On carbon transport and fate in the East Siberian Arctic land–shelf–atmosphere system

Igor P Semiletov1,2, Natalia E Shakhova1,2, Valentin I Sergienko3, Irina I Pipko2 and Oleg V Dudarev2

1 International Arctic Research Center (IARC), University of Alaska Fairbanks (UAF), Fairbanks, AK, USA
2 V I Il’ichev Pacific Oceanological Institute (POI), Far Eastern Branch of the Russian Academy of Sciences (FEBRAS), Vladivostok, Russia
3 Headquarters of the Far Eastern Branch of the Russian Academy of Sciences (FEBRAS), Vladivostok, Russia

E-mail: igorsm@iarc.uaf.edu

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Abstract
This review paper summarizes current understanding of the transport of organic carbon to, and the fate of organic carbon within, the East Siberian Arctic Shelf (ESAS), and of processes determining carbon dioxide (CO2) and methane (CH4) fluxes from the ESAS to the atmosphere achieved from analyzing the data sets obtained on 20 expeditions performed from 1999 to 2011. This study of the ESAS was aimed at investigating how redistribution of old carbon from degrading terrestrial and sub-sea permafrost and from coastal erosion contributes to the carbon pool of the ESAS, how changes in the hydrological cycle of the surrounding land and alteration of terrestrial carbon cycles affect the hydrological and biogeochemical parameters of shelf water masses, and which factors control CH4 and CO2 emissions from the ESAS. This report describes selected results achieved by a developing international scientific partnership that has been crucial at every stage of the study and will be even more important in the future.

Keywords: carbon, permafrost, Arctic Ocean, carbon dioxide, methane, sediments

1. Introduction
The Arctic Ocean is surrounded by permafrost, which is being degraded at an increasing rate under conditions of warming which are most pronounced in Siberia and Alaska (ACIA 2004). The Arctic region contains a huge amount of organic carbon (OC) buried inland and within the Arctic Ocean sedimentary basin (‘Arctic carbon hyper pool’, Gramberg et al 1983), which could become involved in current biogeochemical cycling due to thawing of on-land and sub-sea permafrost (defined by the International Permafrost Association as ground that remains at 0°C or below for at least two consecutive years), increasing coastal and bottom erosion, accelerating river discharge and carbon losses from soils (Semiletov et al 2000, Freeman et al 2001). Thaw and release of OC from Arctic permafrost is postulated to be one of the most powerful mechanisms causing a net redistribution of carbon from land and ocean to the atmosphere (Kelley and Gosink 1979, Oechel et al 1993, Zimov et al 1993, 1996, 1997, Semiletov 1999a, 1999b, ACIA 2004, Gruber et al 2004, IPCC 2007, Canadell and Raupach 2009). Determining the magnitude of particulate and dissolved fluxes of modern and old OC and other terrestrial
material from land is critical to constraining a range of issues in the Arctic shelf-basin system, including carbon cycling, the health of the ecosystem, and the interpretation of sediment records. More than 90% of all OC burial in the Arctic Ocean occurs in sediments deposited on deltas, continental shelves, and upper continental slopes, of which the East Siberian Arctic shelf (ESAS) represents a significant fraction (Gramberg et al. 1983, Vetrov and Romankevich 2004).

The shallow ESAS is a unique area of the World Ocean where ~80% of predicted sub-sea permafrost exists (figure 1) (ACIA 2004). Sub-sea permafrost is much more vulnerable than its terrestrial counterpart, because it experiences a drastic change in its thermal regime due to inundation by the ocean and consequent warming by as much as 12–17°C, prior to the current ongoing climate change (Soloviev et al. 1987). It was therefore hypothesized that combined bottom-up geothermal and top-down seawater heat fluxes, possibly accelerated by amplified Arctic warming, have led to partial destabilization of sub-sea permafrost, causing methane (CH$_4$) production within taliks (a term describing a thaw column or a thaw bulb of sediments within the permafrost) and release from seabed deposits that vent extensively to the Arctic atmosphere (Shakhova et al. 2010a, 2010b, 2010c, Nicolsky and Shakhova 2010).

