Inventories and behavior of particulate organic carbon in the Laptev and East Siberian seas

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[1] Fluvial and erosional release processes in permafrost-dominated Eurasian Arctic cause transport of large amounts of particulate organic carbon (POC) to coastal waters. The marine fate of this terrestrial POC (terr-POC), water column degradation, burial in shelf sediments, or export to depth, impacts the potential for climate-carbon feedback. As part of the International Siberian Shelf Study (ISSS-08; August–September 2008), the POC distribution, inventory, and fate in the water column of the extensive yet poorly studied Eurasian Arctic Shelf seas were investigated. The POC concentration spanned 1–152 μM, with highest values in the SE Laptev Sea. The POC inventory was constrained for the Laptev (1.32 ± 0.09 Tg) and East Siberian seas (2.85 ± 0.20 Tg). A hydraulic residence time of 3.5 ± 2 years for these Siberian shelf seas yielded a combined annual terr-POC removal flux of 3.9 ± 1.4 Tg yr^{-1}. Accounting for sediment burial and shelf-break exchange, the terr-POC water column degradation was ~2.5 ± 1.6 Tg yr^{-1}, corresponding to a first-order terr-POC degradation rate constant of 1.4 ± 0.9 yr^{-1}, which is 5–10 times faster than reported for terr-DOC degradation in the Arctic Ocean. This terr-POC degradation flux thus contributes substantially to the dissolved inorganic carbon excess of 10 Tg C observed during ISSS-08 for these waters. This evaluation suggests that extensive decay of terr-POC occurs already in the water column and contributes to outgassing of CO_2. This process should be considered as a geographically dislocated carbon-climate coupling where thawing of vulnerable permafrost carbon on land is eventually adding CO_2 above the ocean.

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1. Introduction

[2] The Eurasian Arctic Shelf (EAS) is the world's largest continental shelf and covers about one third of the entire Arctic Ocean. The EAS and its vast tundra and taiga drainage basins are predicted to experience the largest temperature increase on Earth due to anthropogenic climate warming [*Zwiers*, 2002; *IPCC*, 2007]. Recent observations are indicating that the region is warming even faster than previous predictions [*Richter-Menge et al.*, 2006], raising issues of climate-carbon couplings with the large carbon store on the nearby land [e.g., *Gruber et al.*, 2004; *IPCC*, 2007; *Schuur et al.*, 2008]. It has already been demonstrated that the enormous export flux of river water to the Arctic Ocean is sensitive to climate [*Savelieva et al.*, 2000; *Peterson et al.*, 2002, 2006] and, by inference, this may also affect the land-ocean fluxes of the organic matter (OM). The pan-Arctic tundra and taiga contain about half of the organic carbon (OC) stored globally in soils, much within shallow permafrost [*Tarnocai et al.*, 2009]. A major part of thaw-released permafrost carbon is found fluvially in the form of particulate organic carbon (POC) [*Guo and Macdonald*, 2006; *Guo et al.*, 2007]. Investigations in the coastal recipients of such river-exported POC thus provide a watershed-integrated view of the released permafrost carbon. It is also plausible that this terrestrial POC (terr-POC) is more degradable in the coastal system than in the terrestrial land-river system due to availability of additional electron acceptors (e.g., SO_4^{-2}). Hence, a hysteresis in the climate-carbon positive feedback process, in terms of atmospheric evasion of CO_2 from degradation of terr-POC displaced to the coastal ocean, should be considered.

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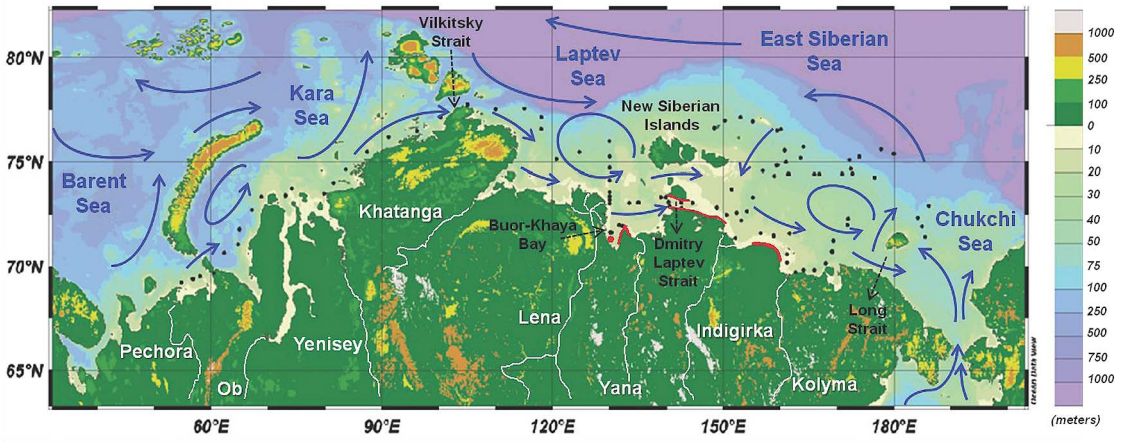

Figure 1. Map of the Eurasian Arctic Shelf, including the Barents, Kara, Laptev, East Siberian, and Chukchi seas, where the International Siberian Shelf Study (ISSS-08) was carried out in the late summer of 2008. Blue arrows indicate main surface currents in the Eurasian Arctic [*AMAP*, 1998]. The 107 sampling stations are represented as black dots, and known areas of intensive coastal erosion are indicated with red lines. The red dot in the Buor-Khaya Bay (Laptev Sea) indicates the location of Muostakh Island, which is particularly important for this study. Color scale for bathymetry and land topography is indicated in the side bar.

[3] While there is a scarcity of studies on early diagenesis of terr-POC and terrestrial dissolved OC (terr-DOC) in the water column in the EAS and elsewhere, some investigations suggest that terr-POC is more active than terr-DOC in biogeochemical cycling, due to its more degradable composition [e.g., *Ittekkot*, 1988; *Stein and Macdonald*, 2004; *Eglinton and Repeta*, 2006; *van Dongen et al.*, 2008a]. Water column degradation of terr-POC may contribute to observations of large CO_2 supersaturation in surface waters of coastal EAS [*Pipko et al.*, 2002; *Semiletov et al.*, 2004; *Semiletov et al.*, 2007; *Anderson et al.*, 2009]. In addition, other processes such as hydrodynamic sorting, deposition-resuspension and uptake by primary production also contribute to the dispersal and processing of the OC in the ESAS. Still, our knowledge of the spatial distribution and fate of terr-POC in the EAS is very limited [e.g., *Stein and Macdonald*, 2004; *Vonk et al.*, 2010a].

[4] The extensive East Siberian Arctic Shelf (ESAS, primarily Laptev Sea, LS, and East Siberian Sea, ESS) is quite shallow (average depth only 50 m). In addition to the massive river input, annual erosion of several meters of permafrosted shoreline of the ESAS is probably an important contribution of terr-OC to these large, yet inaccessible and understudied shelf seas. The ESAS rivers deliver annually ~9 Tg (1 Tg = 10^{12} g) of total OC (TOC) to the Arctic Ocean, with about 18% as POC and 82% as DOC [*Stein and Macdonald*, 2004; *Raymond et al.*, 2007]. Rough estimations of TOC supplied annually by coastal erosion, occurring largely in the LS and western ESS, is lower (~2.3 Tg of TOC), but with higher contribution of POC (~80%) than DOC (~20%) [*Are*, 1999; *Semiletov*, 1999; *Rachold et al.*, 2000; *Stein and Macdonald*, 2004].

