Journal Code				Article ID					Dispatch: 26.03.13	CE: Joyce P. Ogabang
G	R	L		5	0	3	4	8	No. of Pages: 5	ME:

GEOPHYSICAL RESEARCH LETTERS, VOL. 40, 1–5, doi:10.1002/grl.50348, 2013

High biolability of ancient permafrost carbon upon thaw

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Received 11 January 2013; revised 7 March 2013; accepted 8 March 2013.

[1] Ongoing climate warming in the Arctic will thaw permafrost and remobilize substantial terrestrial organic carbon (OC) pools. Around a quarter of northern permafrost OC resides in Siberian Yedoma deposits, the oldest form of permafrost carbon. However, our understanding of the degradation and fate of this ancient OC in coastal and fluvial environments still remains rudimentary. Here, we show that ancient dissolved OC (DOC, >21,000 ^{14}C years), the oldest DOC ever reported, is mobilized in stream waters draining Yedoma outcrops. Furthermore, this DOC is highly biolabile: $34 \pm 0.8\%$ was lost during a 14 day incubation under dark, oxygenated conditions at ambient river temperatures. Mixtures of Yedoma stream DOC with mainstem river and ocean waters, mimicking in situ mixing processes, also showed high DOC losses (14 days; $17 \pm 0.8\%$ to $33 \pm 1.0\%$). This suggests that this exceptionally old DOC is among the most biolabile DOC in any previously reported contemporary river or stream in the Arctic.
Citation: Vonk J. E., P. J. Mann, S. Davydov, A. Davydova, R. G. M. Spencer, J. Schade, W. V. Sobczak, N. Zimov, S. Zimov, E. Bulygina, T. I. Eglinton and R. M. Holmes (2013), High biolability of ancient permafrost carbon upon thaw, *Geophys. Res. Lett.*, *40*, doi:10.1002/grl.50348.

1. Introduction

[2] The northern soil carbon (C) pool contains approximately 1672 Pg C [*Tarnocai et al.*, 2009] of which ca. 25% (>400 Pg C) is stored in frozen Yedoma [*Zimov et al.*, 2006] deposits in the Siberian-Arctic (Figure 1). This is approximately equal to the amount of C stored in total global forest biomass [*Pan et al.*, 2011]. Yedoma deposits formed during the late Pleistocene [*Zimov et al.*, 2006; *Schirrmeister et al.*, 2011] in unglaciated Siberia and cover ca. 1 million km^2 (Figure 1a). Due to a lack of processing and survival of bacteria [*Dutta et al.*, 2006; *Rivkina et al.*, 1998] during formation, organic carbon (OC) held in Yedoma deposits has been hypothesized to be highly biolabile upon thaw. Formation of Yedoma is initiated by accumulation of sediments [*Zimov et al.*, 2006], and is therefore not as much exposed to repetitive freeze-thaw cycles, that process and degrade OC, in comparison to other soil ecosystems. As climate warms, permafrost soils (including Yedoma) will thaw and decompose leading to the production of greenhouse gases, in turn accelerating climate warming [*Dutta et al.*, 2006; *Schaefer et al.*, 2011]. This is referred to as the "permafrost carbon feedback" (PCF). Initial estimates suggest that PCF can be substantial [*Zimov et al.*, 2006; *Schuur et al.*, 2008; *Schaefer et al.*, 2011], but remarkably, this process is not included in any Intergovernmental Panel on Climate Change (IPCC) [*IPCC*, 2007] scenario of future climate change. Understanding how frozen OC pools are processed upon thaw is therefore crucial for understanding future climate change and C dynamics in the Arctic and globally.

