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

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Supplementary Material

S1. Review of Airborne Arctic Methane measurements

CH_4 concentration profiles over the Arctic Ocean were measured on five flights during the HIAPER Pole-to-Pole Observations (HIPPO) campaign [*E A Kort et al*., 2012; *S C Wofsy*, 2011] and produced evidence of sea surface CH_4 emissions from the northern Chukchi and Beaufort Seas in most profiles, up to 82°N. Enhanced concentrations near the sea surface were common over fractured floating ice in sample profiles collected on 2 Nov. 2009, 21 Nov. 2009, and 15 Apr. 2010. On 13 Jan. 2009 and 26 Mar. 2010, when the seasonally highest level of sea-ice coverage occurred, CH_4 emissions were weak or non-existent. Some of the observational variability was correlated with carbon monoxide (CO), indicating terrestrial origin.

The Carbon in Arctic Reservoirs Vulnerability Experiment (CARVE) program sought to quantify Alaskan CO_2 and CH_4 fluxes between the atmosphere and surface terrestrial ecosystems. Intensive aircraft campaigns with ground-based observations were conducted during summer from 2012-2015 [*R Y-W Chang et al*., 2014]. No open ocean measurements were made. Additional Alaskan airborne data were collected summer 2015 (Jun.-Sept.) by the Atmospheric Radiation Measurements V on the North Slope of Alaska (ARM-ACME) project (38 flights, 140 science flight hours), with vertical profile spirals from 150 m to 3 km over Prudhoe Bay, Oliktok Point, Barrow, Atqasuk, Ivotuk, and Toolik Lake. Continuous data on CO_2, CH_4, CO, and nitrous oxide, N_2O, were collected [*S C Biraud*, 2016].

West of Svalbard, an area of known widespread seabed CH_4 seepage aligned along a north-south fault parallel to the coast [*S Mau et al*., 2017; *G K Westbrook et al*., 2008] was the focus of a field airborne campaign June–July 2014 [*C L 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 and was coordinated with oceanographic observations. Seabed CH_4 seepage led to significantly increased seawater CH_4 concentrations. However, no significant atmospheric CH_4 enhancement was observed for the region above the seeps for summer data collected 20 Jun.–1 Aug. 2014 [*C L Myhre et al*., 2016] under mostly light winds.

S2. Satellite Arctic AIRS and IASI Methane Measurement and Validation

A number of current orbital TIR instruments observe CH_4 [*D J Jacob et al*., 2016] including the Tropospheric Emission Spectrometer (TES) [*J Worden et al*., 2012], the Cross-Track Infrared Sounder (CrIS) [*A Gambacorta*, 2013], InfraRed Atmospheric Sounder Interferometer (IASI) [*C Clerbaux et al*., 2009], and the Atmospheric Infrared Sounder (AIRS) [*Hartmut H. Aumann et al*., 2003].

IASI CH_4 validation has been addressed in a number of studies for the lower and mid-upper Arctic troposphere. The EuMetSat IASI instruments are cross-track-scanning Michelson interferometers onboard the MetOp-A and MetOp-B platforms [*C Clerbaux et al*., 2009]. IASI-1 (2007-) and IASI-2 (2013-) follow sun synchronous orbits. Three IASI New Generation instruments [*C Crevoisier et al*., 2014] are planned for launch in 2021, 2028, and 2035 [*IASI-NG*, 2017].

IASI instruments 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 of circular footprints with a nadir spatial resolution of 12 km that is 39×25 km at swath (2400 km) maximum [*C 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, separated by approximately half an orbit, resulting in twice daily, near-global coverage with 29-day revisit. The on-flight noise-equivalent delta temperature at 280K is estimated to be well below 0.1K in the spectral range of interest to CH_4 [*A Razavi et al*., 2009]. IASI has a wide swath with a scan angle of ±48.3°. IASI CH_4 retrieval algorithms are described by *X Xiong et al*. [2013] and *A Gambacorta* [2013].

In the TIR, the AIRS (Atmospheric InfraRed Sounder) mission onboard the Earth Observation Satellite, Aqua satellite [*Hartmut H. Aumann et al.*, 2003] and the EuMetSat IASI-1 mission, on the MetOp-A platform [*C Crevoisier et al.*, 2014] [*C Clerbaux et al.*, 2009] provide long-term arctic CH_4 observations with new IASI instruments planned for launch in 2021, 2028, and 2035 [*IASI-NG*, 2017].

AIRS is a grating diffraction nadir cross-track scanning spectrometer on the Aqua satellite (2002-) that is part of the Earth Observation System [*H.H. Aumann et al.*, 2003]. 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, with a scan angle of ±48.3°. Effective field of view after cloud clearing, is 45 km [*J Susskind et al.*, 2006] and the CH_4 spectral resolution is 1.5 cm^{-1} from the 7.8 μm TIR channel [*Hartmut H. Aumann et al.*, 2003]. Version 6 of AIRS Levels 2 and 3 data are publicly available [*AIRS*, 2016]; see *X 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 are analyzed herein because the AIRS time series is longer than IASI.

AIRS CH_4 validation has been addressed in *X Xiong et al.* [2010], who compared aircraft data taken over Poker Flat, Alaska, and Surgut, Siberia with AIRS CH_4 retrieved profiles. Agreement was within 1.2% with mean measured CH_4 concentration between 300–500 hPa; correlation coefficients were ~0.6-0.7.