[5] Investigations of terr-OC in the EAS have focused either on the nature of riverine OC [e.g., *Holmes et al.*, 2002; *Guo et al.*, 2004; *Neff et al.*, 2006; *Raymond et al.*, 2007; *Elmquist et al.*, 2008; *McClelland et al.*, 2008; *van Dongen et al.*, 2008b; *Goñi et al.*, 2005] or on sediment burial on the Holocene timescale [e.g., *Gordeev*, 2000; *Romankevich and Vetrov*, 2001; *Stein and Macdonald*, 2004]. While there are a few studies on water column POC dynamics on the North American Arctic Shelves [e.g., *Bates et al.*, 2005; *Davis and Benner*, 2005; *Guo and Macdonald*, 2006] and in subarctic northernmost Baltic Sea used as a model system [e.g., *van Dongen et al.*, 2008a; *Vonk et al.*, 2008], basic aspects of POC cycling in the EAS water column such as sources, spatial distribution, inventories and extent of degradation are still largely missing [*Stein and Macdonald*, 2004]. Since transport and decay of POC may play a role in the large CO_2 excess and outgassing to the atmosphere from EAS surface waters [*Semiletov et al.*, 2007; *Anderson et al.*, 2009], it is critical to improve our understanding of the processes controlling the POC distribution and cycling in EAS waters.

[6] The present study provides a geographically extensive baseline of POC in the EAS based on samples obtained along nearly the entire northern seaboard of the Eurasian continent by the International Siberian Shelf Study in August–September 2008 (ISSS-08) (Figure 1). The central objectives of the investigations were to (1) describe the spatial distribution of POC across the different shelf seas and regimes of the EAS, (2) discuss the relative importance of marine versus terrigenous (rivers and coastal erosion) sources of POC and the biogeophysical processes involved in the dispersal and processing of POC, and (3) provide a first estimate of the POC stock and first-order removal rate constant in the LS and ESS.

2. Study Area and Methods

2.1. The Eurasian Arctic Shelf Seas

[7] The EAS, composed of the Barents, Kara, Laptev, East Siberian and Russian part of Chukchi seas (Figure 1) is the widest, shallowest and, by area, largest shelf system in the World Ocean [e.g., *Stein and Macdonald*, 2004]. This study

covers all the EAS shelf seas and almost the entire (~8,400 km) northern seaboard of the Eurasian continent spanning ~140° in longitude, yet the main focus is on the LS and ESS, since these are the largest and least studied EAS seas.

[8] The LS covers almost $500 \cdot 10^3$ km^2 and has an average water depth of 48 m [*Stein and Macdonald*, 2004]. One of its most important characteristics is the high freshwater discharge (~745 km$^3 \cdot$yr^{-1}), which is received mainly from the Lena River (~566 km$^3 \cdot$yr^{-1}) [*Cooper et al.*, 2008]. Rough estimations based on measurements on mainstreams of the Lena River channels establish an annual supply of 20.7–28.6 Tg of total suspended particulate matter (SPM) from the Lena [*Holmes et al.*, 2002]. However, major precipitation (85–90%) of the SPM (and thus POC) on the marginal delta has been proposed [*Lisitzin*, 1995] that may reduce the final amount of particulate material reaching the sea [*Semiletov et al.*, 2010].

[9] The shallow ESS (average depth of 58 m) is the largest ($987 \cdot 10^3$ km^2), most ice-bound and most poorly explored marginal sea of the Arctic Ocean [*Stein and Macdonald*, 2004; *Macdonald et al.*, 2008]. This sea exhibits two distinct physical and biogeochemical provinces with a distinct border (frontal zone, FZ) oscillating around 160°E [e.g., *Semiletov et al.*, 2005]. In the eastern ESS (E-ESS, from ~160°E to ~180°E), influenced by Pacific inflow, primary production is believed to be an important OC source (30 Tg$\cdot$yr^{-1}) [*Stein and Macdonald*, 2004]. In the western ESS (W-ESS, from ~140°E to ~160°E), river runoff and coastal erosion, both rich in products of permafrost degradation, supplemented by relatively high primary production in certain polynya regions, supply the major part of OC [*Kosheleva and Yashin*, 1999; *Semiletov et al.*, 2005]. Coastal thermo-abrasion is common in the W-ESS because extensive parts of the coast are composed of ice-bonded permafrost soils [*Stein and Macdonald*, 2004], which are destabilized upon thawing and washed into the near-coastal waters.

2.2. Sampling Program

[10] In August–September 2008 a complex hydrogeochemical and geophysical sampling and at-sea analysis program (ISSS-08) was accomplished during the 50 days onboard the (H/V) Yacob Smirnitskyi (Archangelsk) [*Semiletov and Gustafsson*, 2009].

[11] Seawater samples for POC and associated analytes were obtained both during steaming, from a 4 m seawater intake (SWI) system, and at hydrographic stations (termed Yacob Smirnitskyi, YS), with a rosette of Niskin bottles arranged around a Seabird CTD (Conductivity-Temperature-Depth) sensor package. The SWI system was composed of stainless steel and HDPE tubing, and retrieved water from underneath the hull at 4 m depth. The SWI system operated exclusively during full steaming. For the YS stations, samples were obtained, based on CTD profiles, from near bottom, middle bottom, pycnoline (4–10 m) and middle mixing layers (2–4 m). For POC, 238 samples were processed from 107 sites (Figure 1); 58 from hydrographic CTD sampling stations (YS) and 49 from SWI during steaming.

[12] The salinity data collected with the SeaBird 19+CTD were compared with deep water samples analyzed onboard with an AUTOSAL lab-salinometer with a good agreement and no need for further calibration. For turbidity monitoring, a Wetlabs turbidity sensor ECO NTU S/N NTURTD-126 was attached to the CTD. This sensor detects light scattered by particles within the water column and generates an output voltage proportional to the particle concentration. The output signal ranged between 0 and 5 V.

2.3. Organic Carbon and Nitrogen Measurements

[13] Between 1 and 3 L of seawater was collected in precleaned clear polycarbonate bottles for OC analysis. The samples were vacuum filtered onboard on precombusted (12 h, 450°C) glass fiber filters (GF/F) of 25 mm diameter, held in an all-glass filtration system. The TOC fraction was separated in two operationally size-defined fractions: the POC (>0.7 μm), retained on the GF/F, and the DOC (<0.7 μm), collected as the filtrate.

[14] The GF/F filters were immediately stored frozen until analysis. Subsamples for elemental analysis (C and N) and stable isotope composition (δ^{13}C) were punched out from the GF/F filters (15–17 punches of 4 mm diameter), placed in precombusted silver capsules (5 × 9 mm, Säntis Analytical AB, Uppsala, Sweden), acidified with HCl (1M) to remove carbonates, and dried overnight at ~50°C. The analyses were performed at the UC Davis Stable Isotope Facility (UC Davis, California, United States), by means of isotope ratio mass spectrometry (Europa Hydra 20/20). The analytical error of the POC and PN quantification was established on the basis of replicate analyses (n = 3) of three SWI samples to range 4.1–11.5% (mean of 6.9%) and 6.4–12.3% (mean of 9.2%), respectively.

[15] DOC was collected from each filtrate in 60 mL Nalgene HDPE bottles. These were stored cold (<4°C) and measured onboard within a few days with a catalytic combustion instrument (SHIMADZU TOC-VCPH). The DOC method and quality checks are described in detail elsewhere [*Alling et al.*, 2010]. In general, the relative error of the measurements was ≤5% but slightly higher for DOC concentrations <80 μM (~8%).

