2015	
1246	A1 0.1	02	3.35	3.26	0.179	0.0750					
1247	A2 0.0	319	3.49	3.38	0.267	0.213					
1248	A3 0.0	0178	3.19	3.17	-0.0185	0.00574					
1249	A4 0.0	867	3.37	3.60	0.310	0.391					
1250	A5 0.0	279	3.10	3.22	0.0105	0.0319					
1251	A6 0.0	0259	3.07	3.24	-0.0123	0.0548					
1252	A7 0.0	323	3.06	3.27	-0.0460	-0.119					
1253	A8 0.0	552	3.11	3.35	0.0642	-0.0544					
1254	A9 0.1	45	3.20	3.44	0.103	0.109					
1255	A100.0	527	3.32	3.51	0.122	0.0613					
1256	^{<i>a</i>} SST – Sea Surface Temperature, CH_4 – methane anomaly.										
1257	^b CH ₄ ' relative to the Barents Sea										
1258	^c CH ₄ ' relative to the Arctic Ocean										





1259 FIGURE CAPTIONS

Figure 1 a) Arctic and sub-arctic methane (CH₄), 0.5° 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 Ocean, 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 line shows approximate
Barents Sea boundaries. Star shows scoping study pixels location. Depth data key on panel.

Figure 3. Comparison of the sea surface temperature (*SST*) and methane (CH₄) 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 CH₄ averages within the study area. Blue and green ovals highlight pixels with different CH₄ trends for *SST* (all CH₄), and (CH₄>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.

Figure 5. a) Surface in situ methane (CH₄) 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 pinru(<u>http://www.pinro.ru/labs/hid/kolsec1_e.htm</u>). Data key on figure. b) CH₄ 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. 3c 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 (CH₄) trends, 2003 to 2015 for a) Arctic Ocean study boxes, b) Northwest of Barents study boxes, and c) Barents Sea focus study boxes. Annual data and 3 year, rollingaverage data shown. Anomaly is relative to entire Barents Sea. Data key on figure.

Figure 9. Trends for focus area A8 (north of Murman) for 2003 to 2015 for CH_4 ' and *SST*. Data key on figure.

1291 Figure 10. Mean values for 2003 to 2015 of sea surface temperature (*SST*) for **a**) June and **b**) September.

1292 Mean methane (CH₄) concentration for **c**) June and **d**) September. Median ice edge for same period is 1293 shown. Years with reduced ice extent contribute to values of *SST* north of this ice edge. Data key on 1294 figure.

Figure 11. Linear trends for 2003 to 2015 of sea surface temperature (dSST/dt) for a) June and b) September. Methane concentration trend (dCH_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.









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1301 **Figure 1** Arctic and sub-arctic methane (CH_4) , 0.5° gridded, 0-4 km altitude, 2016, from the Infrared 1302 Atmospheric Sounding Interferometer (IASI); mountainous regions blanked. Data were filtered as in

Atmospheric Sounding Interferometer (IASI); nYurganov and Leifer (2016a). Data key on panel.







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1323

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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. 3c** and

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1340

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