IASI validation [*X Xiong et al.*, 2013] over a large area was achieved during a quasi pole-to-pole flight of the National Science Foundation's Gulfstream V aircraft [*S C Wofsy*, 2011]. A bias of nearly -1.74% was found for 374–477 hPa and -0.69% for 596–753 hPa. *L Yurganov et al.* [2016] compared 5-year long IASI data for 0-4 km layer over 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 excursions did not correlate. *L Yurganov et al.* [2016] explained the latter by the observatory's location being near the top of the planetary boundary layer.

S3. Currents

S3.1. Barents and Kara Sea Currents

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, Novaya Zemlya archipelago, beyond which is the Kara Sea; to the west lies the Norwegian Sea. In winter the Barents Sea is partially ice-covered, while it is almost ice-free in the summer

The North Atlantic is a significant source of Arctic Basin water, whose density increases by cooling. Some of this water flows into the Barents Sea, ~2 Sv (1 Sv=10^6 m^3 s^{-1}), varying seasonally [*H Loeng et al.*, 1997] with most returning to the North Atlantic as part of the global thermohaline circulation [*K Aagaard and E C Carmack*, 1989; *E Carmack and F McLaughlin*, 2011; *M Yamamoto-Kawai et al.*, 2008]. *T A McClimans and J H Nilsen* [1993] used a laboratory physical model simulation to duplicate most of the observed regional Barents Sea oceanographic features forced by the densities and volume fluxes of water from the Atlantic (the Norwegian Atlantic Current - NAC and the Norwegian Coastal Current - NCC) and Arctic Basin (Persey Current - PC) and Barents Sea hydrography. Key features produced were the general structure of fronts and major currents, etc., which were obtained without regional atmospheric forcing. This highlights the dominant importance of oceanography rather than meteorology to these features.

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

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

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

Although beyond this study's scope, changes in the NAC/MC flow through the Kola Section relate to larger oceanographic trends. *Ø Skagseth et al*. [2008] found good agreement in the Kola Section temperature trend with the Atlantic Multi-decadal Oscillation (AMO) index. *SST* 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 [*J E Stiansen et al*., 2009, Figs. 2.3.3, 2.3.8].

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

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

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

S3.2. Detailed Kara Sea Currents and Bathymetry

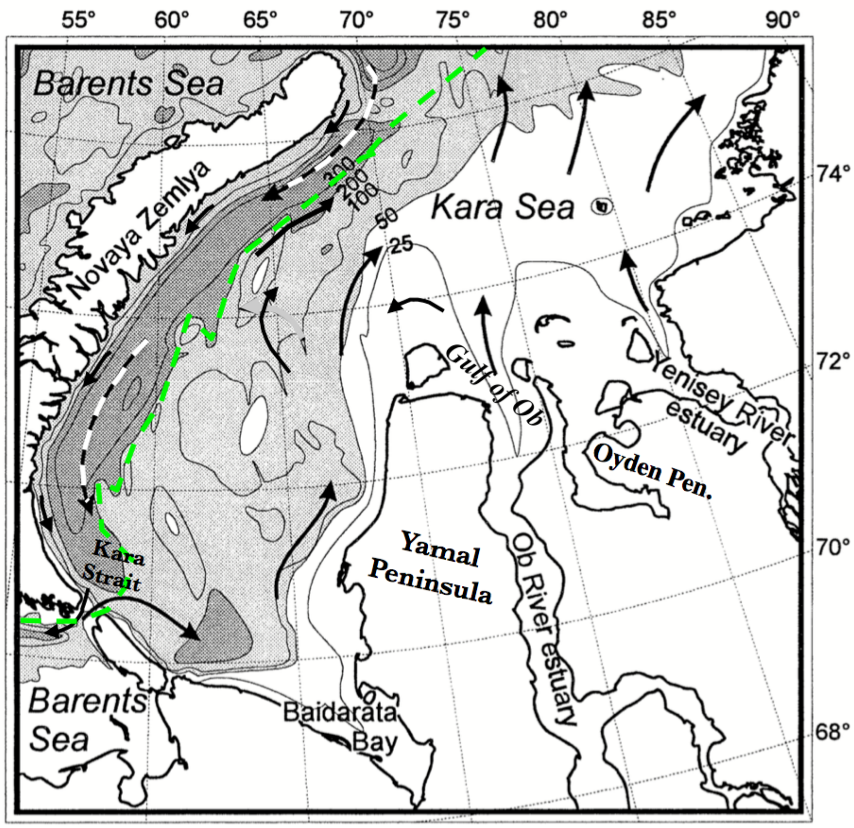

Figure S1. Bathymetry and currents for the Kara Sea. Adapted from *L Polyak et al*. [2002] and *T A McClimans et al*. [2000]. Dashed line indicates subsurface flows. Green line shows approximate edge of submerged permafrost from *T E Osterkamp* [2010].

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^3 yr^{-1}, respectively [*C A 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 [*L Polyak et al*., 2002].

Cold Arctic waters, and ice and melt water from Novaya Zemlya flow southward along the eastern shore of the Novaya Zemlya Archipelago in the narrow, weak Novaya Zemlya Coastal Current (NZCC). Inflow of modified Atlantic water from the Barents Sea (dashed line in **Fig. S1**) accounts for a warm core in the deep Novaya Zemlya Trough [see *T A McClimans et al*., 2000, Section 11]. Part of the NZCC exits through the same Kara Strait that Barents Sea coastal water enters. This, in combination with the rising shallow seabed, causes the Kara Strait to be a site of strong mixing.

Deeper water in the trough is supplied by inflow of modified Atlantic water from the northern Barents Sea. On the surface, inflows to the north Kara Sea come from the MCC, local runoff, and ice in 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 [*T A McClimans et al.*, 1999; *T A McClimans et al.*, 2000].