2.4. Interpolations and Estimation of Inventories

[16] For interpolations and inventory calculations we used the Data Assimilation System program (DAS), developed at Stockholm University [*Sokolov et al.*, 1997] (http://nest.su.se/das/) and used in several previous applications similar to the present one [e.g., *Conley et al.*, 2002; *Savchuk et al.*, 2008]. The advantage of using DAS over more common oceanographic programs is its better resolution of the sea bottom surface in section plots (bathymetry was downloaded from IBCAO homepage; *Jakobsson et al.*, 2008]), as well as its interpolation tool. DAS interpolations allow calculating inventories and mean values in different water bodies, with user-defined vertical and horizontal limits as well as based on salinity (DAS technique detailed in Section A1 in Text S1).[1]

3. Results and Discussion

3.1. Oceanographic and Biogeochemical Settings of the Eurasian Arctic Shelf

[17] The oceanography of the EAS is characterized by a strong stratification, extensive continental shelves and pro-

[1] Auxiliary materials are available with the HTML. doi:10.1029/2010GB003862.

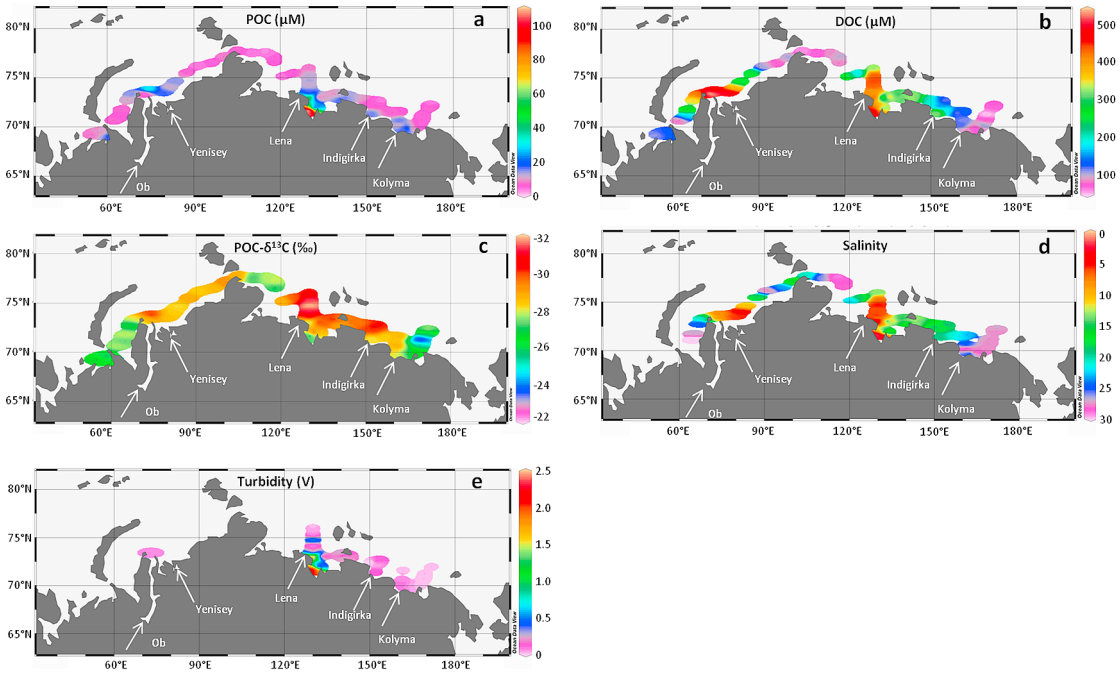

Figure 2. The near-coastal distribution and composition of the surface (4 m depth) water (a) POC, (b) DOC, (c) POC-δ^{13}C, (d) salinity, and (e) turbidity measured during the ISSS-08 cruise.

nounced seasonal forcing (runoff, ice formation, sunlight), all factors that influence the transport of sediments and OC. Much of the water column stratification in the Eurasian Arctic is supported by the large freshwater inflow that sustains a low-density surface layer with important implications for the water circulation. In surface waters, the circulation pattern is dominated by a general eastward drift within the coastal zone (Figure 1), which transports a major portion of the Lena discharge through the Dmitry Laptev and Sannikov Straits toward the ESS [*Semiletov et al.*, 2005]. In late summer 2008, the low-salinity plume of the Lena River reached as far as ~160°E, where it usually meets the more saline Pacific waters and forms the FZ [*Semiletov et al.*, 2005]. Pacific waters enter the Bering Strait and flow into the Arctic Interior via the Chukchi and Herald Canyons, and into the ESS through the Long Strait (Figure 1), to supply deep water in the E-ESS. In contrast, deep waters in the western EAS are mainly supplied by the more dense and saline Atlantic waters. In the Arctic Interior, the dominant feature is the westward water transport (Figure 1), mostly influenced by the Transpolar Drift, which runs from the LS-ESS, across the central Siberian basins, toward the western Fram Strait [*Arctic Monitoring and Assessment Program* (*AMAP*), 1998, and references therein].

[18] Biogeochemical dynamics on the EAS are significantly affected by large seasonal cycles. Nine-months-long winters are characterized by extensive coverage of ice that limits freshwater input and primary production [*Stein and Macdonald*, 2004]. During the partially ice-free season (~June to October), the substantial input of SPM hampers light penetration and results in moderate marine primary production in coastal waters [*Stein and Macdonald*, 2004]. Peak freshet usually takes place in May–June [*Dittmar and Kattner*, 2003; *McClelland et al.*, 2008]. In late August–2008, the freshwater lense from the Lena River extended ~350 km northward and down to ~8 m depth in LS, whereas a combination of Lena outflow and weak plumes from the Indigirka and Kolyma Rivers produced a poorly defined halocline at ~15 m depth in the W-ESS.

[19] The extensive ESAS (~86% of the combined LS and ESS) is an important region for sea-ice production as well as trapping and processing of freshwater, dissolved and particulate inputs from land. The effectiveness of these processes depends on the residence time of shelf waters, which have been estimated to be ~3.5 years for the LS and ESS [*Schlosser et al.*, 1994; *Ekwurzel et al.*, 2001; *Karcher and Oberhuber*, 2002]. Tidal effects are small in the ESAS with tidal ranges of just 20 cm [*Dmitrenko et al.*, 2001]. In the LS, most of the SPM transport takes place within the bottom nepheloid layer. During the ice-free period, SPM is mainly trapped within the shelf through deposition-resuspension dynamics. Net SPM export to the deep Arctic is mainly limited to the freezeup period (~October), through transport via newly formed ice, which in contrast is of minor importance during the ice-covered season [*Wegner et al.*, 2003]. In the central ESS, *Vonk et al.* [2010a] recently documented the importance of the benthic boundary layer transport to disperse coastal erosion-derived OC up to 500 km off the Kolyma River mouth.

3.2. Spatial Distribution of POC and Its Properties

[20] The POC concentration in the EAS water column during ISSS-08 ranged from 1 to 152 μM (mean ±95% confidence interval of 14.7 ± 2.9 μM) (data in Table A1 in Text S1). The LS showed the highest POC concentrations (25.8 ± 8.1 μM, n = 78), with highest values in the SE LS close to Muostakh Island (Figures 2a and 3a–3b), strongly influenced by both the Lena River and coastal erosion

