1	Carbon mineralization in Laptev and East Siberian Sea shelf and slope sediment
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17 Abstract The Siberian Arctic Sea shelf and slope is a key region for the degradation of terrestrial organic material transported from the organic carbon-rich permafrost regions of 18 19 Siberia. We report on sediment carbon mineralization rates based on O₂ microelectrode profiling, intact sediment core incubations, ³⁵S-sulfate tracer experiments, porewater 20 dissolved inorganic carbon (DIC), $\delta^{13}C_{DIC}$, and iron, manganese, and ammonium 21 concentrations from 20 shelf and slope stations. This data set provides a spatial overview of 22 23 sediment carbon mineralization rates and pathways over large parts of the outer Laptev and East Siberian Arctic shelf and slope and allowed us to assess degradation rates and efficiency 24 25 of carbon burial in these sediments. Rates of oxygen uptake and iron and manganese 26 reduction were comparable to temperate shelf and slope environments, but bacterial sulfate 27 reduction rates were comparatively low. In the topmost 20 to 50 cm of sediment, aerobic carbon mineralization dominated degradation and comprised on average 84% of the depth-28 integrated carbon mineralization. Oxygen uptake rates and ³⁵S-sulfate reduction rates 29 integrated over the topmost 30 cm of sediment were higher in the eastern East Siberian Sea 30 shelf compared to the Laptev Sea shelf. DIC/NH₄⁺ ratios in porewaters and the stable carbon 31 isotope composition of remineralized DIC indicated that the degraded organic matter on the 32

Siberian shelf and slope was a mixture of marine and terrestrial organic matter. Based on dual 33 34 end member calculations, the terrestrial organic carbon contribution varied between 32% and 36%, with a higher contribution in the Laptev Sea than in the East Siberian Sea. Extrapolation 35 of the measured degradation rates using isotope end member apportionment over the outer 36 shelf of the Laptev and East Siberian Sea suggests that about 16 Tg C per year are respired in 37 the outer shelf sea floor sediment. Of the organic matter buried below the oxygen penetration 38 depth, between 0.6 and 1.3 Tg C per year are degraded by anaerobic processes, with a 39 40 terrestrial organic carbon contribution ranging between 0.3 and 0.5 Tg per year.

Key words: Carbon mineralization, Arctic shelf and slope sediment, Laptev Sea, East Siberian
Sea

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44 **1. Introduction**

The biogeochemical fate of terrestrial organic carbon deposited on the Arctic shelf and 45 46 slope is one of the most important open questions for the marine Arctic carbon cycle (e.g., Tesi et al., 2014; Macdonald et al., 2015; McGuire et al., 2009; Vonk et al, 2012). The total 47 pan-Arctic terrestrial permafrost carbon reservoir has been estimated at about 1100 - 1500 Pg 48 (Hugelius et al, 2014) – a carbon pool large enough to substantially affect the global 49 atmospheric carbon dioxide pool over the next 100 years, even when only partially 50 decomposed after thawing and oxidation (Schuur et al. 2015; Koven et al., 2015). Yet, there 51 52 remains considerable uncertainty regarding the mineralization of terrestrial organic matter exported by rivers and coastal erosion to the Siberian shelf and slope (Tesi et al., 2014; 53 Karlsson et al. 2015; Semiletov et al., 2011; Salvado et al., 2016). 54

55 Terrestrial organic matter transported to the Siberian shelf is of variable size, age, and molecular composition, which results in a range of qualitatively different carbon degradation 56 rates of bulk carbon and individual molecular components. Size class analysis of the organic 57 matter suggests that coarse organic material settles preferentially in near-shore environments, 58 whereas finer organic fractions disperse offshore in repeated deposition-resuspension cycles 59 gradually losing particular molecular components and overall reactivity (Wegner et al., 2013; 60 61 Tesi et al., 2014, 2016). Substantial oxic degradation of organic matter may occur during near-bottom transport in resuspension-deposition cycles across the shelf (Bröder et al., 62 2016a). Up to 90% of certain biomarker classes may decompose during transport, whereby 63

most of the degradation may take place while the transported organic material resides in the
sediment before being resuspended (Bröder et al., 2016a). However, without making
approximations on transport direction, particle travel time and travel distance these studies
cannot provide direct insights into the rates of carbon degradation and resultant CO₂ fluxes
from sediment. By contrast, direct kinetic constraints provided by sediment carbon
degradation rates can provide testable data for coupled hydrodynamic biogeochemical models
that help assess the fate of land-exported terrestrial carbon pool on the Siberian shelf.

Relatively few studies have directly measured rates of carbon mineralization rates in 71 Siberian shelf sediment (e.g., Boetius and Damm, 1998; Grebmeier et al., 2006; Karlsson et 72 al., 2015, Savvichev et al., 2007). Boetius and Damm (1998) used high-resolution oxygen 73 74 microelectrode data to determine the surface oxygen concentration gradients and oxygen 75 penetration depths in a large number of sediment cores from the shelf and slope of the Laptev 76 Sea. Based on corresponding sediment trap and export productivity data, they concluded that 77 the annual marine organic carbon export in the Laptev Sea shelf and slope was sufficiently high to explain the observed oxygen uptake rates. Current understanding therefore holds that 78 79 due to the long annual ice cover and low productivity on the eastern Siberian Arctic shelf and slope, only a small amount of marine organic carbon is exported and buried in Laptev and 80 East Siberian Sea shelf sediment. The highly reactive fraction of fresh organic matter is 81 thought to degrade in the surface sediment (Boetius and Damm, 1998). Consequently, 82 anaerobic respiration in buried sediment has been thought to be negligible and to reflect the 83 degradation of unreactive terrestrially derived carbon compounds. To our knowledge, with the 84 exception of a recent study by Karlsson et al (2015) a more direct assessment of terrestrial 85 carbon-derived mineralization rates in buried shelf and slope sediment has not been reported 86 for the East Siberian Arctic Sea. 87

In this study, we present data from oxygen microelectrode profiling experiments, porewater data of dissolved inorganic carbon and its stable carbon isotope composition, and ³⁵S-sulfate reduction rate experiments along a shelf-slope transect near 125°E in the Laptev Sea. Samples were taken during the summer 2014 on the SWERUS-C3 expedition with the Swedish icebreaker Oden. We combined these data with porewater analyses of dissolved ammonium, sulfate, iron, and manganese to assess the major carbon degradation pathways and rates across the extensive outer Laptev and Siberian shelf and slope.

96 2. Materials and methods

97 **2.1. Sample collection**

Samples were collected at 20 stations from 40 to 3146 m water depth in the western 98 99 Laptev and East Siberian Sea (Fig. 1 and Table 1). In this study we only report on sampling sites that showed no methane gas plumes, acoustic anomalies in the water column, or 100 101 sediment blankings indicative of rising gas. In areas of active ebullition from the seafloor as seen by video imagery and acoustic gas blankings in the water column, the biogeochemistry 102 of sea floor processes such as bacterial sulfate reduction, DIC concentration and its carbon 103 isotope composition, and oxygen uptake are affected by methane oxidation. These methane 104 105 cycling-related signals overprint the biogeochemistry imparted by carbon mineralization and are reported in a separate study. 106

107 Sediment stations had variable ice cover (Table 1). In the Laptev Sea, except for the deep-water slope stations between 3146 m and 2106m, all stations had open water. By 108 contrast, ice cover exceeded 75% in the East Siberian Sea to the west and east of Bennett 109 island (Station 40 to 63). Sediments with well-preserved sediment surfaces were collected 110 with a Multicorer (Oktopus GmbH, Kiel, Germany) that simultaneously takes 8 sediment 111 cores over an area of about 0.36 m² with acrylic tubes (9.5 cm diameter, 60 cm length) to 40 112 cm depth preserving clear water on top of the sediment. At stations 6, 23, and 24, an 113 underwater video system (Group B Distribution Inc., Jensen Beach, U.S.A.) was mounted on 114 the multicore frame to record the deployment and recovery, and to document the sea floor 115 habitat. For the investigations all cores were taken from the same cast. Two of the cores were 116 used to determine ³⁵S-sulfate reduction rates and porosity. In addition, one core with 117 predrilled 3.8 mm holes sealed with electric tape was used to extract porewaters with rhizons 118 119 (Rhizosphere Research Products BV, Wageningen, Netherlands). A fourth core was used for 120 microelectrode measurements of dissolved oxygen concentration profiling, and finally, four 121 other cores were used for whole-core incubations to determine benthic fluxes of dissolved oxygen, dissolved inorganic carbon, and nutrients. The cores were capped with rubber 122 123 stoppers until further subsampling usually within 30 minutes. For sulfate reduction rates, the cores were subsampled with 40 or 50 cm long acrylic tubes (26 mm inner diameter) prepared 124 125 with silicon-sealed holes, drilled at distances of 1 cm. For whole-core incubations, the cores 126 were sub-sampled with 25 cm-long, 60 mm-wide tubes (56 mm id) to 12 cm depth. Likewise, a 60 mm diameter tube (56 mm id) was collected for microelectrode measurements preserving 127

about 3 cm of the overlying bottom water. For intact whole-core incubations, temperature-

- 129 controlled aquaria were filled with bottom water that was collected from a CTD rosette from
- the same station by collecting water from four ten-liter rosette bottles usually ~5 meters above
- 131 the sea floor. All sediment cores were closed with a stopper retaining the water on top of the
- sediment and stored at 1.5°C in an incubator until further processing.
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134 2.2. Microelectrode oxygen profiles

135 High-resolution O_2 -profiles across the water-sediment interface were obtained to 136 determine oxygen penetration depths and diffusive oxygen uptake (Rasmussen and Jørgensen, 1992; Glud, 2008) (Table 2). The 60 mm tubes were placed in an aquarium filled with bottom 137 138 water from the same station, overflowing the sediment core. The water temperature was kept to ~1°C by a cooling unit (Julabo GmbH, Seelbach, Germany). In exceptional cases when 139 140 there was not sufficient bottom water available to fill the aquarium, bottom water was used from a pump system. A stable diffusive boundary layer above the sediment was created by 141 passing air from an aquarium pump over the water surface with a Pasteur pipette creating a 142 143 slow rotational motion of water inside the core. At each station six to eight O₂ microprofiles were measured using Clark-type oxygen microelectrodes (OX-50, Unisense, Århus Denmark) 144 mounted on a motor-driven micromanipulator (MM33, Unisense, Århus Denmark). O₂ 145 sensors were calibrated with fully oxygenated bottom water from the same station at $\sim 1^{\circ}$ C for 146 saturation and for anoxic conditions by dissolving Na₂SO₃ in the same water. The first profile 147 in each core was measured with a resolution of 1000µm as a quick scan to locate the sediment 148 surface and to adjust the measuring range. Then the vertical resolution was increased to 100-149 500µm and additional five to seven profiles were measured at different points on the surface, 150 151 approximately one cm apart from each other.