The Ob and Yenisei Rivers transport significant sediment, underlying the shallowness of the Kara Sea, with extensive proven and proposed petroleum hydrocarbon reservoirs underlying the east and southeast Kara Sea [*P Rekacewicz*, 2005]. Given the Kara Sea's shallowness, CH_4 seep seabed bubbles can mostly transfer their gas directly to the atmosphere [*I Leifer and R Patro*, 2002; *I Leifer et al.*, 2017] and indirectly from wind mixing [*R Wanninkhof and W R McGillis*, 1999], and also from storm sparging [*V D Boitsov et al.*, 2012; *N Shakhova et al.*, 2013], which in the Arctic can extend to 100-200 m depth, i.e., most of the Kara Sea.

S3.3. Detailed West Barents Sea Currents and Bathymetry

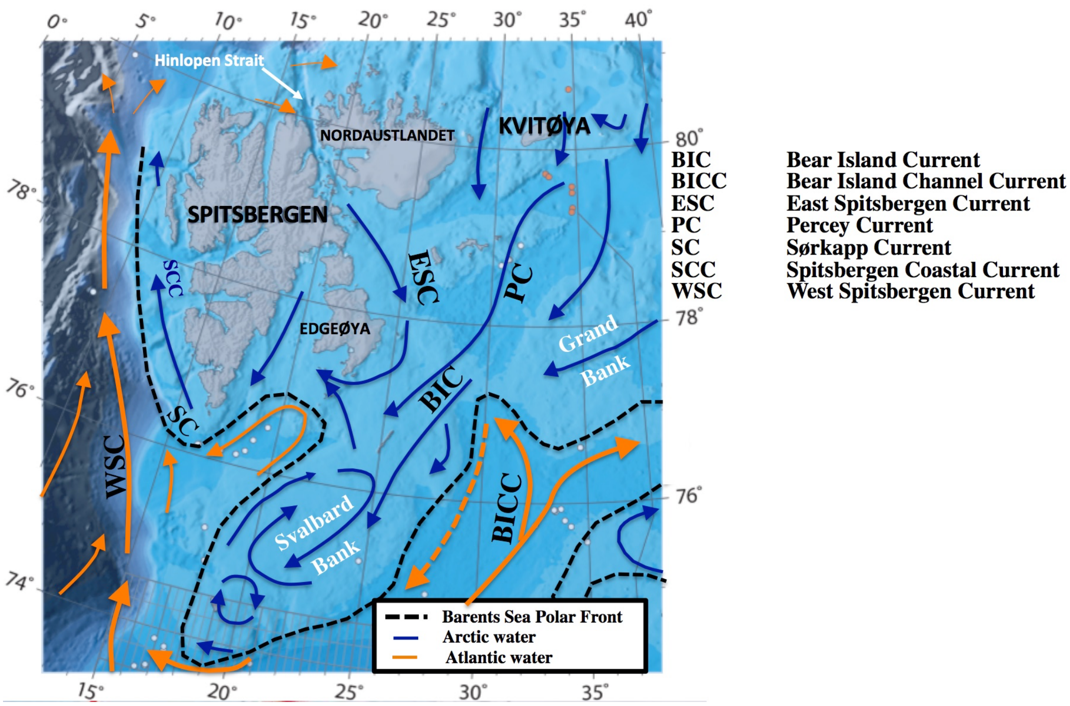

Figure S2. Bathymetry and currents around Svalbard. Bathymetry from *Norwegian Petroleum Directorate* [2016]. Currents from *H Loeng* [1991]. Dashed black line shows location of the Barents Sea Polar Front, Dashed currents are submerged; blue - cold, orange - warm.

Currents and flows around Svalbard Archipelago are complex (**Fig. S2**), dominated by the West Spitsbergen Current (WSC), which is the northerly fork of the Norwegian Atlantic Current (NAC), and flows northwards off the west coast of Spitsbergen. The cold, Percey Current (PC) flows southwest off the eastern shores of the Svalbard Archipelago. The cold East Spitsbergen Current (ESC) flows through the Hinlopen Strait and then joins the PC to flow around the south cape of Spitsbergen as the Sørkapp Current (SC), following the coast northwards as the Spitsbergen Coastal Current (SCC) [*H Svendsen et al.*, 2002]. The cold SCC flows inshore of the WSC, and flows up Svalbard's western coast, inshore and shallower than the warm, Atlantic WSC. The interface between these two currents off west Spitsbergen forms a part of the Barents Sea Polar Front. Thus, coastal waters offshore West Spitsbergen are of Barents Sea / Atlantic water origin, whereas further offshore lies Barents Sea water (origin Atlantic Ocean).

The location of the Barents Sea Polar Front [*L Oziel et al.*, 2016] is semi-permanent and controlled by seabed topography (**Fig. S2**), particularly the Svalbard Bank, the Great Bank, and the trough south of Spitsbergen.

The energy budget of the Barents Sea is driven by Atlantic heat input by the two forks of the NAC (**Fig. S3**) [*V S Lien et al.*, 2013], strongly impacting the Barents Sea *SST* climatology (**Fig. S3**). Along one fork, warmer water flows eastward along the northern Norwegian, Murman, and then western Novaya Zemlya coasts towards the north. The other NAC fork flows northeast along the Svalbard Bank (SB). These flows closely 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 (**Fig. S3a**) and in September in the east Barents Sea (**Fig. S3b**). In June, winds oppose this climatology, i.e., *SST* is most strongly influenced by ocean current transport. In fall, currents and winds are aligned along the Norwegian and Murman and western Novaya Zemlya coasts, reinforcing the transport of heat as indicated in *SST*. Note, though much of the heat that these winds transport originates from the NAC, which maintains Norway at temperatures well above latitudinal averaged. Still, winds cannot explain the spatial distribution of warm *SST*, which extends into the calm around the Central Bank.