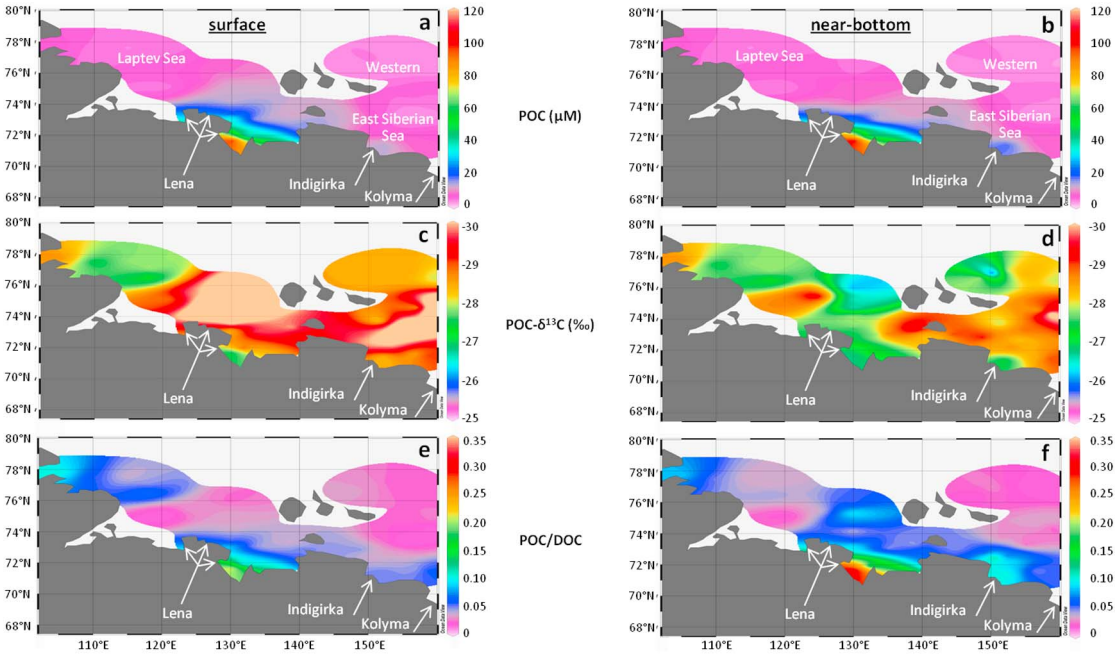

Figure 3. The POC, POC-δ^{13}C, and POC/DOC distributions in surface (4 m depth) versus near-bottom waters in the Laptev and western East Siberian seas.

(Figure 1). In contrast, the lowest POC levels were found in the ESS (8.5 ± 1.1 μM, n = 116) (Figures 2a and 3a–3b), where values as low as 1–3 μM were registered NE of the New Siberian Islands.

[21] The δ^{13}C-POC ratio ranged from −21 to −32‰ (Table A1 in Text S1) in the EAS, with values for the major river estuaries of ∼−30‰ (Ob, Yenisey and Lena) and ∼−28‰ (Indigirka and Kolyma) (Figure 2c). The EAS δ^{13}C-POC signal may be compared with canonical values for marine phytoplankton (−18 to −21‰) and terrestrial C3 plants (−23 to −34‰) [*Meyers*, 1997]. However, there are also different processes that may have produced variation of the δ^{13}C-POC signal. Rayleigh distillation may cause a slight isotopic depletion in dissolved CO_2 of cold polar waters [*Meyers*, 1997], yielding somewhat lower δ^{13}C-POC values. Primary producers assimilating δ^{13}C-depleted terrestrial dissolved inorganic carbon (DIC) may also cause such depletion [*Chanton and Lewis*, 1999; *Peterson and Fry*, 1987]. However, the influence of freshwater phytoplankton in the δ^{13}C-POC signatures is likely minor, due to the poor light penetration in these rivers. The PARTNERS project recorded mean δ^{13}C-POC values (n = 17) in the lower reaches of the major EAS rivers between 2003 and 2006 (Ob: −32.3‰, Yenisey: −30.3‰, Lena: −29.5‰ and Kolyma: −29.9‰) [*McClelland et al.*, 2008] corresponding well with our values during ISSS-08 (Figure 2c). Another recent study found somewhat more enriched δ^{13}C-POC in the Lena River (mean of ∼−28‰) for the repeat summer observations 1999–2007 [*Semiletov et al.*, 2010].

[22] The eastward extension of the depleted δ^{13}C signal (∼−30‰) in surface waters (Figure 3c) likely reflects strong influence of the Lena River, not only in the LS but also in the ESS. According to *Semiletov et al.* [2005], the Lena discharge of SPM is a negligible contribution in the Dmitry Laptev Strait. However, POC may be discharged in several forms, ranging from rapidly settling coarse particles to near-neutrally buoyant large aggregates-flocculates of humic substances [e.g., *Gustafsson and Gschwend*, 1997; *Gustafsson et al.*, 2000]. While POC associated with coarse mineral particles is prone to settle close to land, suspended humic aggregates can travel further, especially when driven by a freshwater plume [*Vonk et al.*, 2010b]. Hydrodynamic sorting of the heterogeneous POC contributes to the non-conservative behavior of the POC (Figure 4a), and the difference of 1–2‰ observed in the near-bottom versus the surface waters (Figures 3a–3b). While surface layers are strongly affected by buoyant freshwater plumes of depleted δ^{13}C-POC, deeper layers are apparently more influenced by either settled primary production–derived POC or erosion–derived POC. The former is known to produce an isotopic fractionation due to preferential settling of δ^{13}C-heavy diatoms [e.g., *Waite et al.*, 2005], whereas the latter is ballasted with poorly sorted mineral particles and thus preferentially settling to deeper strata [e.g., *Gustafsson and Gschwend*, 1997; *Gustafsson et al.*, 2000; *Vonk et al.*, 2010b], influencing the severe nonconservative behavior at salinities <10 (Figure 4a). In addition, nepheloid layer dynamics may also contribute to the variation of δ^{13}C-POC in the bottom waters, through repeated cycles of deposition and resuspension across the inner shelf [*Coppola et al.*, 2007; *Vonk et al.*, 2010a]. Interestingly, the samples exhibiting the highest POC contents (Figure 4a) were collected in the vicinity of the eroding sites of Muostakh Island and the Buor-Khaya Cape. While a clear distinction between riverine and erosion sources is difficult to attain in the Buor-Khaya Bay due to the extensive area of low salinity, the high POC concentrations also at high salinity in this region suggest the contribution of terrestrial sources other than the Lena River (i.e., coastal erosion).

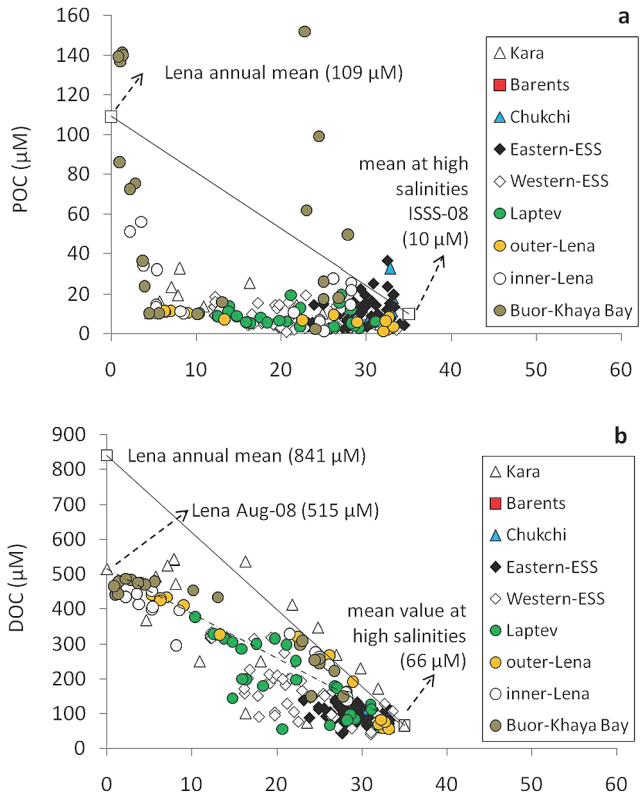

Figure 4. Property-salinity plots for the ISSS-08 samples: (a) POC and (b) DOC. In both Figures 4a and 4b, end-member values from the literature and observations during ISSS-08 are shown, together with hypothetical conservative mixing lines. In Figure 4a, the POC value at low salinity (841 μM) corresponds to the Lena annual mean value [*McClelland et al.*, 2008], and the ocean interior value corresponds to the mean of the high-salinity POC values from ISSS-08. In Figure 4b, Lena annual mean DOC is taken from *Raymond et al.* [2007], Lena Aug-08 DOC is the mean of direct measurements in the three major channels of the Lena River (ISSS-08), and mean value at high salinities is from *Opsahl et al.* [1999] and this study.

