SUPPLEMENTARY INFORMATION

1. FACTORS AFFECTING GAS TRANSPORT IN SUBSEA PERMAFROST

Even if thawing is the most considered parameter known to affect the continuity and permeability of permafrost hence allowing the migration of gas from deeper layer to the surface, other important factors need to be taken into consideration as discussed below.

1.1. Physical factors

When sediments freeze, a certain amount of pore water (depending on water salinity, type of sediments and temperature) remains unfrozen within frozen sediments. The physical properties of frozen grounds/sediments may be influenced significantly by this unfrozen water. For example, when temperature changes from –6.5 to –2.8°C, the content of unfrozen water increases by 22.3% (Khimenkov and Brushkov, 2006). It was shown that within frozen mineralized coarse sands, unfrozen water accumulates in the center of the pore space, forming a net of fissures, building an efficient transportation system within frozen sediments (Arenson and Sego, 2006) This system enables the movement of gaseous and dissolved CH_4 inside the frozen permafrost (predominantly consisted of sandstone and gravel) as it has been demonstrated on Barrow Point (Alaska)(McCarthy et al., 2004).

On a larger scale, the hydraulic system also plays an important role. It represents the layers of mineralized water incorporated above, within and beneath permafrost – so called supra-permafrost, intra-permafrost and sub-permafrost ground water, respectively. The salinity of this cryogenic groundwater usually ranges between 10 and 300psu, in which freezing is prevented by freezing-point depression due to the dissolved-solids content of the pore water (Gilichinsky et al., 2007)in most cases very high as it often includes brines resultant from the freezing of marine sediments. These water layers are usually connected to each other to build a multi-level transport system and allowing gases and geo-fluids to migrate upwards and to be released to the water column through the subsea permafrost (Biggar et al., 1998). The hydraulic system serves as well to connect the deep permafrost to the surface enhancing the development of taliks with a network of mechanical discontinuities existing within the permafrost into an integrated transportation system for ascending gases and geofluids (Makogon et al., 2007).

1.2. Seasonal variations

Since seasonal variations in temperature in Arctic regions are very large, they affect frozen soils/sediments by causing alternations of compression and stretching of frozen grounds. This leads, with time, to the development of numerous cracks filled with water and it results in the formation of cuneiform (wedge-like) shapes of ice inclusions in permafrost body. This mechanism was suggested to be responsible for the observed predominance of wedge-like type of ice within permafrost body widely spread on the Arctic coasts. Such an extensive network of cracks in the structure of frozen rocks serves as well to provide gas migration paths for ascending gases and geofluids (Romanovskii et al., 2000).

1.3. Destabilization of gas hydrate deposits

Another group of factors influencing gas transport within subsea permafrost can be associated to the destabilization of gas hydrates deposits (e.g. Clenell et al., 1999, Romanovaskii et al., 2005, Nicolsky et al., 2012). The thermal and pressure conditions determining the stability/instability of the gas hydrates are very specific (Shakhova et al., 2009). When shallow shelf hydrates destabilize, overly pressured gas from the decaying hydrates accumulates between the lower boundary of permafrost and the upper boundary of hydrate stability (Naudts et al., 2006, Sergienko et al., 2012). This over-pressurized gas front moves both horizontally and vertically following the unconformities and discontinuities of the permafrost body (Cramer et al., 2005). Within developing taliks, this front can also constitute gas migration pathways of high capacity (Osterkamp et al., 1985, Frederick et al., 2014).

2. DEEP CORE LITHOLOGY

The major differences in the lithology of the deep cores drilled in 2011 and 2013 are the thickness and the origin of the Holocene age sediment. In the background core, Holocene age marine sediments compose the upper 5.5m, they represent disperse pelite-aleurite deposits predominantly of alluvial origin, which are accumulations of river-derived matter formed in coastal marine conditions with high rates of sediment accumulation (Fig.S1). Holocene sediments are underlain with terrestrial accumulations (5.8m to 52.3m) of late Pleistocene age, which are represented by consolidated aleuro-sands inter-layered by fine-grained aleurite accumulations with inclusions of pebbles and wood remains.

The other deep cores were drilled in 2013 near the Muostakh Island. In the IID-13 and IIID-13 cores, the Holocene age sediments represent only the upper 0.5m and consist of remains of the coastal ice-complex (IC or Yedoma) of Muostakh Island. This area represents a former part of the coastal alluvial plain, upper part of which is composed of IC that thaws very fast during the last century. Sediment morphology reflects the nature of the sediments: fine-grained sand-aleurite-pelite is interlayered with gravel-pebble material with inclusions of wood remains and plant debris.