Water becomes cooler as it penetrates eastward, and as it reaches the (seasonally varying) ice edge (**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 off western Novaya Zemlya, where the warm NZC flows, water extends further north than elsewhere into areas where winds are from the north (**Fig. 4a**). Moreover, regions with statistically significant warming *SST* trends (*dSST*/*dt*) were in areas of northerly winds both in June and September.

[*J C Comiso et al.*, 2008]

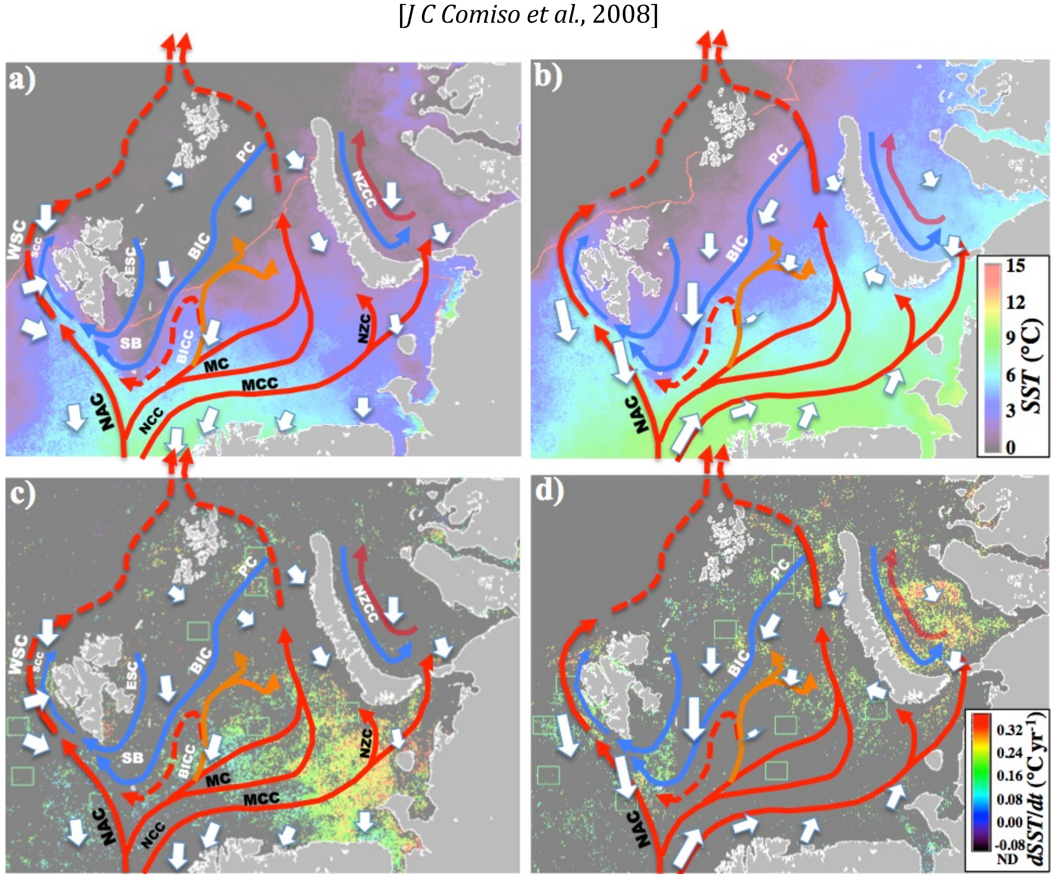

Figure S3. Warm and cold currents (from **Fig. 4a**) superimposed on **a)** June and **b)** September for climatology *SST*, and **c)** June and **d)** September for *dSST*/*dt* trends (ND-no trend detected). The red line shows ice location. Red and blue arrows show warm and cold currents, respectively. Dashed line indicates subsurface flow. Winds (white arrows) are adapted from *E W Kolstad* [2008].

S4. Winds in the Barents and Kara Sea

Accessible meteorological data for the Barents Sea, outside of west Svalbard, whose meteorology and oceanography are affected by the Greenland Sea, are difficult to find, e.g., *V D Boitsov et al*. [2012] for Bear Island, except for sites on the northern Norwegian and Murman coasts. In this regard, the Murmansk airport weather data are the most eastward available long-term data representing southern Barents Sea, coastal meteorology and oceanography. Daily average meteorology data for 2002-2018 were downloaded (https://www.wunderground.com/weather/ru/murmansk) and segregated by month, and found a warming of 0.12°C yr^{-1} in June and 0.11°C yr^{-1} in September. Over this period, winds strengthened slightly (0.0173 m s^{-1} yr^{-1}) with most of the increase in September occurring in 2017 and 2018 (**Fig. S4**). These warming rates are significantly faster than those at Bear Island, which reflects both the greater moderation of the marine rather than coastal atmosphere and the influence of the cold Bear Island Current. Winter temperatures increased even faster.