[3] Four physical mechanisms release OC from permafrost to the atmosphere [*Schuur et al.*, 2008]: active layer deepening, talik formation, thermokarst development, and erosion. Coastal and riverbank erosion of Yedoma deposits can occur over vast distances (e.g., along the > 5000 km East Siberian Arctic coastline) [*Vonk et al.*, 2012] and may release OC from entire soil depth profiles (up to ca. 40 m) directly to aquatic environments. A deepening of the active layer—the seasonally thawed surface soil, may also lead to an increased supply of permafrost derived OC to inland waters over regional scales. After thawing, the crucial current unknowns are how much, and how rapidly does OC held within these deposits enter the contemporary C cycle. Inland and coastal waters are increasingly recognized as important processors of terrestrial organic carbon (OC), generating a substantial flux of CO_2 to the atmosphere [*Aufdenkampe et al.*, 2011; *Battin et al.*, 2008; *Bianchi*, 2011]. Carbon processing during coastal release in the Siberian Arctic has been shown to be substantial [*Alling et al.*, 2010; *Sánchez-García et al.*, 2011]. This study investigates the "hydrological biolability", i.e., the potential biodegradability within the residence time of the aquatic system, of OC from recently thawed Yedoma deposits introduced to aquatic ecosystems.

[4] The "Duvannyi Yar" exposure on the banks of Kolyma River in Northeast Siberia (Figure 1) is a classical and relatively well-studied Yedoma site [e.g., *Vasil'chuk and Vasil'chuk*, 1997; *Dutta et al.*, 2006]. Radiocarbon ages of the ca. 40 m high deposits range between 13,000 and 45,000 years [*Vasil'chuk and Vasil'chuk*, 1997]. Permafrost thaw and/or riverbank erosion result in mean retreat rates of 3–5 m/yr. Water from melting ice wedges feed streams that carry recently thawed Yedoma off the cliff into the Kolyma River (Figure 2). These sediment-laden, first-order streams F2

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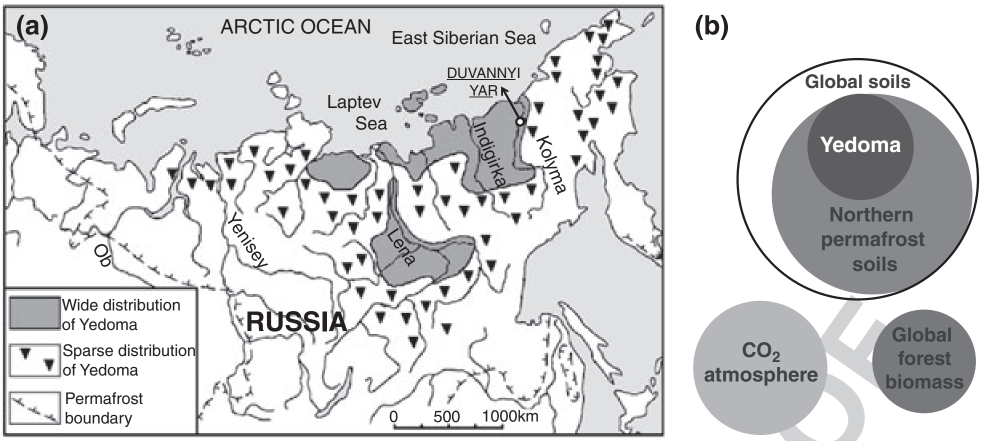

Figure 1. (a) Yedoma extent in the Siberian Arctic [*Romanovskii*, 1993], overlain with major rivers and our study site Duvannyi Yar. (b) Relative carbon pool sizes of Yedoma (450 Pg) [*Zimov et al*., 2006; *Tarnocai et al*., 2009] in comparison with other major global carbon pools (global soils 2400 Pg, northern permafrost soils 1672 Pg, global forest biomass 450 Pg and atmospheric CO_2 750 Pg) [*IPCC*, 2007; *Tarnocai et al*., 2009; *Pan et al*., 2011].

represent an integrated signal of recently thawed Yedoma. These first-order streams are formed early in the summer when ice wedges start to melt, and their course and magnitude depends on local riverbank collapse, thaw rates and relief. We used the stream dissolved OC (DOC) for incubations and mixing experiments with Kolyma River and East Siberian Sea-water (see section 2), with the goal to estimate biolability during fluvial and coastal processing of ancient permafrost OC. Q3