Sediment core VD-13 stands out of all the other cores drilled in 2013. Its morphological structure is different as its frozen fraction is presented by sands interlayered by gravel-pebble accumulations. It is known that coarse sands even frozen remain permeable for gases as within their frozen structure fissures form that allow pore water to remain unfrozen; these fissures were suggested to be serving as gas migration paths (Arenson and Sego, 2006) as discussed in section 1.1. Note, that other drilling data obtained in the study area have shown that at water depth ~ 11m, the upper

57m sediment is entirely unfrozen with typical temperature about 0 degC (Shakhova et al., 2014). Downward extrapolation of the temperature curve together with permafrost modeling show the existence of open taliks in the Buor Khaya Bay where all samples were taken. Similar field and modeling results were obtained for the Dmitry Laptev Strait (Shakhova et al., 2009; Nicolsky and Shakhova, 2010). This means that hydrocarbons from deeper strata could migrate through these migration paths.

3. INTERPRETATION OF THE ^{14}C-CH_4 RESULTS

The observation of unexpectedly high ^{14}C values for the ID-11 background core and water samples from the SE needs further discussion. As explained in the main text, ^{14}C values >200pmC do not exist in nature for any carbonaceous material including CH_4, even not for the peak of surface nuclear bomb tests of the mid-20th century. We assume that a local anthropogenic nuclear contribution is the most likely explanation for our elevated radiocarbon levels, which is justified in this section.

In the ID-11 background sediment, the higher ^{14}C values correspond to the lower CH_4 concentrations. That implies a possible mixture between an old CH_4 source and a background highly enriched in ^{14}C. A Keeling plot shows that the highly enriched ^{14}C contribution is relatively small in terms of CH_4 quantity and that the main CH_4 substrate is relatively old (Fig. S2). For the hotspot sites, where CH_4 concentrations are larger, no mixture with a “younger” source is identified. All data points are showing very low ^{14}C (<1.5pmC) so the main CH_4 substrate at these sites is clearly of Pleistocene age. Note that all points of the IID-13 core were below the analytical detection limit of 0.8pmC hence no conclusions could be drawn from the Keeling plot of this core.

The very high ^{14}C values >200pmC may either originate from “in-situ” cosmogenic or nuclear production of radioactive CH_4 or its substrate. Enhanced ^{14}C has been found in meteorites (Firemann, 1978) and can be produced at the surface of ice sheets (Baudin et al., 1973), but in both cases, the quantity of ^{14}C formed is very small compare to what we observed in the BKB and SE sediment and water samples. Nuclear production of ^{14}C involves formation by neutron activation as consequence of a nuclear chain reaction, which may either take place naturally or artificially. The only place on Earth, where nuclear fission has occurred naturally, was reported to be occurring about 1.7 billion years ago in Oklo, Gabon (Nuclear Wastes in the Arctic report, 1995). However, such natural reactors cannot be active anymore today, as the relative abundance of fissile ^{235}U has now decayed below that required threshold for a sustainable nuclear reaction chain.

The Arctic Ocean has been used as a disposal area for radioactive wastes (Nuclear Wastes in the Arctic report, 1995, Johnson-Pyrtle and Scott, 1991) hence we believe that anthropogenic nuclear contamination is the most likely explanation for these ^{14}C-enriched CH_4 background contribution. Similar cases but with slightly lower values have been observed in gas samples from marine basins along the Californian coast (Kessler et al., 2008). The authors assumed nuclear plant effluents as being the most likely explanation for these unexpected data.

We exclude a possible contamination during sampling, extraction and analysis, because no radioactive tracers were used during the sampling expeditions. The samples affected by enriched ^{14}C values were not sampled in a similar manner. The sediment samples were drilled from the ice in 2011 while the SE water samples were sampled in 2012 from a ship together with other water and surface sediment samples showing no enrichment in ^{14}C values. For the rest of the sampling and analysis process, all samples were handled in a similar way and measured in a random order, but only samples from these two specific locations show highly enriched ^{14}C values. None of the reference and blank measurements was abnormal either.

