



Contrasting regimes for organic matter degradation in the East Siberian Sea and the Laptev Sea assessed through microbial incubations and molecular markers

E.S. Karlsson^{a,c,*}, V. Brüchert^{b,c}, T. Tesi^{a,c}, A. Charkin^{e,f}, O. Dudarev^{e,f}, I. Semiletov^{d,e,f}, Ö. Gustafsson^{a,c,*}

^a Department of Environmental Science and Analytical Chemistry (ACES), Stockholm University, SE-106 91 Stockholm, Sweden

^b Department of Geological Sciences (IGV), Stockholm University, SE-106 91 Stockholm, Sweden

^c Bolin Centre for Climate Research, Stockholm University, SE-106 91 Stockholm, Sweden

^d International Arctic Research Center, University Alaska Fairbanks, Fairbanks, AK 99775, USA

^e Pacific Oceanological Institute, Russian Academy of Sciences Far Eastern Branch, Vladivostok 690041, Russia

^f The National Research Tomsk Polytechnic University, Tomsk, Russia

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ABSTRACT

Compositional studies of organic matter on the East Siberian Arctic Shelf (ESAS) suggest that different terrestrial carbon pools have different propensities for transport and/or degradation. The current study combined laboratory-based microbial degradation experiments with earlier published degradation-diagnostic composition of several classes of terrestrial biomarkers on the same sediments to investigate differences and driving forces of terrestrial organic matter (TerrOM) degradation in two biogeochemically-contrasting regimes of the ESAS. The incubation-based anaerobic degradation rates were consistently higher (by average factor of 6) in the East Siberian Sea Kolyma Paleoriver Channel (ESS-KPC) ($15 \mu\text{mol CO}_2 \text{ g OC}^{-1} \text{ day}^{-1}$) compared to the Laptev Sea Buor-Khaya Bay (LS-BKB) ($2.4 \mu\text{mol CO}_2 \text{ g OC}^{-1} \text{ day}^{-1}$). The reported molecular markers show similarities between the terrestrial carbon pools in the two systems, but impose contrasting degradation regimes in combination with the incubation results. For the LS-BKB, there was a strong relationship between the degradation rates and the three lignin phenol-based degradation proxies ($r^2 = 0.93\text{--}0.96$, $p < 0.01$, linear regression) and two wax lipid-based degradation proxies ($r^2 = 0.71$ and 0.66 , $p < 0.05$, linear regression). In contrast, for the ESS-KPC system, there was no relationship between incubation-based degradation rates and molecular marker-based degradation status of TerrOM. A principal component analysis indicated that short-chain fatty acids and dicarboxylic acids from CuO oxidation are mainly of terrestrial origin in the LS-BKB, but mainly of marine origin in the ESS-KPC. Hence, the microbial degradation in the western (LS-BKB) system appears to be fueled by TerrOM whereas the eastern (ESS-KPC) system degradation appears to be driven by MarOM. By combining molecular fingerprinting of TerrOM degradation state with laboratory-based degradation studies on the same ESAS sediments, a picture evolves of two distinctly different modes of TerrOM degradation in different parts of the ESAS system.

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1. Introduction

There are still major uncertainties regarding the fate of terrestrial organic matter (TerrOM) in the coastal ocean (e.g., Hedges et al., 1997). This is particularly relevant for the Arctic Ocean since it contains extensive shelves that are heavily influenced by terrestrial input from the many rivers and from coastal erosion. Terrestrial pan-Arctic soil holds over half of the global soil carbon in its now thawing permafrost (Tarnocai et al., 2009) and lateral transport of some of this organic matter (OM) to the coastal Arctic Ocean is substantial (e.g. Vonk and Gustafsson, 2013; Gordeev et al., 1996; Raymond et al., 2007). The Eurasian Arctic is anticipated to undergo a more drastic change in OM transport and turnover due to a combination of climate warming,

Abbreviations: OM, organic matter; OC, organic carbon; TerrOM, terrestrial organic matter; MarOM, marine organic matter; ESAS, East Siberian Arctic Shelf; ESS, East Siberian Sea; LS, Laptev Sea; ESS-KPC, refers to the study region of the Kolyma Paleoriver Canyon central-eastern East Siberian Sea; LS-BKB, refers to the study region of Buor Khaya Bay in south-east Laptev Sea; Lignins, lignin phenols from CuO oxidation; Cutins, cutin acids from CuO oxidation; BA, benzoic acids from CuO oxidation; PB, *p*-hydroxybenzenes from CuO oxidation; FA, fatty acids from CuO oxidation; DA, dicarboxylic acids from CuO oxidation

* Corresponding authors at: Department of Environmental Science and Analytical chemistry (ACES) Stockholm University, SE-106 91 Stockholm, Sweden.