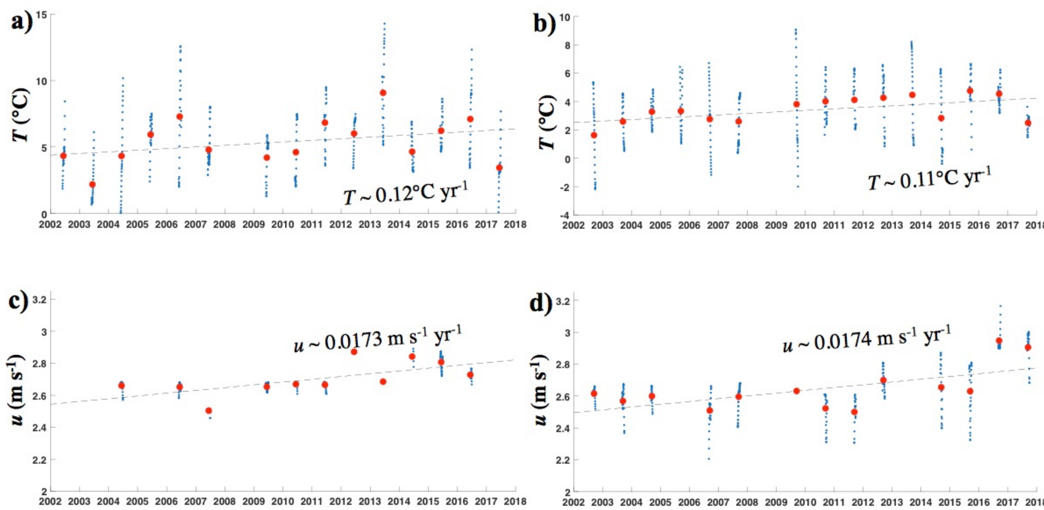

Figure S4. Wind and temperatures for Murmansk airport, Murman, Russia (68.7845°N, 32.7579°E) for **a)** June and **b)** September. Daily-averaged (blue) and monthly-averaged (red) data, and linear polynomial fits (red dashed line) are shown. Data from weatherunderground.com.

S5. Focus Areas

Table S1. Focused study area coordinates

Area	Upper Left	Upper Right	Lower Left	Lower Right
1	79° 16’6.91” N 60° 48’53.42” E	78° 32’12.26” N 62° 49’54.69” E	78° 55’ 5.27” N 57° 43’ 42.58” E	78° 12’27.81” N 59° 52’32.46” E
2	78° 38’25.09” N 55° 34’48.90” E	77° 56’45.71” N 57° 48’15.36” E	78° 14’ 20.21” N 52° 49’ 55.73” E	77° 34’0.24” N 55° 8’13.34” E
3	79° 10’4.24” N 41° 13’50.40” E	78° 36’13.19” N 44° 21’38.03” E	78° 38’ 35.38” N 38° 57’ 26.67” E	78° 6’12.61” N 42° 3’28.98” E
4	79°38’46.04” N 5° 40’51.21” E	79° 31’53.40” N 10° 11’25.49” E	78° 57’ 49.65” N 5° 19’ 46.85” E	78° 51’21.95” N 9° 34’7.45” E
5	78° 8’40.32” N 0° 36’30.89” E	78° 6’24.41” N 4° 35’53.20” E	77° 27’ 29.19” N 0° 34’ 31.46” E	77° 25’20.57” N 4° 20’54.29” E
6	76° 11’22.21” N 1° 16’10.96” E	76° 8’46.20” N 4° 41’44.27” E	75° 30’ 6.27” N 1° 12’35.25” E	75° 27’37.48” N 4° 28’29.63” E
7	74° 48’24.40” N 12° 40’7.63” E	74° 36’9.07” N 15° 40’42.21” E	74° 8’ 1.53” N 12° 7’35.64” E	73° 56’16.20” N 15° 1’6.10” E
8	73° 34’52.37” N 33° 48’43.77” E	73° 6’7.85” N 36° 8’55.31” E	73° 0’12.51” N 32° 31’37.62” E	72° 32’23.29” N 34° 49’20.56” E
9	72° 46’29.04” N 48° 59’20.20” E	72° 8’6.72” N 50° 44’6.03” E	72° 18’49.49” N 47° 18’49.40” E	71° 41’23.16” N 49° 4’27.27” E
10	74° 48’ 6.77” N 38° 38’57.13” Ë	74° 16’3.18” N 41° 0’24.96” E	74° 15’28.49” N 37° 5’34.21” E	73° 44’27.53” N 39° 25’24.39” E

S6. Barents Sea *in situ* data

CO_2 and CH_4 *in situ* data were collected by a Cavity Enhanced Absorption Spectrometer (CEAS), Greenhouse Gas Analyzer (Los Gatos, Research, Mountainview, CA) onboard the *R/V Akademik Fyodorov* during the Nansen and Amundsen Basins Observational System (NABOS) expedition in fall 2013. The *R/V Akademik Fyodorov* is 141-m long with a 25-m beam and 8-m draught. The *R/V Akademik Fyodorov* departed Kirkenes, Norway on 21 Aug. 2013, returning to Kirkenes on 23 Sept. 2013. Analyzer performance information also was recorded for data quality review. Instrument precision was ~1 ppb with a 10 s response time and a 117 s mean layback time. Samples were collected from above the main superstructure, approximately 25 m above the sea surface (**Fig. S4a**), Calibration was daily and used a cylinder standard provided by the Norwegian Air Research Institute (NILU).

The main potential source of ship pollution could be the diesel engine exhaust; however, it appears that the *Akademik Fyodorov*'s engine is not a source of CH_4, with atmospheric CH_4 partially oxidized by the engine leading to exhaust gas having depressed CH_4 compared to ambient air. Data analyzed herein were during steaming transit across the Barents Sea at 26 km hr^{-1}, for which other potential vessel sources, such as the sewage storage venting are not relevant.