Besides mobilization of old OC stored in sub-sea permafrost, there are terrestrial sources of OC delivered to the ESAS. It is highly likely that the Great Siberian Arctic Rivers (GSARs; Ob, Yenisey, Lena, Kolyma and Indigirka rivers) carry and bring to the shelf an integrated signal of terrestrial organic material (OM) released from the soil and the degrading terrestrial permafrost within their vast watersheds (Guo et al. 2004, Dudarev et al. 2006a, Alling et al. 2010, Sánchez-García et al. 2011). The ESAS acts as a Lena, Kolyma and Indigirka estuary; thus, it accepts the integrated signal and transfers it further via the signal carrier, which is shelf water, to the Arctic Ocean (Semiletov et al. 2000, 2007, 2011, van Dongen et al. 2008, Pipko et al.
Figure 2. Process of coastal erosion as it is seen on the northern cape of Muostakh Island (September 2006, photo by I P Semiletov). Rates of coastal erosion can be as high as 20–30 m in two weeks.

2010, 2011, Vonk et al 2010, Charkin et al 2011, Gustafsson et al 2011, Karlsson et al 2011). Another significant source of terrestrial carbon delivered to the ESAS is coastal erosion; on the shores of the ESAS the rates of erosion are the greatest on the globe and can reach up to 80 m yr$^{-1}$ (figure 2; Grigoriev 1993, Are 1999, Rachold et al 2000). The latest estimate produced a value of 4 Tg C-OC released from the coastal ice complex into the ocean annually (Grigoriev et al 2006).

Results of initial offshore studies, performed in the 1990s, demonstrated that the biogeochemical consequences of terrestrial OM transformations within the ESAS could play a significant role in the Arctic marine carbon cycle (Semiletov et al 1994, 1996a, 1996b, Semiletov 1999a, 1999b). Further studies revealed that the eroded terrestrial OM is available for microbial oxidation; this oxidation is followed by a significant carbon dioxide (CO$_2$) buildup in the water column, and consequent CO$_2$ release into the atmosphere (Pipko et al 2005, 2008, 2011, Repina et al 2007, Semiletov and Pipko 2007, Semiletov et al 2007, Anderson et al 2009, 2011). Partitioning between the riverine and eroded terrestrial OM exported onto the shelf is still under discussion (Rachold et al 2000, Dudarev et al 2003, 2006b, Vetrov and Romankevich 2004, Semiletov et al 2005, 2011, Macdonald et al 2008, Vetrov et al 2008, Vonk et al 2010, Charkin et al 2011, Gustafsson et al 2011). Results presented in the current letter serve to contribute to this discussion.

Numerous important and newly emerging problems with potential influence on climate change arise from land–shelf interaction in the ESAS. Addressing these problems requires long-term international cooperation between Russia, to whom the ESAS belongs, and other countries. This brief overview describes the state of progress in establishing such cooperation by reporting results of field research accomplished during 1999–2011 in the ESAS. The objectives of this study were pursued in order to identify and quantify the main processes responsible for carbon cycling in the ESAS land–shelf–atmosphere system and factors altering them. These objectives included the following.

- To investigate how redistribution of old carbon from degrading terrestrial and sub-sea permafrost and from coastal erosion contributes to the carbon pool of the ESAS.
- To study how changes in the hydrological cycle of the surrounding land and alteration of terrestrial carbon cycles contribute to formation and propagation of halocline waters and affect the hydrological and biogeochemical parameters of shelf water masses, and to quantify the area-scaled ESAS contribution of CH$_4$ and CO$_2$ to the atmosphere.
- To define specific factors which control CH$_4$ emission to the ESAS atmosphere in order to develop a conceptual model of CH$_4$ propagation from the seabed to the atmosphere, and to assess the strength, type, and dynamics of the source.