E-mail addresses: emma.karlsson@aces.su.se (E.S. Karlsson), volker.bruchert@geo.su.se (V. Brüchert), tommasso.tesi@aces.su.se (T. Tesi), charkin@poi.dvo.ru (A. Charkin), dudarev@poi.dvo.ru (O. Dudarev), igorsm@iarc.uaf.edu (I. Semiletov), orjan.gustafsson@aces.su.se (Ö. Gustafsson).

changing hydrology, and collapsing permafrost (e.g. Holmes et al., 2013; Luchin et al., 2002; Peterson et al., 2002; Semiletov et al., 2013).

However, the terrestrial carbon pools that may be remobilized across the Eurasian-Arctic are heterogeneous and differences in composition and physical associations may affect their propensity for transport and/or degradation (e.g., Feng et al., 2013; van Dongen et al., 2008a; Vonk et al., 2010b). Indeed, studies in both continental shelf water and sediments of the ESAS have documented different pools of TerrOM that behave differently (e.g. Alling et al., 2010; Karlsson et al., 2011).

The degradability of organic matter has several controls, including intrinsic chemical lability/reactivity related to its molecular structure, physical factors such as sorptive matrix protection, and biological/enzymatic limitations. The different carbon reservoirs around the Arctic include recent vegetation in the active layer, mineral soils, deeper soil compartments accessed through hydraulic conduits and erosion features from, e.g., Ice Complex Deposits (a.k.a. Yedoma) that are ubiquitous in northeast Siberia. In fact, several studies have shown that TerrOM exported to the coastal ocean across the Eurasian Arctic has different sources, as traced by, e.g., ^{14}C -isotopic fingerprint of bulk carbon pools (e.g., Benner et al., 2004; Neff et al., 2006; Vonk et al., 2012) and ^{14}C age of terrestrial molecular markers (e.g., Drenzek et al., 2007; Feng et al., 2013; Gustafsson et al., 2011; Vonk et al., 2010b) as well as by molecular marker composition (e.g., Amon et al., 2012; van Dongen et al., 2008a; Goni et al., 2005). These studies suggest that surface soil permafrost OM degrades to a larger extent in the water column, while terrestrial matter from old Pleistocene coastal deposits may rather accumulate and dominate the sea floor TerrOM composition (Opsahl and Benner, 1997; Opsahl et al., 1999; Sanchez-Garcia et al., 2011; Vonk et al., 2012).

The current study seeks to address the degradability of different carbon pools with two independent approaches, by combining new laboratory-based incubations to assess the intrinsic propensity to microbial degradation with published molecular biomarker signals of the degradation status of the TerrOM on the exactly same sediments. The combined geochemical-microbiological approach was applied to two different regions of the ESAS. Located 1200 km away from each other, the south-east Laptev Sea (LS) and the central-eastern East Siberian Sea (ESS) are potentially influenced by different terrestrial and marine carbon sources (e.g., Semiletov et al., 2005, 2012), affecting the TerrOM degradation process in these marine systems.

2. Study area

The ESAS is the largest yet shallowest continental shelf in the world. Three-quarters are less than 50 m deep, and half of the area is less than 30 m deep (Jakobsson et al., 2008; Vetrov and Romankevich, 2004). The ESAS is the recipient of the inflow from major rivers (Khatanga, Lena, Yana, Indigirka and Kolyma) and of massive erosion of the Pleistocene coastal ice complex (a.k.a. Yedoma; e.g. Günther et al., 2013; Semiletov, 1999; Stein and Macdonald, 2004; Vonk et al., 2012). The eastern areas experience colder weather and contain larger areas of continuous permafrost than the west (Brown et al., 1997; Heginbottom, 1984, 2002). The central-west is warmer, has a longer summer season and more precipitation, but is also part of the high Arctic with extreme continental climate. The study area experiences relatively warm summers and cold winters, but with an annual average temperature well below freezing (Polyakov et al., 2003; Serreze et al., 2000), for example between $-8\text{ }^{\circ}\text{C}$ and $-16\text{ }^{\circ}\text{C}$ on the 135° meridian (from 60° N to the Arctic Ocean Coast) (Romanovsky et al., 2007).