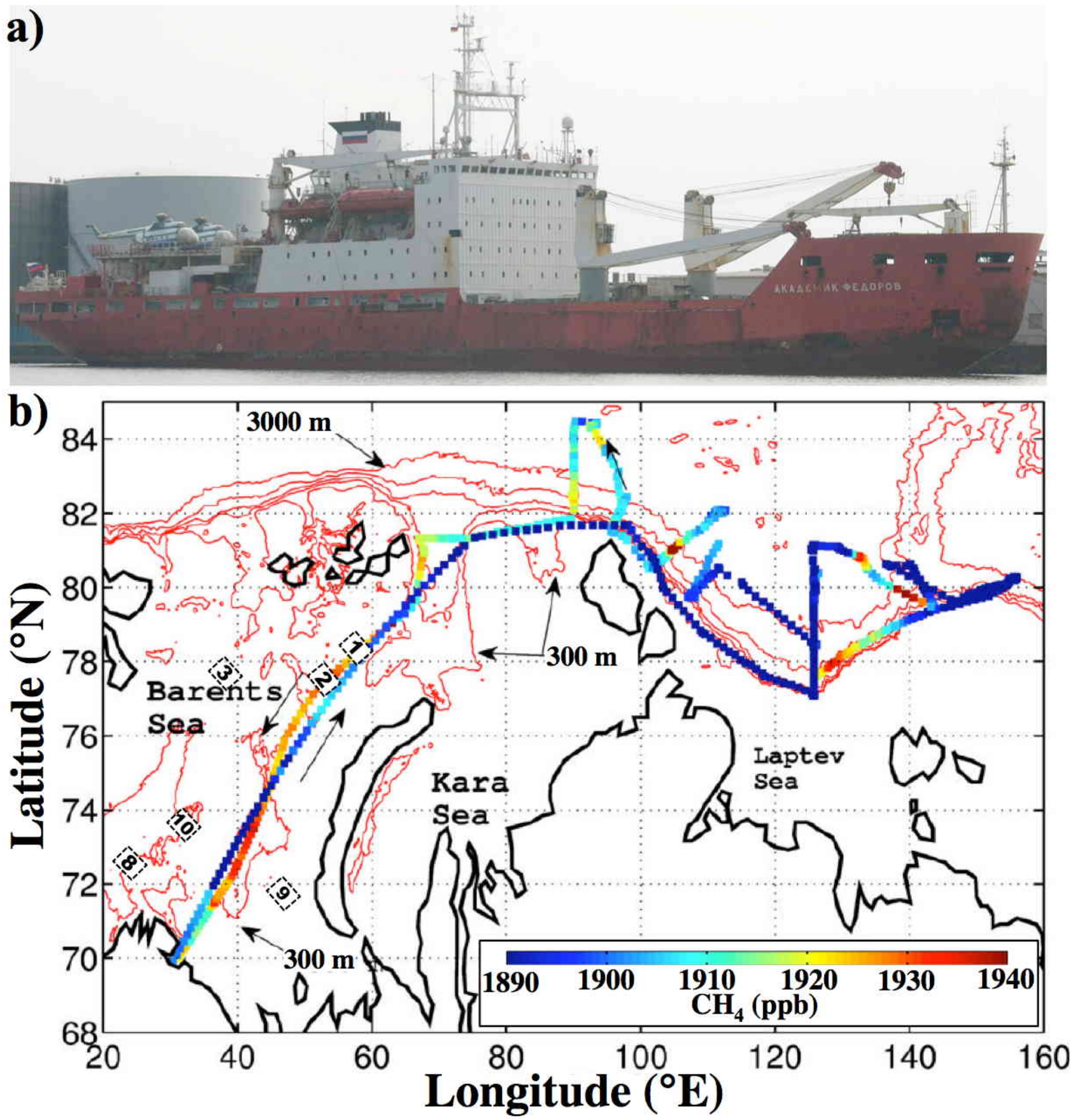

Figure S5. a) Photo of the *R/V Akademic Fyodorov*. **b)** Hourly averaged methane (CH_4) from NABOS expedition. Red shows 300 m depth of the hydrate stability field. Location of focus areas (**Table S1**) shown. Data key on figure.

The month-long data set showed a significant difference between the northwards and southwards transits of the Barents Sea, which were separated by approximately one month and passed directly through Focus areas 1 and 2, as well as between focus areas 9 and 10 in the southeast Barents Sea, approximately along the path of the Murman Current. Most of the CH_4 values in the Laptev Sea were low, although there were several locations of enhanced CH_4.

NABOS values were compared with satellite-retrieved column CH_4 from IASI for 21-24 Aug. 2013 for the northeastwards transit and for 17-22 Sept. 2013 for the southwestwards transit. Agreement between IASI lower tropospheric CH_4 and *in situ* CH_4 for the northwards transit was good, within ~10 ppb, whereas agreement was much poorer for the southwards transit.