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153 **2.3. Whole-core sediment incubations**

Four intact cores with clear overlying water were subsampled in the laboratory in acrylic tubes (i.d. 56 mm, height 25 cm) retaining about 10 cm of the overlying water. The sediment and water height in the tubes were approximately 10 cm. The cores were incubated in a 40-liter incubation tank filled with bottom water from the same station. Before the incubation the overlying water in the cores was equilibrated with bottom water in the tank.

The overlying water in the cores was stirred by small magnetic bars mounted in the core liners 159 and driven by an external magnet at 60 rpm. The cores were pre-incubated uncapped for 6 160 hours and subsequently capped and incubated for a period of 6 to 24 hours depending on the 161 initial oxygen concentration in the bottom water. 2D oxygen sensor spots (Firesting oxygen 162 optode, PyroScience GmbH, Aachen, Germany) with a sensing surface of a diameter of 5 mm 163 were attached to the inner wall of two incubation cores. The sensor spots were calibrated 164 against O₂-saturated bottom water and oxygen-free water following the manufacturer's 165 guidelines accounting for temperature and salinity of the incubation water. Measurements 166 167 were performed with a fiberoptic cable connected to the spot adapter fixed at the outer core 168 liner wall at the spot position. The O₂ concentration was continuously logged during 169 incubations. Sediment total oxygen uptake (TOU) rates were computed by linear regression of the O_2 concentration over time. 5 ml of overlying water were removed over the course of the 170 171 incubation used for nutrient and dissolved CO₂ analysis as described below. Linear regression best fits were used to determine the exchange fluxes of dissolved CO₂. 172

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174 **2.4. Extracted porewater analysis**

Porewater samples for concentration measurements of total dissolved CO₂ (DIC), 175 sulfate, and ammonium were obtained using the methods described in Seeberg-Elverfeldt et 176 al. (2005). Rhizons were treated for 2 hours in 10% HCl solution, followed by two rinses with 177 deionized water for 2 hours and final storage in deionized water. The rhizons were connected 178 to 10 mL disposable plastic syringes with inert pistons (VWR, Stockholm, Sweden) via 179 polyethylene 3-way luer-type stopcocks (Cole-Parmer, U.S.A.) and inserted in 1-cm intervals 180 through tight-fitting, pre-drilled holes in the liner of the sediment cores. The first mL of pore 181 182 water was discarded from the syringe. No more than 5 ml were collected from each core to prevent cross-contamination of adjacent porewater due to the suction effect (Seeberg-183 Elverfeldt et al., 2005). The collected porewater was divided into four different aliquots for 184 later chemical analysis. For dissolved sulfate analysis, 1 ml of porewater was preserved with 185 200 µl of 5% zinc acetate solution and frozen. For ICP-AES analysis of dissolved metals and 186 major cations, 1 ml of porewater was preserved with 100 µl of 10% Suprapur HNO₃ and 187 stored cold. For analysis of dissolved ammonium, 2 ml of porewater were frozen untreated. 188 For analysis of dissolved inorganic carbon, 2 ml of porewater were preserved with 100 µl 189 10% HgCl₂ and stored cold in brown glass vials without headspace. Ammonium was 190

determined on a QUAATRO 4-channel flow injection analyzer (Seal Analytical) on board. 191 All other porewater analyses were performed at the Department of Geological Sciences, 192 Stockholm University. Samples that were analyzed in the home laboratory remained cold or 193 frozen on board until arrival of the icebreaker Oden in Sweden. Sulfate concentration was 194 measured on diluted aliquots on a Dionex System IC 20 ion chromatograph. DIC 195 concentrations were determined by flow injection analysis (Hall and Aller, 1992). Dissolved 196 iron and manganese were determined on diluted aliquots by ICP-AES (Varian Vista AX). For 197 carbon isotope analysis of dissolved inorganic carbon, 1 ml of porewater was filled into 12 ml 198 199 exetainers to which 1 ml of concentrated phosphoric acid was added. The carbon isotope 200 composition of the formed CO₂ was analyzed on a GasbenchII-MAT 253 isotope ratio mass 201 spectrometer coupled to a GC-PAL autosampler. Results are reported in the conventional 202 delta notation relative to PDB. Precision of isotope analysis is 0.1‰.

203 For the calculation of porewater concentration ratios of DIC and NH₄⁺, the effects of different diffusion coefficients and ammonium adsorption were accounted for. We have no 204 205 direct measurements of adsorption coefficients for these sediments. Instead, we used an ammonium adsorption coefficient of 1.3 established for comparable, terrestrially dominated 206 207 silty clays in the East China Sea for which similar porosities and organic carbon 208 concentrations were reported (Mackin and Aller, 1984). The diffusion coefficient of HCO_3^{-1} is about 45% smaller than the diffusion coefficient of NH₄⁺ (Li and Gregory, 1974). The two 209 effects required an upward correction of the ammonium concentration by 40% to facilitate 210 direct comparison in DIC/NH4⁺ ratios. Diffusion- and adsorption-adjusted DIC/NH4⁺ ratios 211 were also corrected for the bottom water DIC and NH₄⁺ concentrations (Table 1). Only 212 concentrations below 4 cm depth were used for comparison to avoid effects of oxidation on 213 214 NH₄⁺ concentrations.

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2.5. Reaction transport modelling 216

Reaction rates and fluxes were estimated from the concentration profiles of dissolved 217 oxygen, manganese, iron, and dissolved inorganic carbon according to the general reaction-218 transport equation at steady state accounting for diffusion and advection exemplified here for 219 220 dissolved oxygen according to

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$$\frac{d}{dz} = \left(\varphi(D_s + D_b)\frac{dO_2}{dz}\right) + \varphi\alpha(O_{2, z=0} - O_{2, z}) + \sum_{z=0}^{\infty} R = 0$$
(1)

At steady state, the rate of the concentration change reflects the balance between the 223 consumption due to respiration and oxidation of reduced inorganic compounds (R) against 224 diffusion and advection due to bioirrigation into sediment (Glud, 2008). D_s (cm² s⁻¹) is the 225 sediment diffusion coefficient and was calculated for the experimental temperature and 226 salinity according to Boudreau (1997). The sediment diffusion coefficient D_s was recalculated 227 from the molecular diffusion coefficient D_0 according to $D_s = D_0 / \theta^2$, where $\theta^2 = 1 - \ln(\varphi^2)$, 228 where ϕ is porosity and θ is tortuosity (Boudreau, 1997). D_b (cm² s⁻¹) is the bioturbation 229 coefficient and α is the irrigation coefficient (cm s⁻¹). D_b and α were estimated by stepwise 230 optimization by fitting a concentration profile to the measured data using the least square 231 fitting procedure of the program Profile (Berg et al., 1998) testing various coefficients until 232 the statistically best fit was obtained. Boundary conditions and the coefficients D_b and α for 233 234 the best fits are shown in the supplemental material Table 1.

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236 **2.6.** ³⁵S-Sulfate reduction rates

237 For the incubations, the whole-core incubation method by Jørgensen (1978) was used. 35 SO₄²⁻ tracer solution was diluted in a 6 ‰ NaCl solution containing 0.5 mM SO₄²⁻ and 2.5 µl 238 239 of the tracer solution (200kBq) was injected through the pre-drilled holes. The cores were then capped and sealed in plastic wrap foil and incubated for 8 hours at the respective bottom 240 water temperatures. After this time, the incubations were stopped by sectioning the core in 1-241 cm intervals to 5 cm depth and in two centimeter intervals below this depth to the bottom of 242 the core. Sediment sections were transferred into 50 ml plastic centrifuge tubes containing 20 243 ml zinc acetate (20% v/v), mixed to a slurry on a vortex stirrer and frozen. The total amount 244 of ³⁵S-labeled reduced inorganic sulfur (TRIS) was determined using the single-step cold 245 chromium distillation method by Kallmeyer et al. (2004). TRIS and supernatant sulfate were 246 counted on a TriCarb 2095 Perkin Elmer scintillation counter. The sulfate reduction rate was 247 calculated using the following equation (Jørgensen, 1978): 248

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$${}^{35}SRR = \left(\frac{TRI^{35}S \times 1.045}{({}^{35}SO_4^{2-} + TRI^{35}S)}\right) \times [SO_4^{2-}]/\rho T$$
 (2)

where $[SO_4^{2-}]$ is the pore water sulfate concentration corrected for porosity ρ , TRI³⁵S and ³⁵SO₄²⁻ are the measured counts (cpm) of sulfate and total reduced inorganic sulfur species, respectively, 1.045 is a correction factor accounting for the kinetic isotope effect of ³⁵S relative to ³²S, and T is the incubation time. The sulfate reduction rate is reported as nmol cm⁻ ³ day⁻¹. ³⁵SRR were measured in two parallel cores for all depth intervals. The incubation
experiments were conducted between July 15 and August 20, but for logistical reasons
(transport to Stockholm) the distillation of the samples was conducted between December 10,
2014 and April 2, 2015 so that between 1.7 and 2.7 half-lives of ³⁵S (87.4 days) had passed
before all samples were processed. The resulting detection limit of the rate measurements
accounting for distillation blanks and radioactive decay of ³⁵S between experiment and
laboratory workup was 0.01 nmol cm⁻³ day⁻¹.