[23] More enriched δ^{13}C values (~−28‰) were found in large parts of the Kara, LS (Buor-Khaya Bay) and ESS (Figure 2c). Since river δ^{13}C-POC signature from at least two studies is around −30‰ (*McClelland et al.* [2008] and this study), those values may reflect either dilution with marine-planktonic POC, or a terrestrial source other than riverine, such as coastal erosion. Similar δ^{13}C values (~−27 to −28‰) in samples from eroding soil from the Quaternary coastal-ice complex along the coastal margin of the ESS [*Stein and Macdonald*, 2004] and the LS [*Schirrmeister et al.*, 2002; *Dudarev et al.*, 2006; *Sánchez-García et al.*, 2010; *Semiletov et al.*, 2010] support the latter hypothesis. Distinctly enriched values (~−24‰) in the easternmost EAS (Figure 2c) indicate marine influence of Pacific waters with higher primary production east of the ESS FZ [*Kosheleva and Yashin*, 1999; *Semiletov et al.*, 2005]. Most of the samples collected from offshore in the Kara, Chukchi, Laptev, Eastern and Western East Siberian seas during the ISSS-08 contained δ^{13}C values more depleted than the canonical −21‰ of typical phytoplankton (Figure S1). Such depletion has been observed in tropical [*Chanton and Lewis*, 1999], as well as in boreal coastal systems [*Gustafsson et al.*, 2001], where the δ^{13}C-POC decreased to below −30‰ with onset of ice coverage and then increased to around −22‰ after break up. Restricted interchange with atmospheric CO_2, together with recycling of DIC in the ice-covered waters were suggested as potential factors of the δ^{13}C depletion. Still, an enriched δ^{13}C (−24.5‰) of highly degraded Yedoma in the lower Kolyma drainage basin [*Dutta et al.*, 2006] complicates these interpretations.

[24] The nonconservative behavior of the POC in the EAS is further observed for the C/N ratio. Overall, the majority of the samples showed C/N ratios lower than those constrained to be possible by mixing of the three putative end-members (i.e., rivers, erosion and primary production; Figure S1). Lowering of C/N ratios has been observed in soils [e.g., *Sollins et al.*, 1984] and in ocean sediments [e.g., *Schubert and Calvert*, 2001], putatively related to mineral (clay) adsorption of inorganic nitrogen (e.g., ammonium) derived from decomposition of OM accompanied by remineralization and release of carbon. In the EAS, this factor is likely of minor relevance, given the lower amount of particulate inorganic nitrogen (0.3775 μM) observed at zero concentration of POC (Figure S2). Alternatively, the lowering of the C/N ratios may be caused by selective degradation of labile carbonaceous forms, as recently suggested by *Hugelius and Kuhry* [2009] to explain the decrease of the C/N ratio in recently exposed peat permafrost after rapid thaw of freeze-locked old carbon. Taken together, the composition and nonconservative geospatial distribution of POC in the EAS may be explained as a combination of variable contributions of three major sources (rivers, coastal erosion and primary production), and various biogeophysical processes, where microbial degradation and sediment burial are likely to play major roles.

3.3. Terrigenous Input of OC in the Eurasian Arctic Shelf

[25] Terrigenous OC from Arctic rivers and coastal erosion are important carbon sources to the Arctic shelf seas [e.g., *Stein and Macdonald*, 2004]. Whereas fluvial OC is released as both POC and DOC, erosion is expected to remobilize OC mainly as POC [*Stein and Macdonald*, 2004; *Guo and Macdonald*, 2006]. At the time of the ISSS-08, the EAS water column exhibited the highest DOC value near the major river mouths of the Kara (Ob and Yenisey) and Laptev (Lena) seas (Figure 2b). Similar distribution was observed for freshwater (traced by salinity; Figure 2d), the eastward extension of which into the ESS illustrated the strength of the Lena River discharge. In contrast to POC that preferentially settles out close to land (Figure 4a), DOC was dispersed further out onto the EAS with variable extents of conservative mixing (Figure 4b) [*Alling et al.*, 2010]. The similar distributions of salinity and DOC in surface waters, detailed by *Alling et al.* [2010], suggests the use of these parameters as markers for river-exported terr-DOC for the EAS.

[26] Turbidity was measured as part of the CTD casts near the major river mouths (Figure 2e). Despite the large freshwater discharge of the Ob and Yenisey rivers [*Cooper et al.*, 2008], turbidity at their mouths is not nearly as high as in the SE LS (Figure 2e). Similar to POC, the highest

turbidities appeared in the Buor-Khaya Bay, with lower values along the northward Laptev plume of the Lena. The similar nearshore distribution of turbidity and POC is supported by a significant positive correlation ($r^2 = 0.77$, at p value <0.001) between these two parameters (Figure S3a). Considering that 75% of the river water discharge [*Dudarev et al.*, 2006] and 64% of the SPM [*Kuptsov and Lisitzin*, 1996] comes from one of the northeastward Lena channels (Trofimovskaya), which is separated by Buor-Khaya maxima in many parameters (Figure 2), it seems likely that a major part of the high turbidity in the Buor-Khaya Bay is derived from a nonriverine source. The steep shorelines of Yedoma in this area are subjected to intensive coastal erosion [*Overduin et al.*, 2007], evident in satellite images (Figure S4). The Bykovsky Peninsula and the Muostakh Island are examples of affected shorelines within the Buor-Khaya Bay [*Schirrmeister et al.*, 2002; *Overduin et al.*, 2007; *Charkin et al.*, 2010; *Semiletov et al.*, 2010]. We hypothesize that coastal erosion plays a major role in the delivery of POC and suspended sediments in this area, causing the observed high turbidity.

[27] The geospatial distribution of the explored geochemical parameters provides useful information about the terrigenous sources of OC in the EAS. While the DOC distribution is clearly influenced by the riverine export, POC and turbidity away from the intermediate river mouths appear to have coastal erosion as an important common source. Thus, the different main origin of DOC and POC may offer an interesting tool to track coastal erosion versus riverine discharge in the EAS. In the next section we explore the possibility of using the POC/DOC ratio to further distinguish between riverine and coastal erosion sources in the ESAS.

3.4. The POC/DOC Ratio and Coastal Erosion

[28] The spatial distribution of the POC/DOC ratio in the ESAS is shown in Figure 3. On average, the near-bottom waters (Figure 3f) exhibited POC/DOC ratios more than twice those of the surface waters (Figure 3e). The highest POC/DOC ratios (up to 0.5) were observed in the Buor-Khaya Bay (Table A1 in Text S1), in both surface and near-bottom waters. In contrast, the lowest ratios (<0.05) were observed in the mid-outer shelf of the ESAS (Figures 3e–3f), where slightly higher ratios (~0.1) in the near-bottom waters illustrate the importance of the nepheloid layer dynamics in the dispersal of POC across the shelf [*Vonk et al.*, 2010a]. The higher contents of POC relative to DOC in the near-bottom waters may be explained by continuous cycles of deposition and resuspension dispersing the POC that has been largely deposited within the Buor-Khaya Bay (Figures 3a–3b). Cross-shelf transport of sediments with POC in the bottom waters may be a prominent feature in the shallow ESAS during the ice-free season [*Wegner et al.*, 2003; *Vonk et al.*, 2010a] as has been reported also for other shelf systems [e.g., *Coppola et al.*, 2007].