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SUPLEMENTARY FIGURES

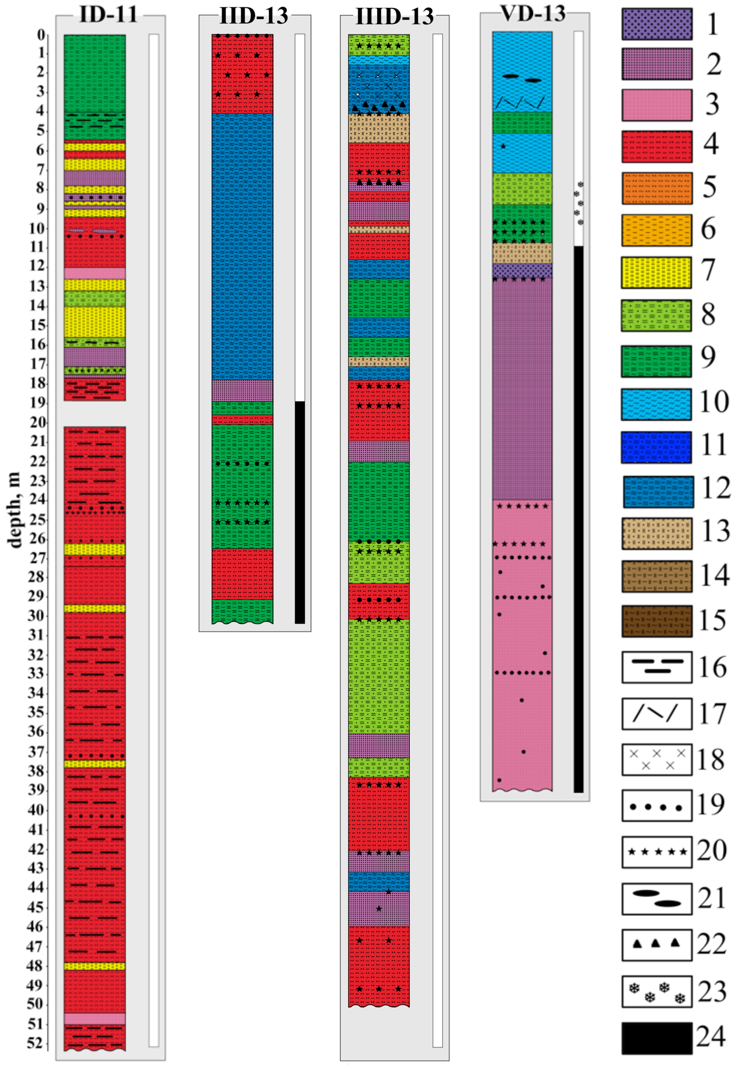

Figure S1. Morphological structure of the sediment cores extracted from boreholes drilled in 2011 (ID-11) and 2013 (IID-13, IIID-13 and VD-13). Lithological structure: 1 – coarse sand, 2- medium-grained sand, 3 – fine-grained sand, 4 – aleuro-sand, .5 –pelite-sand, 6 – coarse-grained aleurite, 7 – fine-grained aleurite, 8 – sand-aleurite, 9 – pelite-aleurite 10 – pelite, 11 – sand-pelite, 12 – aleuro-pelite, 13 – sand-mictite, 14 – aleuro-mictite, 15 – pelite-mictite; Texture characteristics: 16 – horizontal layered texture, 17 – vertical fine banded texture, 21 – horizontal lense-like texture; Diagenesis characteristics: 18 – H_2S presence; Inclusions: 19 – gravel-pebble products, 20 – plant debris, 22 – shell detritus; Cryogenic structure: 23 – ice-lenses/lenticular-layered cryostructure, 24 – frozen ground with massive (in sands) and micro-lenticular cryostructures (shown vertically along the cores).