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262 **2.7.** Carbon equivalent apportionment of terminal electron-accepting processes (TEAP)

The modelled oxygen, iron, manganese, and DIC reaction rates were integrated over a depth 263 264 interval of 30 cm to permit comparison between different stations. The integrated terminal electron accepting process (TEAP) rates were recalculated into carbon equivalents for the 265 266 electron acceptors oxygen, iron, manganese, and sulfate using an idealized $(CH_2O)_x$ stoichiometry for organic matter (Vandieken et al., 2006; Nickel et al., 2008). The 267 contribution of inorganic reoxidation to oxygen consumption and the effects of internal 268 recycling of sulfur, iron, and manganese are discussed in section 4.1. The calculated rates 269 270 were then used to calculate the contribution of the different aerobic and anaerobic electron acceptors to total carbon mineralization for 5 stations in the Laptev and East Siberian Sea 271 (Table 3). 272

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274 **2.8** Marine versus terrestrial endmember partitioning of carbon degradation rates

In order to determine the mineralized proportion of terrestrial and marine organic matter in the sediment directly, the DIC concentrations and the carbon isotope composition of DIC were used as indicators of remineralized organic matter in the sediment. The remineralized fraction (F) of DIC_{total, depth x} at the different sediment depths (x) was defined as

279 $F = (DIC_{total, depth x} - DIC_{bottom water overlying core})/DIC_{total, depth x}$ (3)

and this fraction was plotted against the respective carbon isotope composition of $DIC_{total, depth}$

281 $_x$ (Fig. 6B). The y-intercept of the linear regression was constrained by the carbon isotope

composition of DIC in the bottom water for the respective station so that the slope of the

regression line was the only unknown in this analysis. The gradients for each station yielded

the average stable carbon isotope composition of the remineralized organic matter in the 284 sediment assuming no or very minor isotope fractionation during the oxidation of organic 285 matter. This calculation assumed that porewater removal or addition of DIC by diagenetic 286 processes such as CaCO₃ precipitation or dissolution was minor and time-invariant, which is 287 supported by the observation that Ca^{2+} and Mg^{2+} porewater concentrations at these shelf and 288 slope stations did not change significantly with depth (Brüchert and Sun, unpubl. data). The 289 relative contributions of the terrestrial and marine organic carbon were then calculated with a 290 291 linear two-endmember isotope model:

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$$\delta^{13}C_{\text{DIC, remineralized}} = f_{\text{terr}} * \delta^{13}C_{\text{terr OC}} + f_{\text{mar}} * \delta^{13}C_{\text{marine OC}}$$
(4)

where f_{terr} and f_{mar} are the respective mass fractions of terrestrial and marine-derived organic carbon and $\delta^{13}C_{terrestrial}$ and $\delta^{13}C_{marine}$ reflect the isotope composition of these endmembers. In order to derive specific degradation rates of the marine and terrestrial carbon fractions, the endmember mixing-based assessment of the marine and terrestrial organic carbon contributions to DIC were combined with the mineralization rates derived for the different electron acceptors.

299

300 **3. Results**

301 3.1. Physical and chemical bottom water conditions

Table 1 summarizes the general site characteristics of the investigated sediment 302 stations. Bottom water salinity varied between 34.9 ‰ in the outer Laptev Sea at 3146 m 303 depth (Station 1) to 29.1 ‰ in the East Siberian Sea at 40 m (Station 45). The lower salinity 304 in the East Siberian Sea can be attributed to longshore transport of freshwater eastward from 305 the Lena River. Bottom water temperatures varied between -1.8°C at Station 27 and 0°C at 306 Station 37, but there was no regional trend in the data. Cored sediment consisted of silty clays 307 to clayey silts. Slope sediment had a distinctly brown color throughout the cored interval, 308 309 whereas shelf sediment only had a 1 to 4 cm-thick brown interval, below which the sediment color changed to grey. In the eastern part of the East Siberian Sea, the sediment was mottled 310 black below 10 cm depth. Iron-manganese concretions were found between 2 cm and 10 cm 311 depth at stations 24, 42, and 43, but were also observed at other stations on the shelf that were 312 not part of this study. Benthic macrofauna, when present, consisted mainly of brittle stars, 313 isopods, few polychaetes, and rare bivalves. All bottom waters were well-oxygenated with 314

concentrations higher than 190 µmol/l, but the shelf bottom-waters in the East Siberian Sea 315 316 had generally lower concentrations than in the Laptev Sea and bottom waters on the continental slope had lower oxygen concentrations than on the shelf. Concentrations of 317 bottom-water ammonium ranged between 0.2 µmol/L and 1.8 µmol/L. Generally, the slope 318 stations and the shelf stations nearest to the Lena delta had the highest ammonium 319 320 concentrations, whereas the other shelf stations showed no clear regional trend other than proximity to the Lena delta. Bottom water dissolved inorganic carbon concentrations varied 321 322 between 2086 µM (Station 53) and 2598 µM (Station 27), and the stable carbon isotope composition of DIC in the waters overlying the cores were between -0.5 ‰ and -6.5 ‰ vs. 323 VPDB. 324

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326 **3.2.** Dissolved oxygen, manganese, and iron

We show representative profiles of oxygen concentrations in Figure 2 for the Laptev 327 Sea slope station 1, the Laptev Sea shelf stations 23, 30, 45, and the East Siberian Shelf Sea 328 329 53 and 63. Oxygen penetration depths varied between 3 mm at Station 58 and more than 60 330 mm in all slope sediments (Table 2). For the Laptev Sea slope stations 1, 2, 3, and 4, the maximum depth of oxygen penetration could not be determined, because at penetration 331 332 greater than 60 mm the conical sensor needle opened a hole in the sediment through which oxygen-containing bottom water could potentially have entered the sediment at depth thereby 333 334 artificially extending the oxygen penetration depth. In the slope-to-shelf transects the oxygen penetration depth decreased from >60 mm off-shore to 10 mm at the most inshore station in 335 336 the Laptev Sea and the East Siberian Sea. At the two easternmost shelf stations 58 and 63, we measured the lowest oxygen penetrations depths, 3 mm and 4 mm, respectively. Evidence for 337 338 bioturbation and bioirrigation based on multiple microelectrode profile measurements per core was rare. Only at Station 48 a clear increase in oxygen concentrations below the 339 sediment surface was observed, indicative of active bioirrigating macrofauna. However, even 340 at this station, based on investigations in parallel multicore casts, fauna was not abundant (A. 341 Gukov, pers. comm.). At all other stations, oxygen concentrations decreased exponentially 342 with depth. Fitting of the oxygen concentration profiles to the steady advection-diffusion-343 transport model (Eq. 1) yielded fluxes that varied between 0.81 and 11.49 mmol m⁻² d⁻¹ 344 (Table 2). These calculated O₂ fluxes compared well with total oxygen uptake rates calculated 345 from whole-core incubations using 2D optode sensor spots (Table 2). The good fit between 346

the two methods also supports the notion that bioirrigation and bioturbation effects frommeiofauna and macrofauna were minor.

In the slope sediment at Station 1, concentrations of dissolved manganese and iron 349 350 were low throughout the cored depth interval and below 0.2 and 0.5 µM, respectively. The 351 exception was a small increase for both elements between 4 and 8 cm depth and 14 and 20 cm 352 depth to concentrations of less than 3 µM, possibly due to slightly more degradable organic material in these depth intervals (Fig. 2). A similar concentration profile was found for the 353 other slope station 4 (data not shown), but here concentrations were below $2 \mu M$ throughout 354 for both dissolved iron and manganese and only slightly higher in the topmost cm. On the 355 shelf, in the Laptev Sea (Station 23 and 30), concentrations of dissolved manganese and iron 356 357 were below 0.3 μ M and 1.5 μ M, respectively, in the top 2 cm and 3 cm at Station 23, before 358 increasing to maximum concentrations of 69 and 134 μ M. At both stations, metal 359 concentrations decreased again below the concentration maximum indicating that deeper buried sediment was not a source of the metals and that the dominant source of iron and 360 manganese was reduction in the topmost 5 cm of sediment. There was a general trend of 361 362 increasing manganese concentrations from west to the east. At Station 30 in the Laptev Sea, the concentration of dissolved manganese was less than 0.3 µM in the topmost cm, but 363 increased steeply before increasing to maximum concentrations of 189 µM at 9 cm depth. 364 365 Similarly, dissolved iron concentrations were below 1 μ M to 4 cm depth and then increased to 144 µM. Again, below the maximum, both iron and manganese porewater concentrations 366 367 decreased with increasing sediment depth. Even higher iron and manganese concentrations 368 were found in the East Siberian Sea (Stations 45, 53, 63), where dissolved manganese already increased from the bottom water to concentrations of 20 µM in the topmost centimeter of 369 sediment, and iron increased to above 1 µM below 2 cm depth. The steepest manganese 370 concentration gradient was found at Station 63 in the easternmost East Siberian Sea, where 371 concentrations were 501 μ M in the first cm of sediment with a concentration maximum of 372 373 548 μ M at 2.5 cm depth and decreasing below this depth to 115 μ M at 30 cm depth. Station 63 differed with respect to dissolved iron concentrations from the other stations, because here 374 375 dissolved iron showed two small peaks at 3.5 cm and 7 cm, and concentration increased 376 substantially only below 17 cm depth to concentrations of $189 \,\mu$ M.