2. Fieldwork and research platforms

The location of the main ESAS studies conducted from 2003 to 2009 is shown in figure 3. Vessels of different sizes, from small 15 m boats to mid-size ~70 m hydrographic vessels and heavy diesel ice-breakers, were employed for the summer–fall work. The most comprehensive field research was accomplished during the 2008 International Siberian Shelf Study (ISSS) expedition. This cruise was initiated to alleviate the scarcity of observational data describing the transport and processing of water, sediment and carbon in the ESAS (Semiletov and Gustafsson 2009).
Figure 3. Location of the ISSS (2003–9). More than 1100 hydrological stations, more than 700 sediment coring and grab sites, and four coastal erosion study areas were investigated (2003–7, white circles; 2008, yellow triangles; Nansen and Amundsen Basin Observational System (NABOS) 2009, magenta circles; TB-012 2009, red circles).

The interlinked research themes such as biogeochemistry, geophysics/seismicity, marine chemistry, CH$_4$ dynamics, physical oceanography and sedimentology guided research onboard the ice-strengthened hydrographic vessel Yakov Smirnitsky and two small vessels, the TB 0012 and the Orlan (Semiletov and Gustafsson 2009).

Additional biogeochemical work was done onboard the commercial ice-breaker Kapitan Dranytsin in cooperation with the Nansen and Amundsen Basin Observational System (NABOS) project (www.iarc.uaf.edu/nabos) and onboard the passenger vessel Moskovsky-11, which was utilized for water sampling and taking onboard measurements from the mid-stream to the mouth of the Lena River (for details see Semiletov and Gustafsson 2009). In the winters of 2002, 2007 and 2011 we used a transport caravan and cross-country vehicles provided by Tiksi Hydrobase of the Northern Sea Route service. Both summer and winter helicopter surveys to study the short-term spatial–temporal variability and vertical distribution of the three main greenhouse gases (CH$_4$, CO$_2$ and water vapor) and the thickness of the meteorological boundary layer were conducted in 2006, 2010 and 2011. More than 20 expeditions were accomplished during the last 10 years.

Since 1999 the parameters of the carbonate system have been measured onboard; since 1999 stable C, N, O and H isotopes in the particulate OC (POC) and sediment OM have been measured in the lab. From summer 1999 to summer 2002 we utilized techniques and equipment described in detail in Semiletov et al (1996a, 1996b, 2005, 2007), Semiletov (1999b), Pipko et al (2002, 2011). Since summer 2003 our field activities (figure 3) have included making ‘traditional’ measurements such as in situ temperature, conductivity (salinity), colored dissolved OM, turbidity and photosynthetically active radiation; these parameters were measured using the Seabird SBE 19plus Profiler. Utilizing the same instruments and analyses across years improves our understanding of linkages between various physical and biogeochemical processes and their inter-annual variability in the Arctic land–shelf system, which is an important objective for the ESAS carbon study. All the methods have been described in detail in dedicated papers cited throughout this letter.

3. Selected results

3.1. Western and eastern geochemical provinces of the ESAS

A notable characteristic of the ESAS is large gradients of hydrological and biogeochemical parameters that correspond to geographically critical contrasts in the Arctic system where the Pacific and local shelf waters interact over the shelf. That is reflected in the sedimentation regime (Semiletov et al 2000, 2005, van Dongen et al 2008, Gustafsson et al 2011).

The distribution of $\delta^{13}$C-OC in the surface bottom sediments was used to distinguish between the two geochemical provinces, western and eastern, in the ESAS (Semiletov et al 2005). This approach was later confirmed with more detailed considerations using biomarkers, isotopes, and C/N ratios (Vetrov et al 2008, Vonk et al 2010). The western geochemical province extends from Vilkitsky Strait ($\sim$120E) in the west to Cape Chukochiy (roughly 172E) in the east, where it is approximately bounded by the isolines of OC and nitrogen isotope ratios: $\delta^{13}$C$_{\text{org}} = -24.5\%$ and $\delta^{15}$N$_{\text{org}} = 7\%$. The western province is strongly influenced by particulate material (PM) transport induced by coastal erosion and fresh water (FW) transport by Lena runoff. PM concentrations in the western province are as much as ten times higher than in the eastern geochemical province, which extends from Cape Chukochiy to the Bering Strait and is highly affected by Pacific inflow (Semiletov et al 2005).