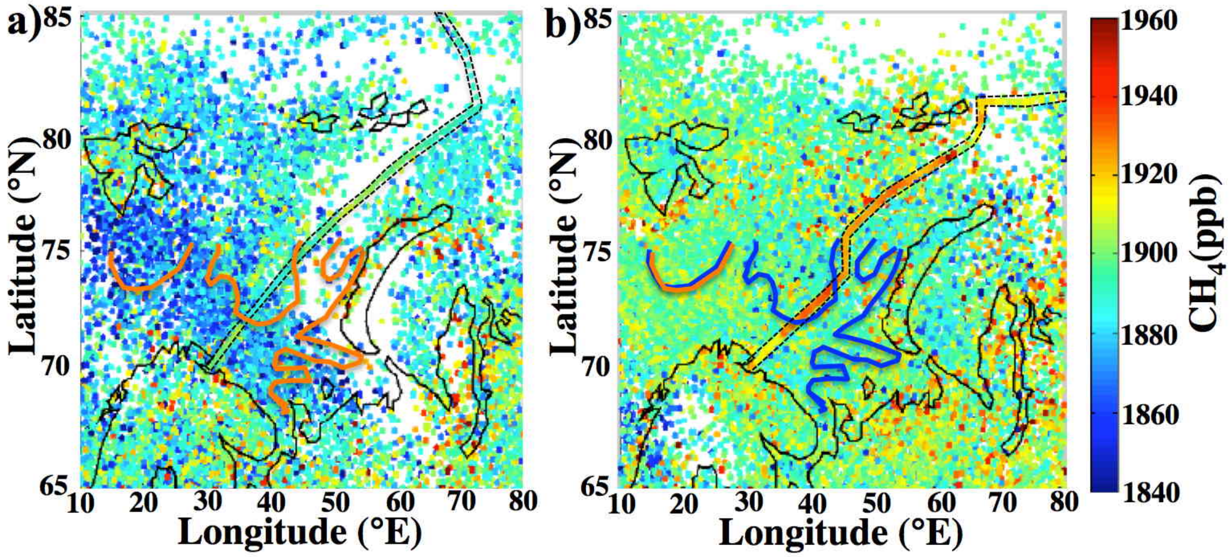

Figure S6. a) IASI retrieved 0-4 km methane (CH_4) for 21-24 Aug. 2013 and hourly CH_4 from the NABOS cruise (outlined in dashed line black). Also shown is the Murman Coastal Current's edges in orange and blue from *A P Alexeev et al*. [2018] and **b)** for 17-22 Sept. 2013. Data key on figure.

S7. Summer month sea surface temperature and methane trends

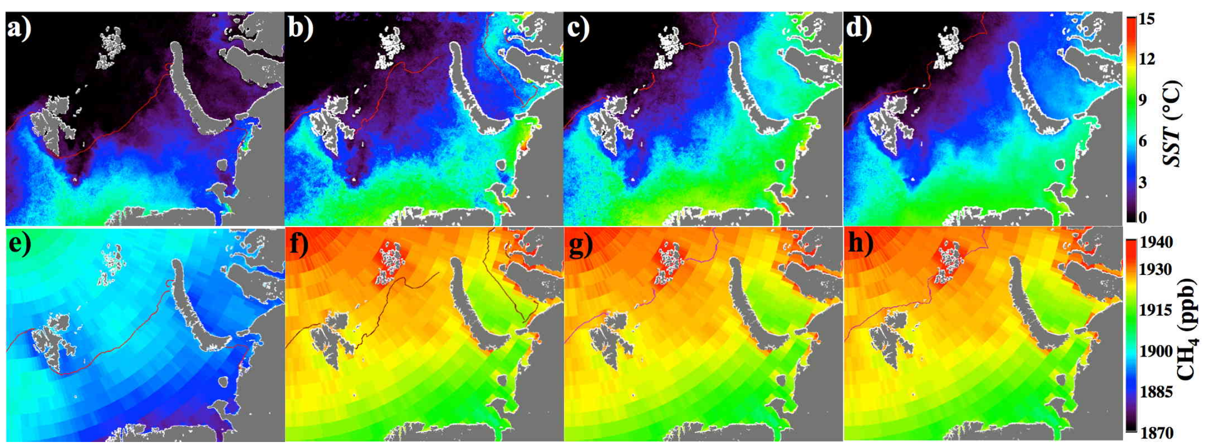

Figure S7. Mean values for 2003 to 2015 of sea surface temperature (*SST*) for **a**) June, **b**) July, **c**) August, and **d**) September. Mean methane (CH_4) concentration for **e**) June, **f**) July, **g**) August, and **h**) September. Median ice edge for same period is shown. Years with reduced ice extent contribute to values of *SST* north of the ice edge. Data keys on figure.