377

378 **3.3.** ³⁵S-sulfate reduction rates and porewater sulfate

Sulfate concentrations showed minor depth gradients at all sampling sites (Fig. 3) and 379 decreased from starting concentrations between 23.9 mM and 28.1 mM by 0.4 mM to 2.5 mM 380 from the top to the bottom of the cores. At all stations, turnover of ³⁵S-tracer was recorded 381 from the topmost sediment interval to the bottom of the core indicating active bacterial sulfate 382 reduction (Fig. 3). Depth-integrated rates over the recovered core lengths varied between 0.03 383 and 1.41 mmol m⁻² d⁻¹ (Table 2). The integrated rates were lowest at Station 1 at 3146 m in 384 the Laptev Sea and highest at the Station 63 in the easternmost East Siberian Sea. Across the 385 shelf (Stations 6 to 24), depth-integrated rates increased from the west to the east. Example 386 depth profiles of depth-specific sulfate reduction rates are shown in Fig. 3 for the same six 387 stations as previously. At Station 1, these rates ranged from 0.03 to 0.38 nmol cm⁻³ d⁻¹. At this 388 station, the variability between replicate cores was large, which is attributed to the fact that 389 many rates were near the detection limit in our handling procedure. Overall, sulfate reduction 390 391 was higher in the top 10 cm of sediment, but showed no pronounced change with depth at this station. This suggests that the reactivity of the organic material did not change substantially 392 393 over the cored depth interval. The second slope station, Station 4, showed a similar rate-depth profile than Station 1. Depth profiles for the mid-outer shelf stations 23 to 63 all showed 394 395 broad sub-surface maxima between 2.5 and 17.5 cm, but the depths of the rate maxima differed between the different stations (Fig. 3). Peak rates varied between 0.6 nmol cm⁻³ d⁻¹ at 396 Station 30 and 39 nmol cm⁻³ d⁻¹ at Station 63. The second highest rate, 7.6 nmol cm⁻³ d⁻¹, was 397 found at the station nearest to the Lena delta, Station 23. At all stations, sulfate reduction rates 398 decreased from the maxima to rates below 1 nmol cm⁻³ d⁻¹ or to below the detection limit at 399 the bottom of the cores. A particularly sharp decrease in the sulfate reduction rate was 400 observed between 8 and 9 cm at Station 63, where rates dropped from 8.5 to 0.1 nmol cm⁻³ 401 day⁻¹ over 1 cm depth. Since sulfate was abundant throughout the cored intervals, this order-402 of-magnitude decrease indicates substantial changes in the reactivity of buried organic matter. 403 Although no abrupt change in grain size or organic carbon was observed in this core, it is 404 likely that a historical change in organic sedimentation took place during deposition across 405 this time interval. 406

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408 3.4. Porewater dissolved inorganic carbon (DIC), ammonium (NH₄⁺), and $\delta^{13}C_{DIC}$

409 Porewater concentrations of dissolved inorganic carbon (DIC) and ammonium (NH_4^+)

410 increased with depth at all stations (Fig. 4). The increase of DIC was between 0.6 mM

(Station 23) and 2.3 mM (Station 45) over the cored sediment depths and ammonium 411 412 concentrations increased between 16.8 μ M (Station 1) and 549 μ M (Station 50). The steepness of the depth gradients was consistent with the rates of oxygen uptake and bacterial 413 sulfate reduction for the different stations. The porewater pattern at Station 63 is an exception, 414 because this station had the highest oxygen uptake and the highest sulfate reduction rates of 415 all stations, but showed only a modest increase in DIC and NH₄⁺ concentrations by 1.5 mM 416 and 57 µM, respectively, over the cored sediment depth. This apparent discrepancy can be 417 explained by the very low rates of sulfate reduction below 10 cm depth. Since these deeper 418 419 layers have not produced large amounts of DIC and NH₄⁺, only the surface 10 cm contribute significantly to total carbon mineralization and ammonium production. 420

For the anoxic parts of the sediment, DIC/NH₄⁺ ratios varied between 8.4 for Station 24 and 421 422 53 and 58, respectively for Stations 1 and 4 in the Laptev Sea, and between 7.2 and 18.8 in the East Siberian Sea, with an overall mean DIC/NH4 ratio of 9.8 for all stations excluding the 423 continental slope stations. The δ^{13} C values of DIC consistently decreased with sediment depth 424 indicating the addition of ¹³C-depleted remineralized carbon to DIC. The greatest downcore 425 depletion in ¹³C was observed at Stations 45, 48, and 50, where δ^{13} C of DIC decreased from -426 2.0 ‰ near the sediment surface to -13.9, -16.4, and -18.6 ‰ at the bottom of the cores (Fig. 427 428 4).

429

430 **3.5. Modelled oxygen, iron, and manganese reduction rates**

431 Results of the reaction transport modelling of dissolved oxygen, iron, and manganese concentration profiles for Station 23 are shown in Fig. 5. O₂ consumption rates exceeded 432 433 sulfate, iron, and manganese reduction rates by a factor of more than 100 (Fig. 5). For the 434 shelf stations, most of the carbon oxidation therefore takes place in the topmost 5 mm. The 435 reaction rate profiles for iron and manganese reduction indicate that manganese reduction dominates in the topmost 2 cm of sediment followed by co-existing iron and sulfate reduction 436 below (Fig. 5). These observations are consistent with results from the northern Barents Sea 437 by Vandieken et al. (2006) and Nickel et al. (2008). Optimal fits of the concentration profiles 438 required a bioirrigation coefficient of $1 \times 10^{-4} \text{ cm}^2 \text{ sec}^{-1}$ in the topmost 2 cm of sediment at 439 Stations 23 and 53. For the other stations, optimal fits required no sediment mixing by 440 bioturbation or advective porewater transport by bioirrigation. This result is consistent with 441 the modelling results of the oxygen microelectrode profiles and the low numbers of 442

bioturbating macrofauna in the outer shelf sediment. Bacterial sulfate reduction was detected 443 444 already at a depth where the sediment was still brown indicating abundant iron oxyhydroxides. It is therefore likely that the modelled negative iron production rates at the 445 sediment surface indicate iron oxidation in the mixed upper layer. This pattern was not 446 observed for manganese, which is consistent with incomplete manganese oxidation at the 447 sediment surface and loss of dissolved manganese to the bottom water. Manganese and iron 448 reduction contributed between 2.3 and 23.7% to the total anaerobic carbon mineralization and 449 between 0.3 and 2.3% to the total carbon mineralization (Table 3). Although these numbers 450 451 may somewhat underestimate the contribution of metals to carbon mineralization, our results 452 indicate that bacterial sulfate reduction is by far the major anaerobic carbon mineralization 453 pathway in these sediments.

454

455 4. Discussion

456 4.1. The relative importance of iron, manganese and sulfate reduction for carbon 457 degradation

There was a statistically significant positive correlation between the dissolved oxygen 458 uptake and anaerobic carbon degradation rates by sulfate reduction with an r^2 of 0.72 (P < 459 0.05). This reflects the coupling of oxygen uptake to the oxidation of reduced inorganic 460 metabolites (FeS and H₂S) produced during the anaerobic metabolism by sulfate reduction 461 (e.g., Glud, 2008; Jørgensen and Kasten, 2006; Thamdrup, 2000; Berg et al., 2003) (Fig. 9). 462 The slope of the regression line for the data set is 6.1 ± 1.1 indicating that about 16.4% of the 463 oxygen uptake is used for the oxidation of reduced manganese, ammonium, dissolved iron, 464 465 and iron sulfides and elemental sulfur. This amount is slightly lower than the average 23% estimated for oxygenated coastal and continental shelf sediment (Canfield et al., 2005), but is 466 467 consistent with the notion that a substantial amount of the buried organic matter in Siberian shelf sediment is oxidized anaerobically. The lower proportion of anaerobic respiration to 468 469 aerobic respiration compared to other shelf environments likely reflects the greater proportion of highly reactive marine-derived organic material in the topmost millimeters of sediment. 470

The prevalence of bacterial sulfate reduction in anaerobic carbon mineralization agrees withthe assessment made by Vandieken et al. (2006) and Nickel et al. (2008), who suggested that

more ice-free stations in the northern Barents Sea supported higher rates of sulfate reductionthan the more permanently ice-covered stations reflecting lower carbon export production.

The efflux of manganese to the bottom water on the eastern Siberian shelf supports the 475 476 assessment by Macdonald and Gobeil (2012) that Arctic shelves can export dissolved 477 manganese to the Arctic interior. Coexistence of net iron reduction and sulfate reduction at the 478 same depths make it difficult to quantify how much of the iron reduction is coupled to heterotrophic carbon oxidation and to the re-oxidation of sulfide produced from bacterial 479 sulfate reduction. Qualitatively, the presence of dissolved iron throughout the measured 480 porewater profile implies that iron reduction exceeded concomitant sulfate reduction, iron 481 sulfide precipitation, and reoxidation reactions, which supports the assessment of net 482 483 heterotrophic iron reduction. However, previous investigations of the importance of iron and manganese reduction in Arctic shelf sediments have emphasized the importance of directly 484 coupled redox processes between iron and manganese (Vandieken et al., 2006). It is also 485 important to note that iron and manganese oxyhydroxides can sorb Mn²⁺ and Fe²⁺ (Canfield 486 et al. 1993). The concentrations of dissolved Fe^{2+} and Mn^{2+} may therefore underestimate the 487 actual concentrations of the reduced forms in these sediments. 488

Iron hydroxide surfaces have been inferred as important mineral surfaces for the 489 490 preservation of organic matter (Lalonde et al., 2012; Salvado et al., 2016). In all cases studied here, the integrated net DIC production based on the porewater gradient of DIC, and the depth 491 492 profiles of iron reduction indicate co-existing heterotrophic and chemical iron reduction and bacterial sulfate reduction. In addition, the porewater modelling results suggest that 493 494 bioturbation can be an important sediment mixing process only for some shelf stations. 495 Organic matter sorbed to mineral surfaces with deposition would thus have been subject to 496 repeated desorption as iron oxyhydroxides were reduced. While this observation does not 497 contradict the observation that some organic material is buried in association with iron oxyhydroxides, the repeated redox cycling of the oxyhydroxides would prevent the sorptive 498 preservation of organic compounds. 499

500

501 4.1. Marine versus terrestrial organic matter contribution

502 Terrestrial organic carbon sources to the Laptev and East Siberian shelf and slope are 503 riverine discharge and coastal erosion of the ice core complex (Stein and Macdonald, 2004;