Recent studies indicate a connection among the atmospheric circulation and the hydrological and geochemical characteristics of the water masses in this region (Semiletov et al 2005, Anderson et al 2009, 2011, Porcelli et al 2009).
Multi-year variability of the carbonate system and dissolved CH	extsubscript{4} dynamics and efflux of CO	extsubscript{2} into the atmosphere confirm the significance of changing atmospheric circulation patterns (Pipko et al 2005, 2008, 2011, Shakhova and Semiletov 2007, 2008). An important effect on distribution of Atlantic and Pacific waters in the upper Arctic Ocean could be exerted by a change in direction of water transport associated with a shift from the cyclonic (Zn) mode to the anticyclonic (Az) mode of atmospheric circulation over the Arctic basin. Under Az mode conditions the trans-Arctic current (which transports FW northward from the Asian shelf) is intensified and shifted toward Siberia (Proshutinsky and Johnson 1997). The switch of atmospheric circulation regimes (from Az to Zn mode) causes a significant inter-annual shift in the position of the trans-Arctic current, and its correlation (0.78) with hydrological conditions (anomalies of temperature and salinity in the East Siberian Sea) is quite high (Shaipkher and Yankina 1969). The river runoff also determines development of the low salinity Siberian coastal current (SCC) that follows the coasts from west to east (Weingartner et al 1999). Due to wind forcing the SCC is highly variable. Under cyclonic atmospheric conditions the SCC exists as a narrow jet along the coast (for example, in summer 2003), while under anticyclonic atmospheric circulation it was less confined and was not observed in the easternmost part of the ESAS (Weingartner et al 1999, Savileeva et al 2008, Pipko et al 2011). Thus, the variability in position of the frontal zone (FZ) between the ‘Pacific waters’ and ‘local shelf waters’ could be considered an indicator of regional climate change as reflected in alternations of the Zn and Az circulation regimes (Semiletov et al 2005). Further comprehensive studies are required to establish process models with different time-scale resolutions, from intra-seasonal to inter-annual to decadal.

3.2. Specific features of sedimentation in the western and eastern provinces

The sediment OM is generally a mixture between marine and terrigenous material that is characterized by a range of C/N ratios between 6 and 15; typically the lower values characterize marine OM, and the higher values characterize the terrigenous material (Stein and Macdonald 2004). The distribution of δ	extsuperscript{13}N	extsubscript{org} also shows an eastward change from its ‘terrestrial’ value of less than 6 to the ‘marine’ value of more than 8 (Walsh et al 1989). The C/N ratio is negatively correlated with δ	extsuperscript{13}N	extsubscript{org} (r = −0.73) and δ	extsuperscript{15}N	extsubscript{org} (r = −0.77). The spatial pattern of δ	extsuperscript{13}C	extsubscript{org} distribution in the bottom sediment was used to quantitatively estimate the contribution of terrestrial OM (CTOM) to the East Siberian Sea sediment west and east of the geochemical FZ. Following Walsh et al (1989), the amount of OC derived from the terrestrial end-member (δ	extsuperscript{13}C	extsubscript{mar}) can be calculated from the data as

\[
\text{CTOM} (%) = \frac{(\delta^{13}\text{C}_o - \delta^{13}\text{C}_\text{mar})100}{\delta^{13}\text{C}_\text{ter} - \delta^{13}\text{C}_\text{mar}},
\]

where ‘o’, ‘mar’, and ‘ter’ refer to the δ	extsuperscript{13}C	extsubscript{org} values of observed, marine and terrestrial sediment, respectively. Given that the two end members are δ	extsuperscript{13}C	extsubscript{ter} of −27%, typical of higher plants, and δ	extsuperscript{13}C	extsubscript{mar} of −21%, typical of phytoplankton (Walsh et al 1989, Naidu et al 2000), the CTOM values for the western and eastern geochemical provinces (bounded by CTOM = 70%) become 86% and 52% of OC	extsubscript{ter}, respectively (Semiletov et al 2005). Calculations indicate that there is a significant amount of OC	extsubscript{ter} stored within sediments, especially in the nearshore zone most strongly influenced by coastal erosion; between the Dmitry Laptev Strait and the Kolyma River mouth, the OC is almost all of terrestrial origin (from 81% to 100%, mean CTOM = 93%). In contrast, almost half of the sediments underneath transformed Pacific-origin water (the eastern province) are marine in origin. To illustrate the anomalous export of terrestrial OM into the ESAS we show the integrated plume of PM and terrestrial OM in POC for the period 2003–7 (figure 4) from data obtained east of the Lena Delta.