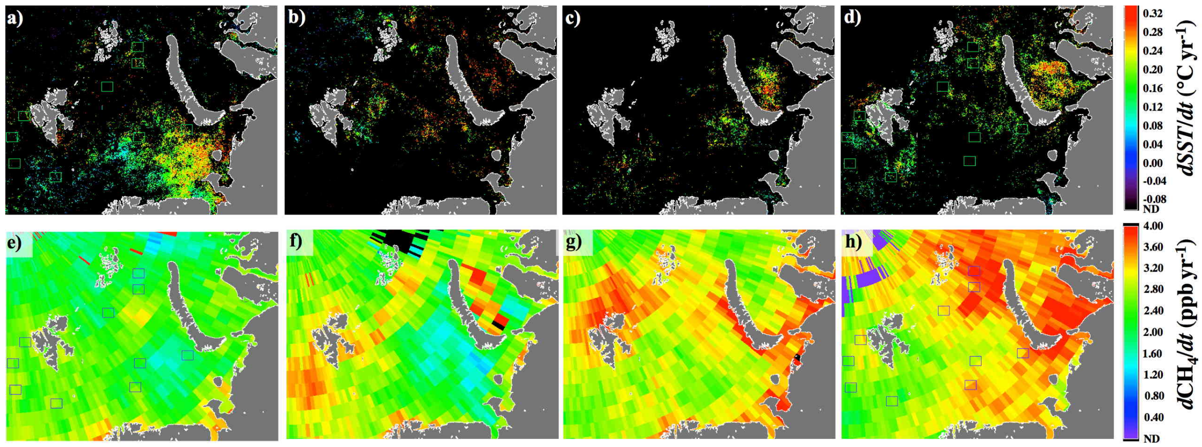

Figure S8. Linear trends for 2003 to 2015 of sea surface temperature (*dSST*/*dt*) for **a**) June, **b**) July, **c**) August, and **d**) September. Methane concentration trend ($d\mathrm{CH_4}/dt$) for **e**) June, **f**) July, **g**) August, and **h**) September. ND – not detectable, i.e., failed statistical test. Blue, black dashed lines shows 100 and 50 m contour, respectively. Data key on figure.

S8. Implications for Svalbard area methane emissions

There are few atmospheric and ocean CH_4 data for the Barents Sea and surrounding areas, the most prominent being associated with CH_4 seepage off Spitsbergen, located immediately south of focus area A4. Studies to date have been in early summer; *S Mau et al.* [2017]; *C L Myhre et al.* [2016] sampled the atmosphere and water column while Westbrook et al. (2009) reported sonar observations of seep bubbles for August-September, and slightly elevated aqueous CH_4 in surface waters immediately above the bubble plumes. All concluded that transport to the atmosphere was not significant, attributed to trapping dissolved CH_4 below the pycnocline. It is important to note that with respect to the overall Barents Sea area CH_4 anomaly, the Svalbard area is far less important than waters around Franz Josef Land, off the west coast of Novaya Zemlya, and the north-central Barents Sea (**Fig. 9**).

Both *SST* and CH_4 in June (**Fig. 9**) and July (**Supp. Fig. S7**) show that much of the active seepage in west Spitsbergen show that much of the area of active seepage was inshore of the Barents Sea Polar Front, and thus under the cooling Arctic waters of the Spitsbergen Coastal Current (SCC), supported by reported salinity data (Mau et al., 2017). Although *SST* remains low 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 *G K Westbrook et al.* [2009].

Although the studies indicate these seeps do not contribute to summer atmospheric CH_4, they did not consider methane shoaling, which would allow seabed CH_4 to reach the atmosphere far downstream. Interestingly, Mau et al. (2017; Fig. 3) show data that could be interpreted as methane shoaling with elevated aqueous CH_4 forced shallower by the north-flowing SCC, crossing subterranean ridges. Focus area A4 shows the strongest increase in CH_4 from 2005-and an increasing *SST* over this time period, consistent with shoaling. Stronger enhancement of CH_4 growth is observed north of Spitsbergen in June (**Fig. 10c**), 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 Current (SCC) that would be CH_4 enriched from seabed seepage, and then flows over relatively shallow seabed towards the Hinlopen Strait. Thus, there is evidence of increasing downstream CH_4 transport to the atmosphere downcurrent of seepage off West Spitsbergen after methane shoaling, albeit not significant to overall Barents Sea emissions.

There is evidence of acceleration in the CH_4 growth nearshore off West Spitsbergen in June, but not in September (**Fig. 10d**) when CH_4 growth enhancement 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.

S9. Arctic Methane Movie

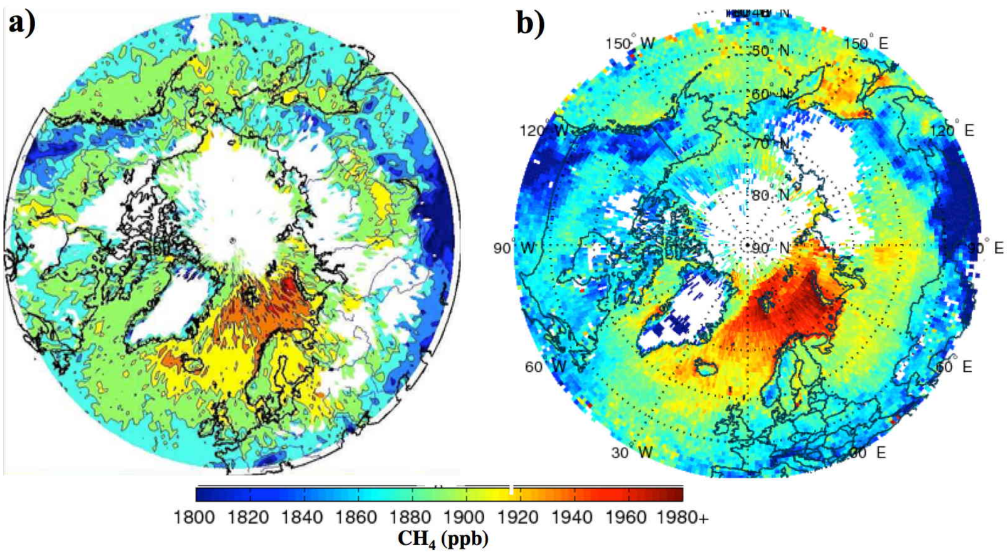

Figure S9. IASI Arctic methane (CH_4) for the lower 4 km for a) March 2012 and b) March 2018.

A movie of Arctic CH_4 from 2012 every 5 days shows a range of variations on a range of different spatial and temporal scales (**Fig. S9**). Strong enhancements are observed that persist in regions for a few days–most likely related to synoptic system flushing in fall to spring. The seasonal variation is easily observed, with highest values often in November and December. In late winter and early spring, large CH_4 anomalies are observed in some years at the ice edge.

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