Vonk et al., 2012; Rachold et al., 2004; Fahl and Nöthig, 2007; Semiletov, 1999). Marine 504 505 organic carbon is derived from open-water production during the ice-free months, export of ice algae, and new production in polynyas (Sakshaug et al., 2004; Nitishinsky et al. (2007). 506 507 Generally, marine productivity in the Laptev Sea is low and controlled by the nutrient concentrations derived from Atlantic water, but spring outflow from the Lena River provides 508 an additional temporary land-derived nutrient source (Pivovarov et al., 1999; Sakshaug et al., 509 2004; Nitishinsky et al., 2007; Bourgeois et al., 2017) during late spring ice melt (Raymond et 510 al., 2007). Terrestrial-derived nutrients can also affect marine productivity either directly by 511 512 new production, or indirectly, due to plankton production from remineralized terrestrial DOC/POC (Alling et al., 2012). In the eastern East Siberian and Chukchi Sea, the inflow of 513 514 nutrient-rich Pacific water supports higher marine primary productivity (e.g., Grebmeier et al., 2006). Ice-rafted transport and bottom boundary layer transport are the two most important 515 516 modes of particle transport (Wegner et al., 2005; Bauch et al., 2009). Since all sediments sampled in this study were fine-grained siltey clays and clayey silts, coarse-grained woody, 517 518 ice-rafted material played only a minor role for deposition of organic matter on the outer shelf and slope sediment. The transport direction of inner shelf sediments has been suggested to 519 520 follow the predominant atmospheric regime, which is thought to be linked to the Arctic 521 Oscillation (AO) (Dimitrenko et al., 2008; Guay et al., 2001; Weingartner et al., 1999). During positive AO southwesterly winds lead to generally eastward transport and repeated 522 inshore transport in the BBL, whereas negative AO favors southerly winds and a 523 predominantly northward transport (Guay et al., 2001; Dmitrenko et al., 2008). Offshore 524 525 transport of dissolved and particulate organic matter from the Lean delta to the north can occur with the Transpolar Drift, but terrestrial organic material is also transported eastward 526 527 and obliquely offshore with the Siberian coastal current receiving additional organic material from the Indigirka and Kolyma rivers (Guo et al., 2007; Dudarev et al., 2006). East of 140°E, 528 529 the influence of Pacific-derived nutrient-rich water supporting marine production is stronger the further east and offshore the sampling stations are located (Semiletov et al., 2005) (Fig. 1). 530

Carbon degradation rates in the sediment across the whole Siberian shelf and slope
reflect this temporally and spatially diverse distribution of nutrient availability, ice cover,
sediment deposition, and current flow regime (Rachold et al., 2004; Dudarev et al., 2006;
Semiletov et al., 2005; Sakshaug et al., 2004; Dmitrenko et al., 2005). The proportion of
degradable marine-derived organic material at the eastern Stations 50 to 63 on the East
Siberian shelf is higher than at the western stations in the Laptev Sea, in line with higher

nutrient availability due to the Pacific influence. Ice-free conditions and the opening of water 537 538 due to northward migration of ice shortly before the sampling likely supported new algal primary production at the shelf stations closest to land leading to enhanced export and 539 deposition on the seafloor. During the time of sampling, only Stations 6 to 27 were ice-free, 540 while Stations 23 and 24 had the longest ice-free condition before sampling. By contrast, 541 542 Stations 30 to 63 were still covered by ice during sampling. New export of reactive organic material explains why O₂ uptake rates were the highest at stations 23 and 24 along the shelf to 543 544 slope transect from station 1 to station 24 (Boetius and Damm, 1998). The same pattern as for 545 the O₂ uptake rates is also observed for the sulfate reduction rates indicating that reactive organic matter is also buried below the oxygen penetration depth and mixed layer into the 546 547 sulfate-reducing zone. This indicates that a greater portion of reactive organic material is buried closer to the Lena delta. 548

Published organic carbon budgets for the Arctic shelves infer an average burial 549 efficiency of about 1% of exported marine OC (Stein and Macdonald, 2004), while terrestrial 550 organic carbon, only accounting for about 10% of the organic carbon delivered to the Arctic 551 552 Ocean bottom, has been suggested to be preserved with about 90% efficiency (Macdonald et al., 2015). Recently, Semiletov et al. (2016) compiled a large dataset indicating substantial 553 aragonite undersaturation of Arctic shelf bottom waters from the Laptev, the East Siberian, 554 and the Russian part of the Chukchi Sea, which was interpreted due to the remineralization of 555 terrestrial organic matter. The observation of strongest aragonite undersaturation in the 556 bottom waters supports a sediment-derived CO₂ source or a stagnant bottom boundary layer 557 558 (Semiletov et al., 2013). It is therefore possible that oxic carbon mineralization in the topmost mm of sediment is a major CO₂ source for the overlying water. 559

560 Fig. 6 shows exemplary gradients of the regression for the six stations presented before and 561 Table 4 lists the derived carbon isotope compositions of remineralized organic matter for all stations. The range of δ^{13} C of remineralized DIC varied between -18.8 ‰ ± 1.1 ‰ (Station 562 53) and -35.8% \pm 3.0 % (Station 1). The strongly ¹³C-depleted isotope composition of -35.8 563 ‰ for remineralized DIC at Station 1 suggests the mineralization of strongly ¹³C-depleted 564 organic matter and possibly a strong contribution of terrestrial organic matter to carbon 565 mineralization far offshore, in line with the very high DIC/NH₄⁺ ratio of the porewaters at the 566 slope stations. The potential existence of degradable terrestrial organic matter in slope 567 sediments of 3000 m water depth is intriguing, since it would imply downslope transport and 568 569 degradation of terrestrial organic matter. Northward off-shelf transport of terrestrial organic

matter with the Transpolar Drift is a viable transport mechanism. The contribution of
degradable terrestrial organic matter to DIC in lower slope sediments is also supported by the
observation of terrestrially derived biomarkers in porewater DOC of central Arctic Ocean
sediment analyzed by FT-ICRMS (Rossel et al., 2016) and deep-water sediment trap data in
the central Arctic Ocean (Fahl and Nöthig, 2007), but requires further investigation.

575 The isotope composition of the remineralized DIC therefore reflects mineralization of a mixture of organic molecules of different origins – interpreted here as a mixture of 576 terrestrial and marine-derived organic matter. For the following discussion, given the 577 uncertainty of the organic matter origin in slope sediment, we exclude data from the slope 578 579 stations and restrict the discussion to the use of the following end member compositions for 580 the shelf stations. For the Laptev Sea shelf, we account for the fact that a fraction of the DIC 581 used for marine production is derived from remineralized terrestrial DOC and POC in shelf waters. Alling et al. (2012) report $\delta^{13}C_{DIC}$ values for offshore DIC samples below the 582 halocline varying between -2 and -4 ‰. We therefore use an isotope endmember for marine 583 584 organic matter of -24 ‰ and an isotope composition of -28 ‰ for the terrestrial organic carbon contribution (Alling et al., 2010; Vonk et al., 2012). For the East Siberian Sea East of 585 586 140°E, the heaviest calculated isotope composition of remineralized DIC was -19 ‰ and is 587 used here as the marine endmember (Station 53). The same carbon isotope composition of -28 ‰ as for the Laptev Sea was used as the terrestrial end member. The heavier marine δ^{13} C 588 value in the East Siberian Sea is supported by the slightly heavier $\delta^{13}C_{DIC}$ values reported for 589 the offshore East Siberian Sea, which vary between 0 and -2 ‰ (Alling et al., 2012). 590

The calculated mass fractions of the two endmembers are listed in Table 4. Based on this analysis the porewaters on the Laptev Sea shelf contain a significant proportion of terrestrially derived organic matter, comprising on average 36 % of the remineralized DIC. This proportion decreases to average values of 32% in the East Siberian Sea, in line with a greater marine production in this area due to the inflow of Pacific water (Semiletov et al., 2005, Dudarev et al., 2006; Naidu et al., 2000).

597 Since ³⁵S-sulfate reduction rates comprise most of the anaerobic carbon mineralization 598 of sediment buried below the oxygen penetration depth, our assessment includes, in contrast 599 to earlier studies, the mineralization rates of terrestrial organic matter beyond the short time 500 period of oxygen exposure in the topmost mm of sediment. Using sedimentation rates of 0.8 501 mm y⁻¹ for the outer Laptev Sea (Strobl et al., 1988) and 1.4 mm y⁻¹ for the outer East

Siberian Sea (Bröder et al., 2016b), the recovered sediments record a time interval of 250 to 602 700 years since burial. Using the mass fractions of terrestrial and marine-derived organic 603 carbon listed in Table 4, respective mineralization rates of the terrestrial and marine carbon 604 fractions were calculated from the product of the mass fractions and the depth-integrated ³⁵S-605 sulfate reduction rates (Table 2). This approach is only applicable in combination with depth-606 607 integrated anaerobic carbon mineralization rates, but would be biased if used in combination with total oxygen uptake rates. The reason for this is that the depth of oxygen penetration 608 609 varied only between a few millimeters to little more than a centimeter on the shelf, whereas 610 the corresponding DIC concentrations, even in the topmost centimeter of sediment, are 611 affected by diffusive exchange along the 30 cm-long concentration profile smoothing out 612 depth-dependent changes in the source signal of organic matter. It is therefore not possible to assess the relative fractions of terrestrial and marine organic matter mineralized for discrete 613 614 depth intervals. Our combined radiotracer and DIC stable isotope approach suggests that both marine and terrestrial organic matter are degraded in the buried sediment and that both pools 615 616 contribute to degradation products in anoxic buried sediment. This assessment is a significant modification to earlier studies by Boetius and Damm (1998) and Bourgeois et al. (2017), who 617 618 have described organic matter mineralization in Siberian Arctic sediments largely as a 619 function of oxygen uptake.

620 Carbon mineralization rates measured along the transect near 130°E (Stations 1 through 24) reflect the influence of gradual offshore transport of terrestrial organic material (Bröder et 621 622 al., 2016a) (Fig. 7A, B). A comparison with the oxygen uptake rates reported by Boetius and 623 Damm (1998) indicates that all rates measured in 2014 were significantly higher than the rates measured in 1993 by Boetius and Damm (1998). Although the different rates may reflect a 624 seasonal effect since Boetius and Damm's data were acquired later in the year than our data, 625 the increase may also point to higher organic carbon mass accumulation rates compared to 20 626 years ago, consistent with a decrease in the annual ice cover over the past 20 years in the 627 628 Arctic (Arrigo and van Dijken, 2011; Stroeve et al., 2012; Walsh et al., 2017). Whether these rates reflect higher marine and/or terrestrial accumulation cannot be answered satisfyingly 629 630 with this data set.