[29] To further assess the vertical distribution of POC/DOC with respect to sources of POC, we traced a coastline section including the major part of the sampling stations next to land in the eastern LS and W-ESS (Figure 5a). The POC/DOC ratios varied between ~0 and 0.5 along the coastline section, with higher values within the Buor-Khaya Bay (around 132°E; Figure 5a). Although the large discharge of the Lena makes it difficult to clearly distinguish between river and coastal erosion, there was a distinct difference in the composition of the water north of the Lena Delta (salinity of ~15, POC of ~21 μM and turbidity of 0.56 V) and that in the vicinity of the Muostakh Island (salinity of ~8, POC of ~85 μM and turbidity of 2.6 V). Despite both waters being influenced by the Lena River (low salinity), the higher content of POC and turbidity around Muostakh Island suggested erosion as an additional source of POC here. Also, water near the mouth of Ob and Yenisey Rivers, with discharges comparable to Lena [*Cooper et al.*, 2008], presented similar salinity (~10) but much lower POC (~18 μM) and turbidity (0.06 V) than those around Muostakh Island, supporting the importance of coastal erosion in the supply of POC in these waters.

[30] A perpendicular section tracing the transition from the outer surface plume of the Lena River to the interior of the Buor-Khaya Bay (Figure 5b) vividly illustrates the high POC concentration measured around Muostakh Island. Along the section, including nine of the ISSS-08 YS stations, the surface water POC/DOC ratio increased from values ~0.01, in the outer plume, to ratios ~0.2 in the proximity of the delta, to reach the highest POC/DOC ratios (~0.5) in the Buor-Khaya Bay around Muostakh Island (Figure 5b). Whereas the POC in the Buor-Khaya Bay in principle can be supplied by either the Lena River or coastal erosion, the distinctly higher POC/DOC ratios in the vicinity of Muostakh than in any other location along the Lena transect suggest a dominant input from the island. The erosion evolution of this island has been observed over the last decades [*Overduin et al.*, 2007], showing a retreat rate of 10–13 m between 1951 and 2002, and up to 20 m during the summer months since 2001 [*Semiletov et al.*, 2010; *Grigoriev*, 2010]. The abrasion and degradation of the island is likely delivering large amounts of POC, which settle close to the island and contribute significantly to the enhanced POC/DOC signatures in surrounding near-bottom waters.

[31] Although of lower magnitude, relatively high POC/DOC ratios (~0.2–0.3) were also observed at other locations along the ESAS coastline, such as ~133°E (Buor-Khaya Cape), ~151°E and 164–166°. All these coastal segments are known to be subjected to intense thermo-abrasion (Figures 1 and S4) [*Stein and Macdonald*, 2004; *Overduin et al.*, 2007]. In the ESS, the characteristic coastal landscape of smooth relieves and rounded shapes (Figure S4) can be explained by the action of erosional processes. The depleted $\delta^{13}C$ ratios (~−26‰) observed in the surrounding waters of these coasts support the contribution of terrestrial sources at a time of the year when the riverine signal was weak (section 3.1), likely making coastal erosion the dominant source of terrestrial POC. All in all, this suggests that the POC/DOC ratio combines with turbidity and $\delta^{13}C$ as potentially useful proxies to qualitatively differentiate between coastal erosion and river sources of the EAS POC. Future studies may explore the quantitative use of $\Delta^{14}C$ as a putatively even more powerful tracer of erosional input of POC from the Pleistocene coastal-ice complex [*Vonk et al.*, 2010a].

3.5. POC Inventory and Removal Flux in the East Siberian Arctic Shelf Seas

[32] The dense sampling coverage in the ESAS (81 stations) allowed us to constrain robust estimates of the POC

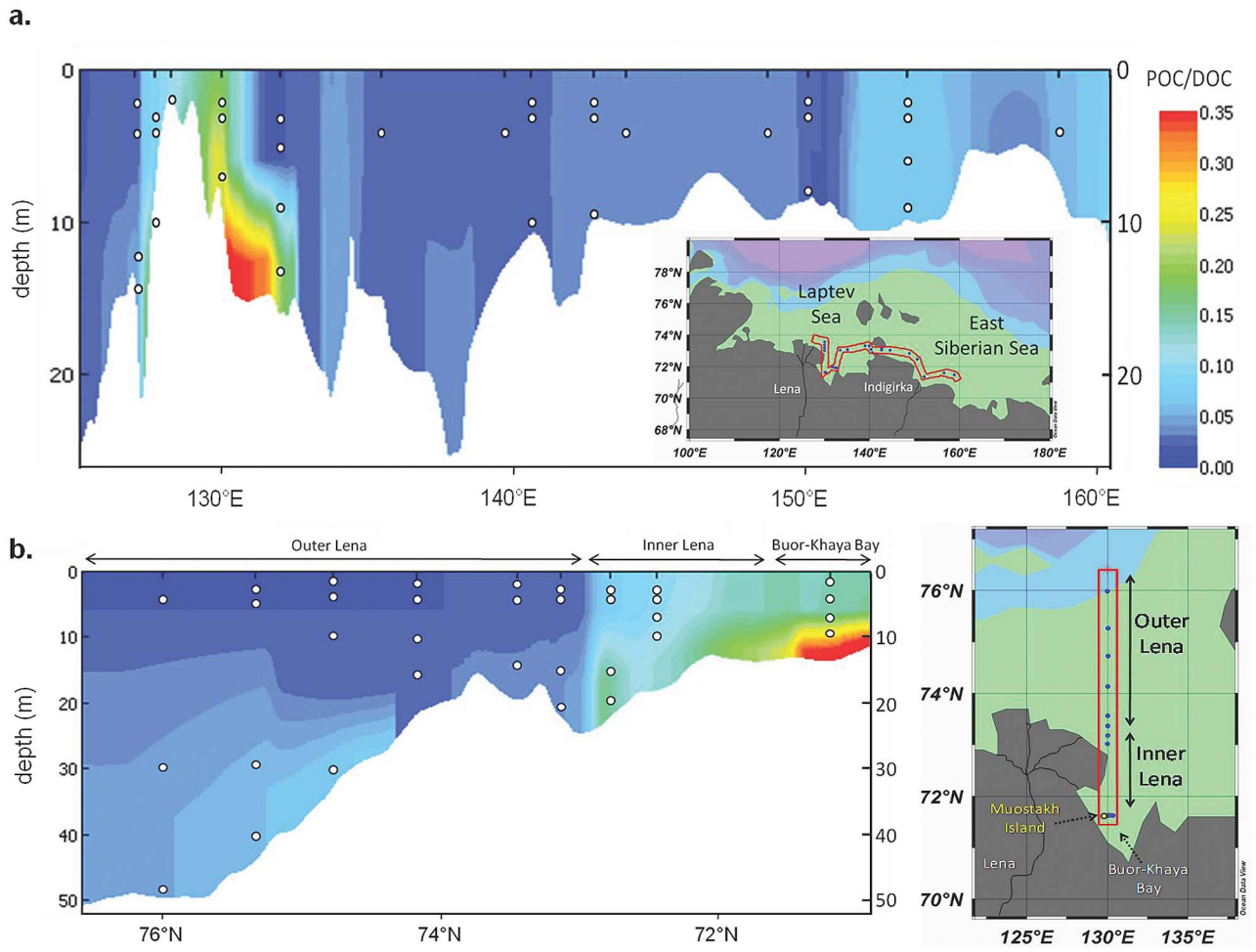

Figure 5. Sections of the POC/DOC ratio (a) near the coast spanning between the eastern Lena Delta and 160°E, where intensive coastal erosion is expected (Figures 1 and S4), and (b) along the Lena-Laptev transect, where the POC concentration was among the highest measured during the ISSS-08.

inventories for the LS and ESS. These estimates (Table 1) are based on DAS interpolations extending to the outer border of the shelf (defined at 200 m isobath). For the LS, the water column POC inventory was 1.32 ± 0.09 Tg with a DAS-based volume-weighted mean salinity of 25, for POC concentration of 5.68 μM, and a clear terrigenous imprint in δ^{13}C of −28.1‰. For the ESS, the water column POC inventory was 2.85 ± 0.20 Tg, with a major proportion (83% or 2.36 ± 0.16 Tg) residing in the more voluminous E-ESS. The influence of Pacific-derived waters [*Semiletov et al.*, 2005] explains the higher mean salinity of 30 measured in the E-ESS. The mean δ^{13}C of −25.9‰ suggests about equal contributions of marine-planktogenic and terrestrial sources to its POC stock. In contrast, the POC stock in the less voluminous W-ESS (0.48 ± 0.03 Tg) appears, similarly to the LS, to be dominated by terrestrial sources (mean δ^{13}C of −28.9‰). As far as we know, these are the first estimates of water column POC stocks in the ESAS and thus serve as a useful baseline for assessment of the terr-OC fate in the Arctic Ocean.