These observations support the hypothesis of a dominant role for coastal erosion (Semiletov 1999a, 1999b) in ESAS sedimentation and in the dynamics of the OC and carbonate system. It can be seen that the PM and the terrestrial OM/POC plume from the Lena is limited to the narrow nearshore zone where most of the Lena River burden settles. The pattern also shows a negligible direct influence of Lena transport of PM into the East Siberian Sea as was earlier suggested (Semiletov et al 2005).

General differences between the oceanographic and geochemical regimes of the western and eastern provinces in the East Siberian Sea are reflected in the size fraction distribution of the surface sediments. The mean psammitic (sand) fraction more than doubled from the western province to the eastern province, while the aleurite (silt) fraction was nearly halved (Semiletov et al 2005). The percentage contribution of the pelite (clay) fraction (particle size = <0.01 mm) is almost the same in the western and eastern provinces; however, the pelite fraction is almost completely terrestrial in origin in the western province, but exhibits a mix of marine and terrestrial origins in the eastern province. The highest OC concentration (up to 1.9%) exists in the pelite fraction. The dominant presence of terrestrial eroded material in ESAS sedimentation is illustrated by the distribution of surface sediments in the ESAS (figure 5): >95% of the entire ESAS area is covered by clayey silt, the main product of coastal erosion (Dudarev et al 2006b).

3.3. Molecular and radiocarbon compositional variations of the terrestrial OM exported by coastal erosion and the GSARs

It is necessary to quantitatively assess the fraction of terrestrial OC (OC	extsubscript{ter}) contributed by riverine runoff and the fraction from coastal erosion; in order to provide a benchmark of the composition, provenance, and extent of degradation of the OC	extsubscript{ter}, it is very important to distinguish between these two sources. The molecular and radiocarbon composition of the OC	extsubscript{ter} released by the GSARs indicate clear patterns relatable to vegetation and climate variations across the 4000 km continent-scale climosequence of the Siberian Arctic (Guo et al 2004, van Dongen et al 2008, Vonk et al 2010, Gustafsson et al 2011). Black carbon ranged from 3 to 10%
Figure 4. Secondary role of the Lena River and dominating role of coastal erosion in ESAS sedimentation is illustrated by surface distribution of (a) particulate materials (mg l$^{-1}$): (1) <2, (2) 2–12, (3) 13–24, (4) >24; (b) contribution of terrestrial OM to POC (%): (1) <25, (2) 25–49, (3) 50–75, (4) >75.

Figure 5. Surface sediments of the ESAS: 1—gravel and pebble, 2—coarse and medium sand, 3—fine sand, 4—silty sand, 5—sandy silt, 6—muddy sand, 7—clayey silt, 8—silty clay, 9—clay, 10—sandy sand–silt–clay, 11—silty sand–silt–clay, 12—clayey sand–silt–clay.
of total OC; the highest fractions in the permafrost-covered East Siberian region showed a $^{14}$C (radiocarbon) mass balance, suggesting about 20% from biomass burning and the balance from fossil fuel combustion or relict black carbon in uplifted source rocks (Elmqquist et al. 2008). Among the lipid biomarkers, there is a large contribution of C$_{23}$ and C$_{25}$ n-alkanes relative to other homologs, suggesting a substantial contribution from sphagnum, with a decreasing trend eastward. Increasing concentrations of both high-molecular-weight (HMW) n-alkanoic acids and beta-sitosterol relative to HMW n-alkanes toward East Siberia both high-molecular-weight (HMW) n-alkanoic acids and beta-sitosterol relative to HMW n-alkanes toward East Siberia is consistent with increasing permafrost and a shorter annual thaw period (Vonk et al. 2010, Gustafsson et al. 2011).