Fig. 8A compares the oxygen uptake rate of the stations of this study with averaged
oxygen uptake rates from the literature for different shelf, slope, and abyssal plain
environments worldwide (Canfield et al., 2005). The data suggest that there is no significant
difference in the oxygen consumption rates between the Siberian shelf and slope and other

continental margin environments. ³⁵S-sulfate reduction rates in Sea Siberian slope sediment 635 are also comparable rates to those in other slope environments (Fig.7B and 8B), but the 636 sulfate reduction on the shelf are lower by a factor up to 15. Another difference apparent from 637 this comparison is the similarity in sulfate reduction rates for the outer shelf and continental 638 slope sediments of the Siberian Arctic (Fig. 8B). This similarity is noteworthy for several 639 reasons: 1) it suggests that the kinetics of anaerobic carbon degradation in the shelf and slope 640 sediments reflect similar reactivity of the organic matter. This is surprising since 641 accumulation rates on the continental slope are significantly slower than on the outer shelf. 2) 642 643 The absolute magnitude of the sulfate reduction rates in shelf and slope sediment indicate significant rates of organic matter mineralization long after burial consistent with the 644 substantial DIC flux and the strongly ¹³C-depleted DIC carbon isotope composition. Overall, 645 the data indicate that organic matter reactivity substantially changes during burial in shelf 646 647 sediment, but that the reactivity of transported organic matter that is exported to deep water across the shelf does not decrease significantly supporting long-term slow mineralization rates 648 649 in the slope environment. Accumulation of the organic material on the slope may therefore be related to rapid downslope transport of organic material or a rapid offshore transport, e.g., due 650 651 to transport with ice or as bottom nepheloid layers cascading from the shelf edge (Ivanov and 652 Golovin, 2007).

653

654 4.2. Assessment of carbon burial efficiency

Reported ²¹⁰Pb-based sediment accumulation rates in outer Siberian shelf sediment 655 range between 0.05 ± 0.02 g cm⁻² y⁻¹ in the Laptev Sea (Strobl et al., 1988) and 0.24 ± 0.04 g 656 cm⁻² y⁻¹ in the East Siberian Sea (Bröder et al., 2016b). Given surface sediment organic 657 carbon concentration for this area between 1% and 1.5%, the resulting organic carbon mass 658 accumulation rates vary between 1.1 mmol m⁻² d⁻¹ and 1.7 mmol m⁻² d⁻¹ for the Laptev Sea 659 (area near Station 23) and 5.5 and 8.2 mmol m⁻² d⁻¹ in the East Siberian Sea (data for Station 660 63). We estimated the burial efficiency of terrestrial organic carbon from the ratio of the 661 depth-integrated sulfate reduction rates relative to the ²¹⁰Pb mass accumulation rate of organic 662 carbon. This treatment assumes that the reported organic carbon mass accumulation rates 663 largely reflect the refractory component of organic matter. While it is possible that a fraction 664 of terrestrial and marine organic matter is degraded on shorter time scales than captured by 665 the ²¹⁰Pb method, we assume that the fraction of highly reactive terrestrial organic matter 666

667 missed in this treatment is small. The resulting burial efficiency of the terrestrial carbon 668 fraction is on average 91 ± 6 % in the Laptev Sea and 94 ± 4 % for the East Siberian Sea. We 669 also calculated apparent degradation rate constants of organic matter assuming first order 670 degradation kinetics for the time duration of sediment burial recorded in the sediment cores. 671 For this assessment, we used the total depth-integrated anaerobic carbon mineralization 672 determined from the combined manganese, iron, and sulfate reduction rates for the recovered 673 sediment. The apparent annual degradation rate constant (k) was then calculated from

674
$$k_{terrestrial} = \frac{\left(-ln \frac{\int_{0}^{30} OC_{accumulation} - \int_{0}^{30} OC_{total mineralization}}{\int_{0}^{30} OC_{accumulation}}\right)}{t_{burial}}$$
(5)

where the integrals of $OC_{accumulation}$ and $OC_{mineralization}$ cover a period of 250 years to 700 years based on the ²¹⁰Pb mass accumulation rates. The resulting annual degradation rate constant (*k*_{terrestrial}) ranges between 1 x 10⁻⁴ y⁻¹ and 5 x 10⁻⁴ y⁻¹ averaging 1.5 x 10⁻⁴ y⁻¹ in the Laptev Sea and between 8 x 10⁻⁵ and 3 x 10⁻⁴ y⁻¹ averaging 1.2 x 10⁻⁴ y⁻¹ in the East Siberian Sea.

A comparison of the total oxygen uptake with the Corg mass accumulation rates 679 indicates that the 210 Pb-based C_{org} mass accumulation rates on the shelf are equal or 680 significantly lower than the oxygen uptake rates, with a discrepancy of up to a factor 10. 681 Since the derivation of the 210 Pb-based C_{org} mass accumulation rates is based on the same 682 depth range as our direct ³⁵S-based degradation rate measurements (30 cm of sediment, Vonk 683 et al., 2012), Corg mass accumulation rates and degradation rate measurements cover the same 684 time window of sediment burial. Temporal variation in sediment accumulation therefore 685 cannot explain the discrepancy. In addition, methane seep sediments where upward transport 686 687 of methane from deeper sediment layers contributed to oxygen uptake were excluded from this data set. The best explanation for the discrepancy is therefore that the ²¹⁰Pb-mass 688 689 accumulation rates underestimate the true mass accumulation rate of highly reactive organic material and that this material is oxidized at the sediment surface. Based on the measured 690 691 oxygen uptake rates this freshly deposited organic material has substantially higher degradation rates within the top mm of sediment as reflected by the steep O₂ gradients. ²¹⁰Pb-692 693 based organic carbon accumulation therefore reflects the long-term burial of less reactive organic material in the top 30 cm of sediment. Since anaerobic degradation processes prevail 694 below the O₂ penetration depth, the measured burial efficiency of the accumulating organic 695 material is therefore a function of the anaerobic bacterial degradation rather than the aerobic 696 697 degradation efficiency. This conclusion has implications regarding the assessment of potential aerobic degradation of reactive terrestrial organic matter, since degradation of such material
 would have gone undetected with ²¹⁰Pb-based accumulation rate measurements.

700

701

702 **4.5. Regional estimates**

703 We present areal estimates of sediment carbon mineralization by extrapolating the 704 measured carbon mineralization rates over the outer Laptev Sea and East Siberian Sea shelf. 705 Such extrapolations of benthic carbon mineralization rates are notoriously difficult given sediment heterogeneity and insufficient temporal data coverage of benthic carbon 706 707 mineralization rates. For this investigation, no near-shore or slope stations were included in the assessment. The near-shore Siberian shelf environments are under much stronger 708 709 influence by coastal erosion and riverine discharge than the outer shelf stations and have considerable longer open-water conditions than the outer shelf stations investigated here. In 710 711 addition, the sedimentation pattern in the near-shore environments is significantly more diverse, which will affect sedimentation rates, grain size distribution, and carbon contents. For 712 this reason, we did not extend our extrapolations to the inner shelf environments. Some of 713 these inner shelf settings likely have much higher benthic carbon mineralization rates and 714 additional studies are required to constrain these better. Our coverage of the slope stations is 715 insufficient for meaningful spatial extrapolations. A large data set for this region has been 716 717 analyzed by Miller et al. (2016) and the reader is referred to this work.

We estimate the extent of the outer shelf area with depositional conditions comparable 718 719 to those investigated here to cover approximately 280,000 km² of the Laptev Sea. For the East Siberian Sea, we estimate the respective area of the outer shelf to be 340,000 km². Due to the 720 stronger terrestrial influence in the Laptev Sea, we calculated rates separately for the two shelf 721 seas. The areal coverage with sediment stations was too sparse for statistically significant 722 interpolations between stations that would give reliable spatial accounts of the gradients in 723 rates between the stations. Instead, arithmetic averages of sediment mineralization rates and 724 fluxes were calculated for these regions. Accepting the uncertainties in our assessment and 725 726 data density, we estimate that the calculated areal rates could deviate by up to 50%. Table 5 727 lists the calculated rates based on the average flux calculated per square meter per day for 728 oxygen uptake, DIC flux, bacterial sulfate, and total anaerobic carbon mineralization. For the

latter three methods, the total flux was calculated for the marine and terrestrial component, 729 730 respectively. The same analysis cannot be performed for the oxygen uptake for the reasons discussed in section 4.3. Since the major part of the oxygen uptake is likely associated with 731 degradation of a highly reactive marine organic carbon component, the proportions calculated 732 based on the δ^{13} C composition of DIC would not necessarily apply to the topmost mm of 733 sediment. It is noteworthy to say that the rates calculated with our data set agree well with the 734 O₂ uptake rates recently published by Bourgeois et al. (2017) for the Laptev Sea. Our 735 calculations suggest that 5.2 and 10.4 Tg O₂ y⁻¹, respectively are taken up by the outer shelf 736 sediment in the Laptev and East Siberian Sea, respectively, totaling 15.9 Tg y⁻¹ for the whole 737 investigated area (Table 5). Anaerobic carbon mineralization based on DIC, ³⁵S-SRR and 738 combined manganese, iron, and sulfate reduction range between 0.62 and 1.28 Tg y^{-1} . Of the 739 total anaerobic carbon mineralization, between 0.25 and 0.48 Tg y⁻¹ can be attributed to the 740 oxidation of terrestrially derived organic material. This rate is five to ten times lower than the 741 742 estimated annual water column degradation of particulate terrestrial organic matter in the Eastern Siberian Arctic shelf system of $2.5 \pm 1.6 \text{ Tg y}^{-1}$ (Sanchez et al. 2011), and only 743 between 0.5% and 2% of the annual organic carbon export from land (Stein and Macdonald, 744 2004; Vonk et al., 2012). 745