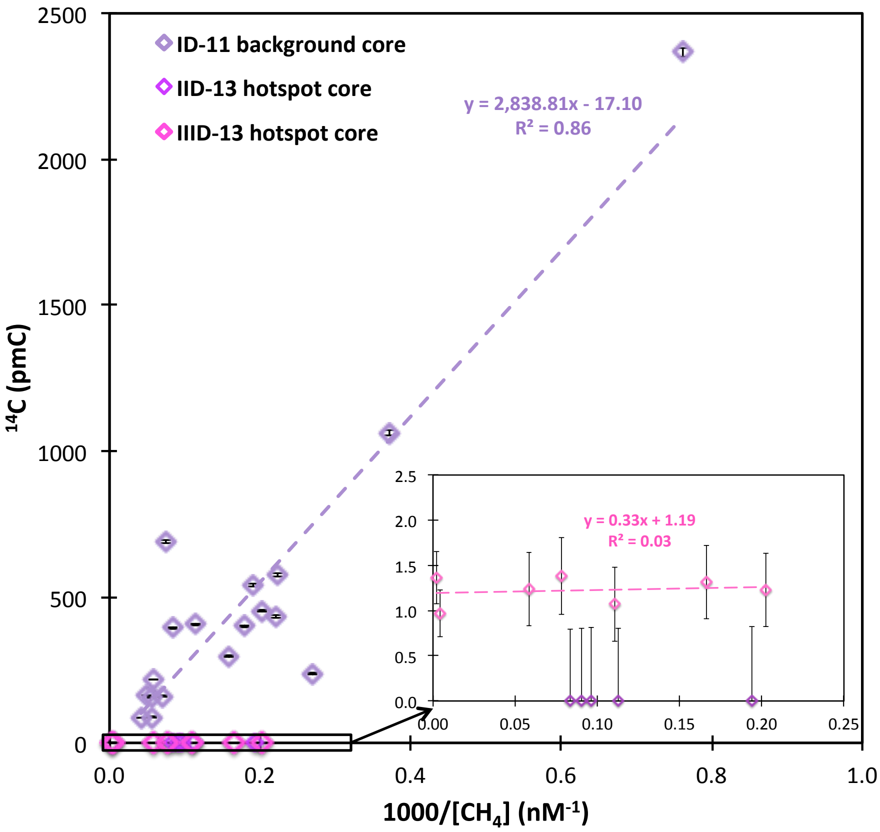

Figure S2: Keeling plot: inverse CH_4 concentration versus ^{14}C data for sediment samples in the partially thawed subsea permafrost. The diamonds are "deep" core sediment data and the dashed lines represent the linear regressions for the ID-11 (purple) and IIID-13 (pink) cores. All values of the IID-13 core are close to zero so no linear regression line is depicted for this core. The intersections with the y-axis correspond to the ^{14}C pmC values of the main CH_4 substrate.

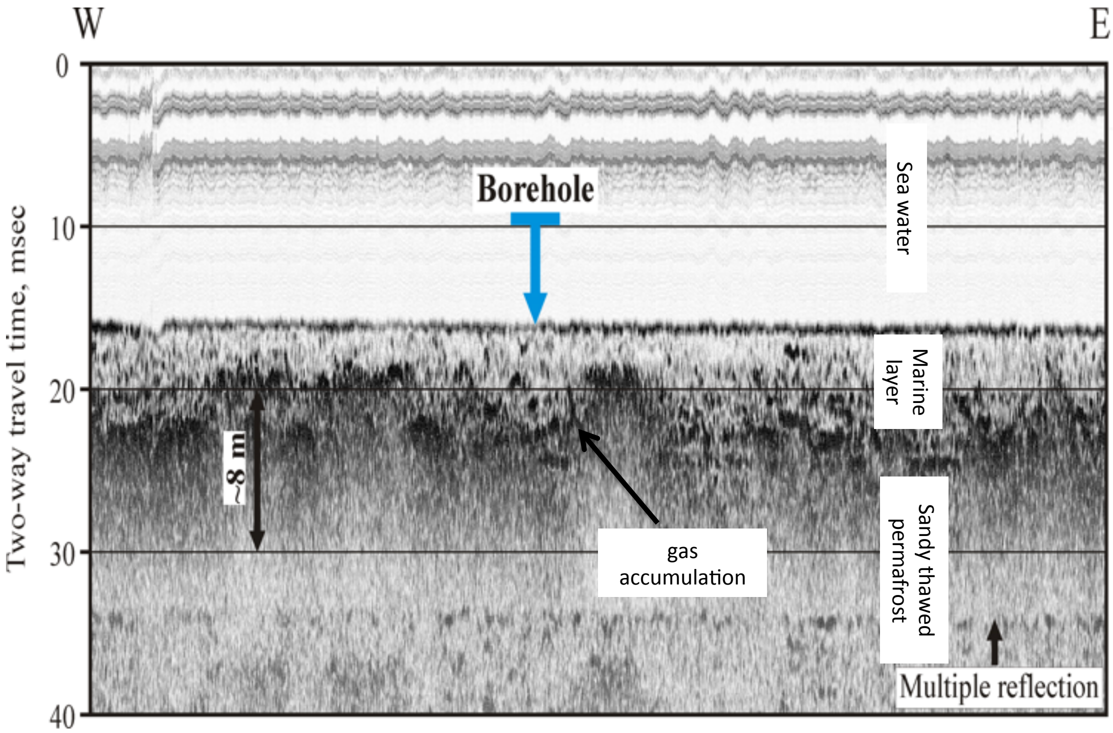

Figure S3: Acoustic profile of the borehole of the ID-11 drilling site. Darker areas represent changes in density between the different horizontal layers (Sergienko et al., 2012).