The terrestrial carbon input to the ESAS is high and varies seasonally as a function of river discharge coastal erosion and sea ice cover (Charkin et al., 2011; Guo et al., 2004; Holmes et al., 2012; Semiletov et al., 2013). Phytoplankton deposition to the sea floor varies with season, since nutrient concentrations depend on the amount of river discharge and on Pacific inflow from the east (Belicka et al., 2002; Kosheleva and Yashin, 1999; Semiletov et al., 2005). The riverine

nutrient input to the Arctic Ocean is generally low (Cauwet and Sidorov, 1996; Dittmar and Kattner, 2003a; Lobbes et al., 2000). Coastal erosion and subsequent oxidation of eroded OM may serve as the predominant source of nutrients in the shallow near-shore Laptev Sea (Semiletov et al., 2013). Taken together, these conditions create a permanently cold sedimentary environment with high spatiotemporal variability of OM and nutrient input (Charkin et al., 2011; Cooper et al., 2008; Yunker et al., 1995).

These characteristics also generate potentially contrasting degradation regimes of the different parts in the ESAS sea floor, here represented by the two study sites located 1200 km apart, Buor-Khaya Bay (LS-BKB) (Fig. 1A, B) and the cross-shelf transect in the Kolyma Paleoriver Canyon (ESS-KPC) (Fig. 1A, C). Buor-Khaya Bay is situated right at the mouth of the Lena River, and is the recipient of the main branches of the Lena delta (Fig. 1A, B). In addition to the large river input, the LS-BKB receives a large portion of terrestrial matter from erosion of the coastal ice-complexes, including a coastal erosion hot spot, Muostakh Island. The study area ESS-KPC starts at the Kolyma river mouth and covers hundreds of kilometers offshore across the shelf (Fig. 1A, C). The Kolyma River is smaller than the Lena River and discharges in the central-eastern ESS, an area that is also affected by Pacific inflow from the east.

3. Materials and methods

3.1. Sample selection

The sediment samples selected for this coupled biomarker-incubation study were collected during the International Siberian Shelf Study 2008 (ISSS-08), a campaign onboard R/V Yakob Smirnitkiy over extensive regions of the ESAS (Semiletov and Gustafsson, 2009). For the current study, ISSS-08 samples from two contrasting regimes were selected based on published biomarker patterns of degradation. First, six surface sediments from a 13 to 46 m depth transect were selected from across the central-eastern ESS in the Kolyma Paleoriver channel (Fig. 1), reported on earlier for the wax lipid, lignin and cutin type biomarkers (Vonc et al., 2010a). Secondly, to provide a contrasting regime, five surface sediments (5–21 m depth) were investigated from a sub-campaign of the ISSS-08 to the Buor-Khaya Bay in south-eastern LS, where biomarker distribution was also detailed earlier (e.g., Charkin et al., 2011; Karlsson et al., 2011; Tesi et al., 2014) (Fig. 1). Both of these sets were selected to provide clear gradients in biomarker-indicated organic matter sources and degradation status (Vonc et al., 2010a; Karlsson et al., 2011; Tesi et al., 2014).

3.2. Microcosm setup – incubation

Most continental shelf sediments have an oxygen penetration depth of only a few mm (Glud, 2008). A large amount of the microbial degradation in continental shelf sediments such as the ESAS therefore takes place under anaerobic conditions. Sediment incubations in this study were consequently conducted under anaerobic conditions and these apparent rates were compared to the ambient biomarker patterns. For comparison, a subset of samples was also incubated aerobically. However, it is likely that oxygen was diffusion-limited even in the aerobic setup so that anoxic interiors in the sediment samples were retained.

Around 5 g of frozen (wet) sediment was weighed into 25 mL serum bottles, whereupon bottles were sealed with rubber stoppers and aluminum crimp caps. Bottles had been acid-rinsed, washed, and baked at $450\text{ }^{\circ}\text{C}$. Rubber stoppers and crimp caps were autoclaved. 5 mL of Artificial Sea Water (ASW) [Recipe ASW: (concentrations, given in g L^{-1}) NaCl 20, $\text{MgCl}_2 \times 6\text{H}_2\text{O}$ 3, $\text{CaCl}_2 \times 2\text{H}_2\text{O}$ 0.15, Na_2SO_4 0.15, NH_4Cl 0.25, KH_2PO_4 0.2, KCl 0.5] was added to the frozen sediments. The DOC that may have been added to the system from the DOC in MilliQ would be low. Concentrations of MilliQ from the used system are consistently $<10\text{ }\mu\text{g/L}$ and often $<1\text{ }\mu\text{g/L}$. We added 5 mL of ASW to each sample bottle,

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