746

747 **5.** Conclusions

748 Directly measured carbon mineralization rates together with stable isotope and concentration data of East Siberian Arctic shelf and slope porewaters indicate that about one 749 750 third of the remineralized organic carbon in porewater DIC is derived from terrestrial organic 751 matter. This conclusion confirms and extends previous observations that terrestrial organic 752 carbon buried in Siberian shelf and slope sediment is not conservative (Semiletov et al., 2013; 753 Karlsson et al., 2015; Bröder et al., 2016b). While mineralization of terrestrial organic material has been described for the water column and resuspended surface sediment, our data 754 indicate that mineralization also proceeds long after burial in sediment. The estimated 755 apparent carbon degradation rate constants of transformed terrestrial organic matter on the 756 outer shelf are slow ($< 3 \times 10^{-4} \text{ y}^{-1}$) and the overall terrestrial carbon burial efficiency is 757 relatively high (> 87 %) and only slightly lower than previously reported based on millennial-758 scale carbon burial rates (> 90 %, Stein and Macdonald, 2004). Area-integrated rates of 759 carbon mineralization in the outer shelf sediments (0.29 to 0.48 Tg y^{-1}) represent about 0.5 % 760

to 8 % of the annual terrestrial organic matter load to the Laptev and East Siberian Sea 761 ranging from 6 Tg y⁻¹ (Stein and Macdonald, 2004) to 22 ± 8 to 44 Tg y⁻¹ (Vonk et al., 2012). 762 There are large uncertainties associated with these estimates, given that our calculations do 763 not account for carbon mineralization of resuspended terrestrial organic material and likely 764 higher rates of mineralization in the inner shelf sediments. Nevertheless, these data indicate 765 that the contribution of the benthic DIC flux to the total CO₂ production in the outer Eastern 766 Siberian Sea and Laptev Sea is small. This conclusion, however, does not necessarily extend 767 to the inner parts of the Laptev Sea and the western parts of the East Siberian Sea, where CO₂ 768 769 supersaturation has been reported by Semiletov et al. (2012) and Pipko et al. (2011). Anderson et al (2009) estimated a DIC excess of 10 Tg C by evaluating data from the Laptev 770 771 and East Siberian Seas collected in the summer of 2008 and suggested that this excess was caused mainly by terrestrial organic matter decomposition. Their estimate can be compared to 772 773 our sediment oxygen uptake for the outer Laptev and East Siberian Sea shelf of almost 16 Tg y^{-1} , which would demand that 62.5 % of the oxygen uptake was due to terrestrial organic 774 775 matter mineralization. However, the reported annual production of marine organic matter for the total Laptev and East Siberian Sea is about 46 Tg y⁻¹ (Stein and Macdonald, 2004). Even 776 777 if only half of this amount is produced in the outer shelf region and only another half of that amount was deposited, there would still be more than 10 Tg y^{-1} of reactive marine organic 778 matter at the sediment surface. Our data would therefore suggest that at least in the more 779 productive East Siberian Sea the pronounced aragonite undersaturation reported for bottom 780 waters in the East Siberian Sea is due to aerobic mineralization of a significant amount of 781 marine organic matter, which extends the assessment for the western Chukchi Sea and the 782 central Arctic Ocean by Qi et al. (2017). It is apparent that these sediments play a major role 783 in the recycling of marine organic carbon on the Arctic shelf. Future changes in marine 784 production on the Siberian shelf under longer ice-free conditions (Arrigo and van Dijken, 785 2011) will likely change the relative proportions of degrading marine and terrestrial organic 786 matter further so that this particular shelf system may in the future more strongly resemble 787 788 that of other ice-free shelf-slope environments.

789

790 6. Acknowledgements

Funding for this investigation came from the K&A Wallenberg foundation, the SwedishPolarsekretariat, and the Bolin Centre for Climate Research at Stockholm University. Igor

- 793 Semiletov acknowledge support from the Russian Government (No.
- 14.Z50.31.0012/03.19.2014). We would like to thank the members of the SWERUS-C3
- consortium, the shipcrew on icebreaker Oden, and Heike Siegmund, Lina Hansson, Barkas
- 796 Charalampos, and Dimitra Panagiotopoulou for help with the laboratory work. We dedicate
- this publication to our friend and colleague Vladimir Samarkin, who unfortunately passed
- away before publication of this work. This manuscript benefitted from discussions with
- 799 Patrick Crill, Örjan Gustafsson, Christoph Humborg, Julia Steinbach, Clint Miller, Marc
- 800 Geibel, Emma Karlsson, Brett Thornton, Jorien Vonk, Leif Anderson, and Magnus Mörth.

802 List of Tables

803

Table 1. Physical and chemical characteristics of sediment and bottom water chemical

805 characteristics at the sampled stations.

- Table 2. Summary of oxygen penetration depth, O_2 uptake, and integrated ³⁵S-sulfate reduction rates.
- 808 Table 3. Anaerobic rates of carbon mineralization by manganese, iron, and sulfate reduction
- Table 4. Calculated carbon isotope composition of remineralized DIC and mass fractions of
- 810 the marine and terrestrial end member and corresponding terrestrial carbon degradation rates
- 811 based on ³⁵S-SRR and DIC
- Table 5. Regional estimates of sediment carbon mineralization in the outer Laptev and East
- 813 Siberian shelf sea

814 List of figures

Figure 1: General map of the Laptev and East Siberian Sea with sediment stations and majorcurrent features

817

Figure 2: Depth profiles of dissolved O₂ measured with O₂ microelectrode sensors for Stations
1, 23, 30, 45, 58, and 63 and profiles of porewater concentrations of dissolved iron and
manganese.

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Figure 3: ³⁵S-SRR rates and corresponding porewater sulfate concentrations for Stations 1, 23,
30, 45, 58, and 63.

824

Figure 4: Depth profiles of dissolved inorganic carbon (DIC), $\delta^{13}C_{DIC}$, and dissolved ammonium (NH₄⁺) for Stations 1, 23, 30, 45, 58, and 63.

827

Fig. 5. Comparison of reaction rates of oxygen, manganese, iron, and sulfate reduction at
Station 23. Note the different depth scale for the O₂ consumption rate. The dashed line marks
the oxygen penetration depth.

831

Fig. 5 A, B. Map of field area and sampling stations showing oxygen uptake rates in panel A
and depth-integrated sulfate reduction rates in panel B. For comparison, oxygen uptake rates
reported in Boetius and Damm (1998) using the same color coding are shown as triangles for
comparison.

836

Fig. 6. A: Crossplot of dissolved NH4⁺ and porewater DIC* after correction for bottom water
DIC concentrations. The slopes of the regression lines for the individual stations are shown in
Table 2. B: Crossplot of the fraction of remineralized DIC calculated from a two endmember

840 mixing model versus $\delta^{13}C_{DIC}$. The slope and y-intercept of the regression for each station are 841 shown in Table 3.

842

Fig. 7 A, B. Map of field area and sampling stations showing oxygen uptake rates in panel Aand depth-integrated sulfate reduction rates in panel B.

845

Fig. 8A. Water depth variation of sediment oxygen uptake. 8B: Water depth variation of

847 integrated ³⁵S-sulfate reduction rates (0-30 cm sediment depth). For reference average rates of

848 abyssal plain, continental rise, slope, and shelf sediments, deposition and non-depositional,

849 are shown for reference.

850

Fig. 9. Crossplot of diffusive oxygen uptake and integrated sulfate reduction rates. The black line is the linear regression and yielded a y-intercept of 2.1 mmol $m^{-2} d^{-1}$ and a slope of 5.55.

Blue and red lines show the 95% and 99% confidence interval.

854

856 7. References

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Station	Latitude "N	Longitude °E	Date	Water depth	Ice cover	Bottom water salinity	Bottom water temperature	Bottom water 02 concentration	Bottom water NH4 ⁺ concentration	Bottom water DIC above sediment	8 ¹³ C DIC bottom water	Sediment description
			Month/Day/Year	m	%	%0	°C	µmol/L	μmol/L	µmol/L	‰vs. VPDB	
1	78.942	125.243	7/15/2014	3146	50 - 75	34.9	-0.9	271.9	1.65	2151.5	-0.5	clay, brown
6	78.581	125.607	7/16/2014	2900	25 - 50	34.9	-0.9	275.0		n.a.	n.a.	clay, brown
3	78.238	126.150	7/16/2014	2601	< 25	34.9	-0.9	280.0		n.a.	n.a.	clay, brown
4	77.855	126.664	7/16/2014	2106	< 25	34.9	-0.8	289.4	1.81	2164.5	-1.6	clay, brown
9	77.142	127.378	7/17/2014	89	0.0	34.6	0 -	327.0	1.30	2213.0	-2.2	clay, top 3 cm brown, then gray, fauna on top of sediment
:				i			-1.8					silty clay, top 4 cm brown.
53	76.171	129.333	7/22/2014	56	0.0	34.2	-1.8	303.2	1.34	2246.3	-3.2	then gray, brittle stars
24	75.599	129.558	7/24/2014	46	0.0	34.0	-1.7	283.8	0.89	2244.1	-2.0	silty clay, top 4 cm brown, then gray
27	76.943	132.229	7/23/2014	4	0.0	34.2	-1.8	332.3	0.94	2595.0	-6.5	silty clay, top 2 cm brown, then gray, fluffy surface layer,
												brittle stars
30	78.181	138.354	7/24/2014	69	0.0	34.1	-1.6	334.8	0.79	2178.4	-3.7	silty clay, top 4 cm brown, then grav
31	79.396	135.497	7/25/2014	3056	0.0	34.9	-0.9	270.9	0.74	2161.7	n.a.	clay, brown
35	78.600	137.061	7/26/2014	541	0.0	34.9	0.4	288.1	0.43	2183.7	n.a.	clay top 15cm brown, fluffy, inhomogeneous, surface-
												dwelling fauna
37	78.521	137.170	7/26/2014	205	0.0	34.7	0.0	295.4	0.89	2171.1	n.a.	clay, top 5cm brown, then
40	77.670	144.668	7/27/2014	45	0.0	31.5	-1.3	190.3	0.53	2213.7	-1.6	silty clay, top 3cm brown,
												then gray, brittle stars
												Survey the start of the start starts of the starts of the starts of the starts of the start starts of the starts of t
43	76.780	147.791	7/28/2014	42	25-50	30.1	-1.2	256.4	0.61	2086.7	n.a.	small durface-dwelling
												animals
45	76.416	148.115	7/29/2014	40	< 50	29.1	-1.3	319.9	0.57	2576.0	-2.1	silty clay to clayey silt, 2cm brown, then gray-black, rather
												stiff silty clay to clayary silt_ton
8	76.615	153.345	7/30/2014	49	>75	30.6	-1.6	315.9	0.50	2075.1	-2.2	3cm brown, then grayblack
50	75.764	158.529	8/1/2014	44	>75	31.1	-1.4	311.0	0.51	2068.7	-2.1	silty clay to clayet silt, top 2cm brown, then grayblack
												silty clay to clayey silt, top 3
53	74.957	161.088	8/2/2014	47	>75	31.0	-1.6	253.3	0.16	2086.1	-2.5	cm brown, then 3 cm gray,
												then grayblack silty clay to clayev silt,
58	74.440	166.050	8/4/2014	54	> 75	31.4	-1.7	254.3	0.65	2154.9	-1.5	slightly resuspended, top 2 cm
63	74.685	172.361	8/7/2014	67	> 75	32.4	-1.4	186.0	0.61	2240.8	-2.2	DIOWI, UKU BIAY, SOIL silty clay to clayey silt, top lcm brown, then gray