Compound-specific radiocarbon analysis yielded similar modern $^{14}$C fraction values for long-chained n-alkanes and n-alkanoic acids, which increased toward the east. We suggest that these gradients reflect decoupled hydraulic conduits for fresh and aged OC$_{terr}$. Since this decoupling is related to the extent of permafrost, there may be repercussions for OC$_{terr}$ remodeling under a warming climate (van Dongen et al. 2008, Vonk et al. 2010, Gustafsson et al. 2011). If the climate in the ESAS region becomes more like the current climate in the West Siberian region, these results would predict a greater degree (compared to that in western Siberia) of decomposition of the old terrestrial OM released by coastal erosion (figure 2) and the Lena and other eastern GSARs, and thus greater remineralization and release as CO$_2$.

3.4. Transport of terrestrial OM to the ESAS and dynamics of the carbonate system

Rivers transport large quantities of both dissolved and particulate matter and therefore they are carriers of a wide variety of chemical signals to the sea margin (Semiletov 1999a, 1999b, Guo et al. 2004, Semiletov et al. 2007, Pipko et al. 2008, 2010, 2011, van Dongen et al. 2008, Anderson et al. 2009, 2011, Vonk et al. 2010, Charkin et al. 2011, Karlsson et al. 2011). The degradation of particulate OM (POM) causes the formation of an anomalously high partial pressure of CO$_2$ ($p$CO$_2$) and nutrient concentrations in the nearshore zone, where the river signal overlaps with the biogeochemical signal from highly eroded coastal ice complexes enriched by old terrestrial organics (Semiletov 1999a, 1999b, Semiletov et al. 2007, 2011). The POM signal of coastal erosion leads to a $p$CO$_2$ increase in bottom water near the highly eroded ice complexes by up to $\sim$4000 $\mu$atm, while $p$CO$_2$ values in the surface water strongly impacted by river runoff were usually observed to be $\sim$1000 $\mu$atm or less (figure 6).

This is consistent with levels of $p$CO$_2$ measured in carbon plumes from the GSARs along the northern sea route (figure 7); $p$CO$_2$ usually appears in surface water at levels $<500$ $\mu$atm (Semiletov et al. 2007, 2011).

Note that rates of oxidation of POM and OM from surface sediments must be quite high to produce a homogeneous distribution of anomalously high $p$CO$_2$ values ranging between 2000 and 3000 $\mu$atm in the completely mixed water column (figure 6(a), Stations 13–15); the highest values were found in the bottom water of the Buor-Khaya Gulf (figure 6(b), Stations 123–125) where the water column is strongly stratified (Charkin et al. 2011). High rates of eroded OM oxidation were confirmed by direct measurements of CO$_2$ release (measured in September 2006 and 2008 using the Li-Cor chamber technique) in the air above the bluff of the highly eroded ice complex at the northern cape of Muostakh Island in the Laptev Sea; CO$_2$ efflux to the atmosphere usually ranged from tens up to hundreds of mM per square meter per day.

Recycling of limiting nutrients from old terrestrial OM could influence the element stoichiometry (and ‘new production’) (Codispoti and Richards 1968, Cauwet and Sidorov 1996, Jones et al. 1998, Kattner et al. 1999). Our data show that the decay of POM and OM at the sediment surface results in low in situ pH values (as low as 7.24, total scale) and a naturally acidified marine environment. The ESAS exhibits the lowest values of calcium carbonate saturation ever reported for the open marine environment. In some areas of the Laptev Sea (such as at Stations 123–125, figure 6(b)) acidification lowers the saturation state of calcite and aragonite in the surface waters to 0.4 and 0.2, and in the bottom water to 0.2 and 0.1, respectively; these values are even lower than those measured in the East Siberian Sea (Anderson et al. 2011). In contrast to eroded POM, dissolved organic carbon (DOC) was believed to be bio-unavailable (Dittmar and Kattner 2003). However, the DOC concentrations measured in samples from the ISSS 2008 cruise showed a strong non-conservative pattern in areas with FW residence times of several years (Alling et al. 2010). The average DOC losses during mixing along the ESAS shelf were estimated to be 30–50%, suggesting an additional and unaccounted source of CO$_2$ to the atmosphere.