Table 1. Estimates of the Water Column POC Inventory in the Laptev and East Siberian Seas[a]

Shelf	Mean Salinity	Mean δ^{13}C (‰)	Water Volume (km^3)	POC Inventory[b] (Tg)	Mean POC (μM)
Laptev Sea	25	−28.1	19,374	1.32 ± 0.09	5.68
East Siberian Sea	29	−26.5	51,340	2.85 ± 0.20	4.63
Western part (140°E–160°E)	24	−28.9	8,750	0.48 ± 0.03	4.14
Eastern part (160°E–180°E)	30	−25.9	41,623	2.36 ± 0.16	4.73

[a]Estimations based on Data Assimilation System (DAS) interpolations, which returns water volume-weighed integrations.
[b]POC inventories are expressed as the mean ± standard deviation, which is estimated from the propagation of the analytical error of POC replicates (relative standard deviation of 6.9%, n = 234).

Table 2. Estimates of Terrestrial POC Input and Removal Fluxes for the Laptev and Western East Siberian (140°E–160°E) Seas

Shelf	Laptev Sea	West-East Siberian Sea	Together
Riverine input ($Tg \cdot yr^{-1}$)	1.30	0.17[a]	1.47
Coastal erosion input ($Tg \cdot yr^{-1}$)	1.44	1.32[b]	2.76
Eolian input ($Tg \cdot yr^{-1}$)	0.09	0.09[c]	0.18
Total terrestrial input ($Tg \cdot yr^{-1}$)	2.83	1.58	4.41
Annual POC removal ($Tg \cdot yr^{-1}$)	2.5 ± 0.8	1.4 ± 0.7	3.9 ± 1.4
Annual POC degradation[d] ($Tg \cdot yr^{-1}$)	1.5 ± 0.9	1.0 ± 0.7	2.5 ± 1.6
First-order removal rate constant[e] (yr^{-1})	1.9 ± 0.6	3.0 ± 1.4	2.2 ± 0.8
First-order degradation rate constant[f] (yr^{-1})	1.1 ± 0.7	2.1 ± 1.5	1.4 ± 0.9

[a]The Indigirka River is the only major riverine POC source in the W-ESS.

[b]Taken as 75% of that for the total ESS (1.76 $Tg \cdot yr^{-1}$) based on the areal extent of apparent erosional input from satellite images (Figure S4).

[c]Taken as 50% of the total ESS POC eolian input (0.18 $Tg \cdot yr^{-1}$), considering that the longitudinal extent of the W-ESS is about half of the entire shelf sea.

[d]Values obtained after subtraction of the OC burial flux for the LS (0.98 $Tg \cdot yr^{-1}$) and W-ESS (0.42 $Tg \cdot yr^{-1}$, as 44% of the total ESS bottom area as derived by DAS [*Stein and Macdonald*, 2004]) from the respective annual POC removal estimated in this study.

[e]Estimated as: k (yr^{-1}) = Flux removal ($Tg \cdot yr^{-1}$)/POC inventory k (Tg).

[f]Estimated as: k (yr^{-1}) = Flux degradation ($Tg \cdot yr^{-1}$)/POC inventory k (Tg).

[33] To seek further constraint on the fate of terr-POC in the ESAS, the input and removal processes were quantitatively assessed on the basis of the available information on hydraulic and other system features of the ESAS. For this, we focused on the LS and the W-ESS; the two systems overwhelmingly influenced by terr-POC. The terr-POC fluxes to the LS, suggested by the compilation of *Stein and Macdonald* [2004], are 1.30 $Tg \cdot yr^{-1}$ for rivers, 1.44 $Tg \cdot yr^{-1}$ for coastal erosion and maximum 0.09 $Tg \cdot yr^{-1}$ for eolian input (Table 2). Similarly, the terr-POC sources to the W-ESS includes 0.17 $Tg \cdot yr^{-1}$ from rivers (Indigirka as the only major river), whereas POC fluxes from coastal erosion and eolian sources are only available for the entire ESS. The coastal erosion POC flux to the W-ESS was taken as 75% of that for the total ESS, based on the areal extent of apparent erosional input from satellite images (Figure S4), and thus 1.32 $Tg \cdot yr^{-1}$. Since the longitudinal extent of the W-ESS is about half of the entire shelf sea, its eolian POC input was taken as 50% of that total and thus 0.09 $Tg \cdot yr^{-1}$.

[34] There are several limitations to these available literature data. First, the important Lena-Laptev River flux is based on a study that only considered outflow through one, albeit the main, of the several important Lena delta channels. Multiyear studies of POC from several of the delta channels and in the SE-LS demonstrates large interannual variability [*Semiletov et al.*, 2010]. For the erosional-POC input to the shelf seas there is a scarcity of investigations, with no peer-reviewed studies conducted from the sea side yet published [*Semiletov et al.*, 2010; *Charkin et al.*, 2010]. The current estimates must thus be regarded as uncertain. However, a recent study of surface sediment OC along a 500 km offshoreward transect in the mid ESS suggested a dominant contribution from erosion [*Vonk et al.*, 2010a]. For the eolian POC input, based on one single study, it is unclear how the quality assurance with respect to contamination from the ship's exhaust was ascertained. Nevertheless, with need for further refinement, the currently available data suggest a total terr-POC flux of ~4.4 ± 1.39 $Tg \cdot yr^{-1}$ entering the combined LS and W-ESS (Table 2), based largely on the *Stein and Macdonald* [2004] literature compilation.

[35] To make progress toward estimating the removal fluxes indicated by the strongly nonconservative POC-salinity plot (Figure 4a), we need to take into account the hydraulic residence time of this system. The deficit in the observed terr-POC inventory (Table 1) relative to the expected terr-POC input over 3.5 ± 2 yr [*Schlosser et al.*, 1994] represents the total terr-POC removal flux (Table 2). This missing POC is on the order of 3.9 ± 1.42 $Tg \cdot yr^{-1}$ with about 60% of the removal in the LS water column and about 40% in the smaller W-ESS (Table 2). These preliminary estimations are based on a steady state mass balance, where the only significant removal pathways are sedimentation and water column degradation, since the net POC export to other water masses, primarily across the shelf break, was estimated to be on the order of <2% of the POC removal due to degradation and sediment burial reported here (calculations detailed in Section A2 in Text S1). Minor SPM transport during great part of the year [*Wegner et al.*, 2003] supports the lesser importance of off-shelf export processes for the POC water column mass balance on these wide shelves. A recent box model of the fate of terr-POC exported from a subarctic river found only a minor fraction hydraulically exported out of the box volume despite it constituting a much smaller volume in northernmost Baltic Sea, thus also inferring rapid removal due to POC degradation and settling [*van Dongen et al.*, 2008a].