Table 1. Physical and chemical characteristics of sediment and bottom water at the sampled stations

						DIC flux (modelled in	
Station	Water depth	mean O ₂	mean O ₂ at 60mm	O ₂ uptake	³⁵ S-SRR (0-30 cm)	anaerobic	Average
Station	water ueptii	depth	depth	(modelled/measured with 2D optode)	duplicates	whole core	DIC/NH ⁺
				(init 22 optione)		incubation)	
	m	mm	µmol/L	mmol $m^{-2} d^{-1}$	mmol $\mathbf{m}^2 \mathbf{d}^1$	mmol $m^{-2} d^{-1}$	
	2146		217	1.40 0.00	0.05 (0.01	0.11	
1	3146	> 60	217	1.48 ± 0.08	0.05 / 0.21	-0.11	
2	2900	> 60	213	1.32 ± 0.05			
3	2601	> 60	194	0.81 ± 0.06			
4	2106	> 60	89	1.32 ± 0.05	0.17 / 0.17	-0.15	
6	89	36	0	2.61 ± 0.01	0.03 / 0.05	-0.08	
23	56	13	0	$5.00 \pm 0.09 \ ; \ 5.3 \pm 0.2$	0.56	-0.12; -5.1 ± 0.4	13
24	46	10	0	7.95 ± 0.14		-0.22	10
27	44	16	0	3.75 ± 0.08	0.37 / 0.20	-0.27	12
30	69	16	0	2.61 ± 0.11	0.06 / 0.03	-0.12	15
31	3056	> 60	194	1.78 ± 0.07			
35	541	> 60	30	2.43 ± 0.32			
37	205	44	0	2.51 ± 0.10			
Average Laptev S	Sea shelf			4.20	0.19	0.2;5.1	12
40	45	12	0	4.62 ± 0.08	0.33 / 0.24	-0.19	16
43	42	13	0	4.7 ± 0.10			
45	40	10	0	4.02 ± 0.10	0.23 / 0.19	-0.37	13
48	49	5	0	9.14 ± 0.22	0.68 / 0.53	-0.71	10
50	44	9	0	$5.65 \pm 0.43; 5.2 \pm 0.1$	1.32 / 1.40	-1.01 ; -5.2 ± 0.2	12
53	47	10	0	$4.53 \pm 0.08 \ ; 4.7 \pm 0.1$	0.10 / 0.17	-0.20	14
58	54	3	0	11.49 ± 0.52	1.01	-1.27	24
63	67	4	0	0.72 ± 0.15 ; 10.8 ± 0.12	1.41	-1.35 ; -10.8 ± 0.6	12
Average East Sib	erian Sea shelf			7.2		0.7;8.0	14

Table 2. O_2 uptake, integrated ³⁵S-sulfate reduction rates, DIC flux, and porewater DIC/NH₄⁺ ratios

Average East Siberian Sea shelf

Table 3. Anaer	obic rates of	carbon mine	ralization by n	nanganese, in	ron, and sulfate re	duction			
	Net Fe ²⁺	Net Mn ²⁺	C-equivalent	³⁵ S-Sulfate	C-equivalents	Oxygen	% Fe + Mn	Percentage	Percentage
	production	production	Fe + Mn	re duction	total anaerobic	uptake	reduction of	anae robic C	Fe and Mn
	1		reduction		mineralization		total	mine ralizatio	mineralizatio
							anae robic	n of total	n of total
			Iomm	$\mathbf{m}^{-1} \mathbf{d}^{-1}$				%	
Station 23	0.05	0.03	0.03	0.56	1.1	5.0	2.3	22.9	0.5
Station 30	0.02	0.04	0.03	0.05	0.1	2.6	21.9	4.4	1.0
Station 45	0.14	0.12	0.09	0.21	0.5	4.0	18.3	12.8	2.3
Station 53	0.15	0.09	0.08	0.14	0.4	4.5	23.7	7.8	1.8
Station 63	-	0.50	0.25	1.41	3.1	10.7	8.1	26.0	2.3

Station	Average δ ¹³ C _{DIC} remineralized	Marine end member	Terrestrial end member	³⁵ S-SRR-based terrestrial degradation rate	DIC-based terrestrial degradation rate
	‰vs. VPDB	Mass	s fraction	mmol m ⁻² d ⁻¹	mmol $m^{-2} d^{-1}$
1	-35.8	0.0	1.0	0.13	0.11
4	-24.7	0.73	0.27	0.05	0.04
6	-25.1	0.65	0.35	0.01	0.03
23	-24.5	0.78	0.22	0.12	0.03
24	-24.7	0.73	0.27		0.06
27	-25.4	0.58	0.42	0.12	0.11
30	-28.5	0.00	1.00	0.05	0.13
Average Laptev Sea shelf	-25.6	0.53	0.47	0.08	0.07
40	-21.4	0.72	0.28	0.08	0.05
45	-22.2	0.63	0.37	0.08	0.14
48	-23.0	0.54	0.46	0.28	0.32
50	-24.0	0.43	0.57	0.77	0.57
53	-18.8	1.00	0.00	0.00	0.00
58	-22.6	0.59	0.41	0.42	0.53
63	-20.3	0.84	0.16	0.25	0.22
Average East Siberian Sea shel	f -21.8	0.68	0.32	0.27	0.26

Table 4. Calculated carbon isotope composition of remineralized DIC and mass fractions of the marine and terrestrial endmembers and corresponding terrestrial carbon degradation rates based on ³⁵S-SRR and DIC flux

Table 5. Regional estimate	es of sediment	carbon mineral	ization in the o	outer Laptev and	l East Siberian s	shelf sea	
			Dissolved O ₂ uptake	Upward DIC flux (anaerobic)	Terrestrial OC- derived DIC flux (anaerobic)	Marine OC- derived DIC flux (anaerobic)	Depth-integrated ³⁵ S-SRR (C equivalent)
Outer Laptev Sea	Average	mmol m ⁻² d ⁻¹	4.2	0.16	0.07	0.09	0.09
Outer East Siberian Sea	Average	mmol m ⁻² d ⁻¹	7.2	0.73	0.26	0.47	0.34
Outer Laptev Sea	280,000 km ²	Tg C y ⁻¹	5.2	0.20	0.09	0.11	0.11
Outer East Siberian Sea	$340,000~{\rm km}^2$	Tg C y ⁻¹	10.8	1.09	0.39	0.70	0.50
Total outer shelf area	$620,000~{\rm km}^2$	Tg C y ⁻¹	15.9	1.28	0.48	0.81	0.62
			³⁵ S-SRR- based terrestrial C degradation	³⁵ S-SRR- based marine C degradation	Total TEAP- based anaerobic OC degradation rate	Total TEAP- based anaerobic terrestrial OC degradation rate	Total TEAP- based anaerobic marine OC degradation rate
Outer Laptev Sea	Average	mmol m ⁻² d ⁻¹	0.04	0.05	0.15	0.05	0.10
Outer East Siberian Sea	Average	$mmol \ m^{-2} \ d^{-1}$	0.13	0.21	0.42	0.16	0.26
Outer Laptev Sea	$280,000~{\rm km}^2$	Tg C y ⁻¹	0.05	0.07	0.18	0.06	0.12
Outer East Siberian Sea	$340,000~{\rm km}^2$	Tg C y ⁻¹	0.20	0.31	0.62	0.23	0.39
Total outer shelf area	$620,000~{\rm km}^2$	Tg C y ⁻¹	0.25	0.37	0.80	0.29	0.51



1102 Fig. 1. Map of the Eastern Siberian Sea and slope and station locations.



Fig. 2. Depth profiles of dissolved O₂, Fe²⁺, and Mn²⁺ at Stations 1, 23, 30, 45, 53, and 63. For
 microelectrode profiles, 4 replicates are shown for each station. Depth resolution of measurement

- 1108 for O₂ was 100 μm.
- 1109

1105

1110



1111

1112 Fig. 3. Depth of profiles of ³⁵S-sulfate reduction rates and porewater concentration of dissolved

sulfate for Stations 1, 23, 30, 45, 53, and 63. A replicate incubation was conducted for each depth

1114 except for Station 63.



1116 Fig. 4. Depth profiles of porewater dissovled inorganic carbon (DIC), δ^{13} C DIC and porewater NH₄⁺ at

1117 stations 1, 23, 30, 45, 53, and 63.



1118

Fig. 5. Comparison of reaction rates of oxygen, manganese, iron, and sulfate reduction at Station 23.
Note the different depth scale for the O₂ consumption rate. The dashed line marks the oxygen
penetration depth.





Fig. 6. A: Crossplot of dissolved NH₄⁺ and porewater DIC* after correction for bottom water DIC
concentrations. The slopes of the regression lines for the individual stations are shown in Table 2. B:

- 1127 Crossplot of the fraction of remineralized DIC calculated from a 2-endmember mixing model versus
- δ^{13} C DIC. The slope and y-intercept of the regression for each station are shown in Table 3.





160°E

140°E

Fig. 7 A, B. Map of field area and sampling stations showing oxygen uptake rates in panel A anddepth-integrated sulfate reduction rates in panel B.

120°E



Fig. 8A. Water depth variation of sediment oxygen uptake. 8B: Water depth variation of integrated
 ³⁵S-sulfate reduction rates (0-30 cm sediment depth). For reference average rates of abyssal plain,
 continental rise, slope, and shelf sediments, deposition and non-depositional from Canfield et al.
 (2005).





Fig. 9. Crossplot of diffusive oxygen uptake and integrated sulfate reduction rates. The black line is the linear regression and yielded a y-intercept of 2.1 mmol m⁻² d⁻¹ and a slope of 5.55. Blue and red lines show the 95% and 99% confidence interval.