3.5. Factors controlling CH$_4$ release and atmospheric emissions from the ESAS

Permafrost degradation takes place during climate warming and marine transgression. At present, the mean annual onshore permafrost temperature ranges from $−11^\circ$C in the coastal zone (Grigoriev et al. 2006, Rachold et al. 2007), to $−12$ to $−15^\circ$C in the northern ESAS (Novosibirsky Islands, 73°N–76°N), giving a mean latitudinal gradient of about 1.5°C/100 km (Romanovskii et al. 2005). The bed of the high-latitude, shallow ESAS has been alternately exposed to the air and inundated with seawater during glacial and interglacial periods, respectively. Because of its shallowness and location, the ESAS experiences unique climatological transitions. During previous glacial periods, when sea level was more than 100 m lower than at present, the coastline extended up to 1000 km further northward, exposing the continental shelf above the sea surface and, thus, increasing the area of the Siberian coastal accumulative plain by a factor of five (Romanovskii et al. 2005). During the last glaciations, the exposed ESAS permafrost was 4–6°C colder than now (Romanovskii et al. 2000); therefore, the mean annual permafrost temperature ranged from $−15^\circ$C in the southern part of the ESAS to $−19$ to $−21^\circ$C at its northern periphery.
Extensive permafrost formation occurred because freezing extended hundreds of meters deep. Upwardly migrating CH$_4$ became trapped below the impermeable permafrost and was transformed into permafrost-related Arctic hydrates (Soloviev et al. 1987, Kvenvolden 1988). Because they are permafrost-controlled, these hydrates destabilize when sub-sea permafrost experiences warming (Makogon et al. 2007). The transition from the last glacial period to the current warm Holocene was accompanied by a sea level rise that inundated the previously exposed shelf area 5–12 kyr ago (Fleming et al. 1998). Shallow shelf water annual mean temperatures ranged from $-1.8^\circ$C to slightly above $0^\circ$C; therefore, inundation resulted in a warming at the seafloor boundary by up to $13-20^\circ$C (versus $\sim12-17^\circ$C suggested by Soloviev et al. 1987) relative to the temperatures under which permafrost originated in cold epochs when sediments were not covered with water. Over time, ocean heat together with geothermal heat flux-drive submerged permafrost to a new equilibrium state and cause it to thaw. This process first occurs in the areas of fault zones and river estuaries where geothermal heat flux and annual water temperatures are the greatest (Romanovskii et al. 2005, Nicolsky and Shakhova 2010, Charkin et al. 2011). Thus, as permafrost degradation takes place, thaw bulbs, called taliks, form in limited areas (Soloviev et al. 1987, Kvenvolden 1988). Thaw bulbs that penetrate through permafrost are
called open taliks (Romanovskii et al 2005, 2000). Taliks could be considered, first, to be an environment favorable for modern methanogenesis from thawed old carbon and, second, to form gas migration pathways by which CH$_4$ can be released from seabed deposits to the water column and further to the atmosphere (Shakhova et al 2010b, 2010c). The discontinuous presence of taliks is manifested by extremely high spatial variability in concentrations of dissolved CH$_4$, which correlate with spikes in mixing ratios of CH$_4$ in the atmosphere like those shown in figure 8.

Additional destabilization of seabed CH$_4$ deposits could be caused by sediment settlement and adjustment caused by partial hydrate destabilization, seismic and tectonic events, and formation of drainage features in permafrost, where they are interpreted as providing possible migration pathways (Hyndman and Dallimore 2001, Wright et al 2008). Recently detected warmer water temperatures near the seabed may also impact the stability of the ESAS shelf’s submarine permafrost (Holemann et al 2011, Shakhova and Semiletov 2007, 2008).