[36] The only estimate available for sediment sequestration of water column POC is a Holocene-averaged POC burial of 1.4 $Tg \cdot yr^{-1}$ (estimated from *Stein and Macdonald* [2004]). While this is clearly over a different temporal scale than the other processes considered here, subtracting this estimate from the total POC (deficit-based) removal flux yields a water column terr-POC degradation flux in the LS and W-ESS on the order of 2.5 ± 1.6 $Tg \cdot yr^{-1}$ (Table 2). This degradation flux may be compared with terr-OC degradation fluxes inferred for the same ISSS-08 model box from the carbonate system [*Anderson et al.*, 2009] and from the DOC mass balance [*Alling et al.*, 2010]. *Anderson et al.* [2009] constrained a DIC excess of ~5 Tg C each for the LS and W-ESS for the ISSS-08 observations in summer of 2008 and inferred a key role for transport and decay of particulate eroded terr-OC. *Alling et al.* [2010] used an extensive coverage of DOC from the ISSS-08 campaign to estimate a DOC degradation flux for the W-ESS of 5.1 $Tg \cdot yr^{-1}$. Given our estimate of POC degradation flux for the same W-ESS of 1.02 ± 0.70 $Tg \cdot yr^{-1}$ (Table 2), it becomes clear that the water column remineralization fluxes are of similar size for these two different OC pools, despite very large (factor of near 35) difference in POC and DOC stocks. This ~17% of terr-OC degradation occurring

in the POC pool for this system is lower than the ~44% estimate for the POC pool in the coastal waters receiving subarctic river discharge in the northernmost Baltic Sea [*van Dongen et al.*, 2008a]. In contrasts, the *Stein and Macdonald* [2004] compilation on the OC cycle in the Arctic Ocean suggest a zero degradation of the DOC component over the Siberian-Arctic shelf seas. The herein estimated terr-POC decay flux is consistent with the carbonate budget and represents ~25% of the excess DIC measured in the area, eventually to be released to the Arctic atmosphere as CO_2.

[37] Apparent first-order degradation rate constants were derived to facilitate comparison with other systems and incorporation into models. Considering a combined POC stock of 1.8 ± 0.09 Tg for the LS and W-ESS (Table 1), the POC losses observed here produce a first-order degradation rate constant of 1.4 ± 0.9 yr^{-1} (Table 2). These estimates are conservatively low, since we are considering only terr-POC for the degradation fluxes, whereas total POC (including marine-derived POC) for the POC inventory. A smaller number for the exclusively terr-POC inventory would produce higher degradation rate constants. In comparison, *Alling et al.* [2010] reported a first-order DOC degradation rate constants of 0.30 yr^{-1} for the W-ESS, very similar to that of 0.24 ± 0.07 yr^{-1} reported by *Letscher et al.* [2011] for the entire eastern Arctic Ocean (from 0° to 180°E) and in contrast to that suggested by *Manizza et al.* [2009] for the same latter region (0.1 yr^{-1}). *Stein and Macdonald* [2004], due to lack of information at that time, relied in their box modeling of the terr-OC on a "35% universal terr-POC degradation" [*Ittekkot*, 1988] in every Arctic shelf sea, irrespective of hydraulic residence time and other factors. Considering an average residence time on the Siberian-Arctic shelves of 3.5 ± 2 years [*Schlosser et al.*, 1994], that corresponds to using a first-order degradation rate constant of roughly 0.1 yr^{-1} for POC [*Stein and Macdonald*, 2004], which is an order of magnitude lower than the present estimate based on actual observations in the ESAS. All in all, degradation of terr-POC in the ESAS water column is nearly as important as DOC degradation and consistent with excess DIC and, thus, substantially more important than earlier suggestions.

3.6. Consideration of Uncertainties

[38] The uncertainties involved in this assessment of POC dynamics on the EAS were carefully considered through a combination of available analytical uncertainties (of our >200 measurements), literature-provided uncertainties and, when not available, estimations of uncertainties based on an expert judgment of how well a parameter in the literature is constrained. For the POC inventories (Table 1), the reported uncertainties correspond to the analytical error of the POC replicates (6.9%; relative standard deviation (rsd)). An additional sensitivity analysis of the inventory calculation was performed by performing the DAS interpolation estimates with first the central value minus 1sd of all POC observations and then repeating with central value plus 1sd. The range in POC inventories for our three ESAS systems were in all three cases between 7 and 9% of uncertainty.

[39] The estimations of the POC removal fluxes (Table 2) involved several parameters from the literature. Since it was desired to provide uncertainty estimates on the POC removal and degradation fluxes, uncertainties of each parameter were considered to allow for error propagation. Hence, the propagated uncertainty of the POC removal fluxes were estimated based on the rsd of the corresponding (1) inventories, (2) hydraulic residence time, and (3) terrestrial influx. The uncertainty of hydraulic residence time was provided in the literature (3.5 ± 2 years; *Schlosser et al.* [1994]), whereas that of terrestrial influx had to be derived from the propagation of the estimated uncertainties of the riverine, erosional and eolian inputs. Given the absence of provided uncertainties in those published inflow fluxes, we applied conservative rsd to reflect the relative scale of uncertainty of these terrestrial input. For the riverine influx, most likely the best constrained vector, a 10% rsd was chosen assuming that the uncertainty of this well-studied parameter should not be much larger than the analytical error estimated here for the POC determination (6.9% rsd). Considering the larger uncertainty of the other two terrestrial inputs, a rsd of 50% was applied for both coastal erosion and eolian inflows. Finally, for the propagated uncertainty of the POC degradation flux (Table 2), the additional uncertainties of the sediment burial and off-shelf export fluxes were added. In the absence of available uncertainty estimates in the literature for these processes in the ESAS, rsd of 50% were assigned also to these processes. Taken together, the propagated uncertainty for the combined LS and W-ESS annual POC degradation flux and first-order degradation rate constant was 64% rsd (Table 2).

4. Summary and Conclusions

[40] This geographically extensive study covering almost the entire northern seaboard of Eurasia provides a benchmark on POC distribution and behavior in the EAS. A combination of geochemical proxies indicated that the 2.36 ± 0.16 Tg POC contained in the E-ESS in late summer 2008 were about equally influenced by terrestrial and marine (Pacific) sources, whereas the 1.8 ± 0.09 Tg POC held in the combined LS and W-ESS reflected dominantly terrestrial sources. Higher POC concentrations in areas known to be affected by intensive coastal erosion in the LS and W-ESS suggest that this process should be investigated in future studies.

[41] The strongly nonconservative behavior of POC was quantified in a steady state mass balance assessment to correspond to an annual terr-POC removal flux of ~3.9 ± 1.4 Tg and a first-order terr-POC removal rate constant of 2.16 ± 0.8 yr^{-1} for the combined LS and W-ESS. Accounting for sediment burial and net hydraulic exchange, this suggested that about two thirds of the terr-POC lost annually in the LS and W-ESS is caused by POC degradation in the water column (~2.5 ± 1.6 Tg yr^{-1}; ~1.4 ± 0.9 yr^{-1}). This degradation flux is much larger than realized earlier [*Stein and Macdonald*, 2004] and the annual flux similar to that from the DOC pool for the W-ESS [*Alling et al.*, 2010]. The degradation rate for POC is ~7 times faster than for DOC in the W-ESS. Furthermore, the combined POC and DOC degradation fluxes is consistent within a factor of ~2 with the water column DIC excess for the same study area and period [*Anderson et al.*, 2009]. This estimate thus suggests that about 25% of what is eventually outgassing as CO_2 could have been caused by degradation

of terr-POC in the LS and W-ESS. This terr-POC is originating from the land-based region of continuous permafrost in the river drainage basins and from coastal Yedoma. Given that enhanced climate warming in this region may accelerate release of this old and large carbon reservoirs, the current finding of extensive degradation of the terr-OC in the Arctic shelf waters, up to 1000 km away from the point of release, should be considered as a geographically dislocated part of the total feedback process between climate warming and permafrost carbon.

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