Our measurements of CH$_4$ taken in 1994–9 and 2003–10 over the ESAS demonstrate that the system is in a destabilization period (Semiletov 1999a, Shakhova et al 2010a, 2010b, 2010c). Moreover, unlike most other sub-sea locations where CH$_4$ oxidizes within the water column, a substantial amount of CH$_4$ released at the seafloor in the shallow ESAS is delivered to the atmosphere (Shakhova et al 2010b, 2010c). Among the factors controlling the fluxes of CH$_4$ to the atmosphere over the ESAS, we discovered seasonal features such as ice coverage, wind speeds and wave heights (frequency of storm events); depth of the water column, which affects transition time from the bottom to the surface and the amount of CH$_4$ removed by oxidation as well as water mixing during deep fall convection; and fraction of CH$_4$ transported by bubbles relative to that transported through diffusion, because bubble-mediated CH$_4$ transport is far more efficient than diffusion (Shakhova et al 2010b, 2010c). We suggest that fluxes from the ESAS could contribute to rising atmospheric CH$_4$ because the ESAS serves as a source of CH$_4$, annually releasing ~8 Tg CH$_4$ to the atmosphere.
Figure 8. Distribution of mixing ratios of CH$_4$ in the atmospheric layer above the sea water (a) and concentrations of dissolved CH$_4$ in the surface layer of shelf water (b) (September 2005).

The atmosphere. A number of flux components remain to be quantified and added to the existing evaluation (Shakhova et al. 2010c).

4. Summary statements and future work

To investigate processes of carbon transport and fate in the ESAS, we employed the ESAS as an integrator of ongoing changes in the surrounding land, which create a terrestrial or exogenous signal, and in situ changes that present a marine or endogenous signal which is generated by submarine permafrost destabilization, increasing coastal and bottom erosion, and involvement of old carbon in the modern biogeochemical cycle. Because two different hydrologic and biogeochemical regimes exist in the western and eastern provinces of the ESAS, the features of carbon cycling are different and specific to each province. In the western province the main processes that drive the marine carbon cycle (except for CH$_4$ emissions) are determined by the combined influence of coastal erosion and riverine runoff from the GSARs. Transformation of the OC$_{terr}$ leads to oversaturation of shelf waters in CO$_2$; CO$_2$ is highest where the contribution of coastally eroded OC$_{terr}$ prevails. The decay of OC$_{terr}$ also lowers pH leading to the acidification of shelf water in the ESAS, which is especially pronounced in the western biogeochemical province. In the eastern province, which is largely influenced by Pacific Ocean inflow, the contribution of OC$_{terr}$ is much lower and marine primary production dominates; there, pCO$_2$ indicated under-saturation of shelf water in CO$_2$.

Since the compound-specific signal of the bulk OC is related to the extent of permafrost, there may be repercussions for OC$_{terr}$ remobilization under a warming climate. Observations showed a highly mosaic pattern of dissolved CH$_4$ spatial distribution in the shelf water. The ESAS serves as a source of CH$_4$ to the atmosphere. CH$_4$ release from the ESAS is determined by the current state of sub-sea permafrost, which has failed to seal ancient
carbon pools sequestered in seabed deposits within and beneath permafrost, and which provides CH₄ emissions to the atmosphere are determined primarily by seasonal features specific to the ESAS such as ice cover, water mixing during storm events, and deep fall convection, rather than by factors influencing the type and the strength of the source. We can conclude that the ESAS accumulates the integrated biogeochemical signal produced by terrestrial and marine ecosystems in response to ongoing climate change. To further assess and quantify the contribution of the ESAS to the regional and global carbon cycle, we plan to obtain new data to answer the following overarching questions.

(1) How does the contribution of the ESAS affect the role of the Arctic Ocean in influencing the regional CO₂ balance?
(2) How do processes of terrestrial OM decay in the ESAS water contribute to Arctic Ocean acidification?
(3) How much CH₄ could be released to the atmosphere from the ESAS due to degradation of sub-sea permafrost and decay of seabed deposits? What is the current state and projected future dynamics of sub-sea permafrost?

To answer these questions we call for extended international cooperation in studying the ESAS. Such study will require multiple year-round exploration campaigns, including drilling of sub-sea permafrost to evaluate the sediment CH₄ potential and comprehensive atmospheric measurements to assess the ESAS strength as a greenhouse gas source. International effort should also be joined in order to quantitatively assess future changes in greenhouse gas emissions in response to ongoing climate change by establishing and developing regional numerical models with the aim of incorporating them into global climate models.

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