2. Methods

[5] In July 2010, we sampled six thaw streams along the Duvannyi Yar exposure (on a line from 68.631°N–159.156°E to 68.631°N–159.143°E, Figure 2) on the Kolyma River. The streams have all freshly formed from thawing ice-rich Yedoma permafrost, and meander down the cliff in a course that is dependent on local bank collapse, thaw rates and relief. Estimated flow rates on our sampling day were ca. 2.5–10 L/s, stream temperatures just above 0 °C and transport time from thaw to entry into Kolyma River < 1 h. The water was filtered through precombusted 0.7 µm glass fiber filters (Whatman) and analyzed at the Northeast Science Station in Cherskii (Russia), for DOC on a Shimadzu TOC-V analyzer using established protocols [*Mann et al*., 2012]. Frozen water samples were exported, acidified (to remove inorganic carbon) and analyzed for ^{14}C-DOC at the U.S. National Ocean Sciences Accelerator Mass Spectrometry facility of the Woods Hole Oceanographic Institution (USA). Frozen particulate

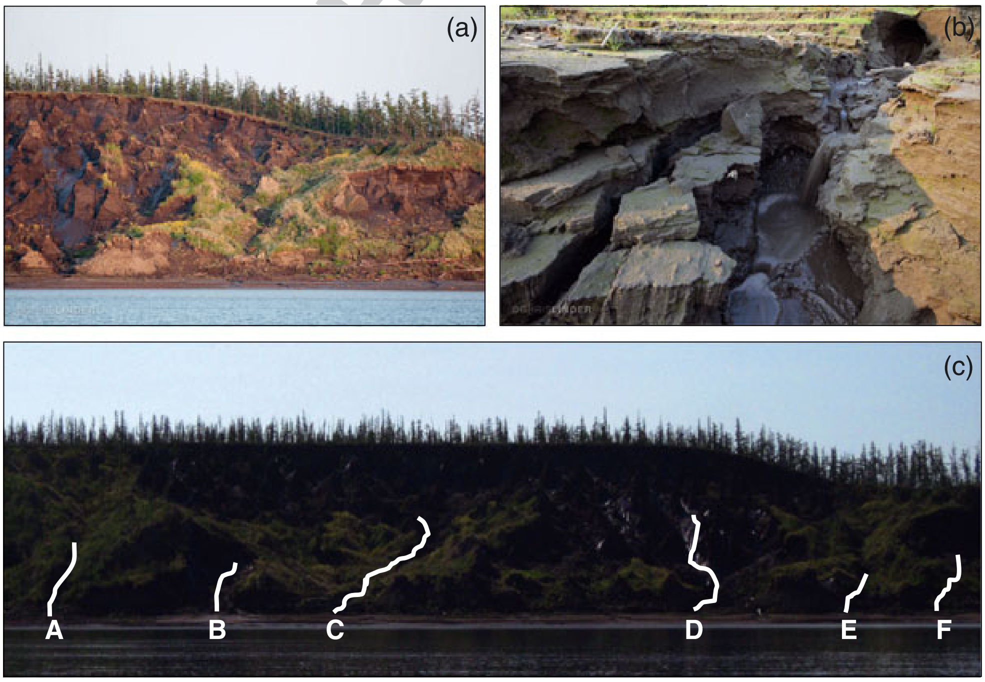

Figure 2. (a) Duvannyi Yar exposure on the Kolyma river bank containing (b) freshly thawed Yedoma organic matter is exported down the cliffs merging in mud streams that drain into Kolyma River. (c) Location of thaw streams sampled in 2010. (photos © Chris Linder).

matter samples were acidified (to remove inorganic carbon) and analyzed for percent OC at the Stable Isotope Laboratory at the Department of Geological Sciences at Stockholm University (Sweden) and for ^{14}C-POC at the AMS facility of the Laboratory of Ion Beam Physics of the Swiss Federal Institute of Technology (Zürich, Switzerland). In July 2011, we returned to Duvannyi Yar and collected a composite sample of four thaw streams (approximate locations of the streams B, C, D, F in 2010, Figure 2c). The samples were kept cool during transport back to the Northeast Science station and filtered (pre-combusted GF/F 0.7 µm, Whatman). The same day, we prepared 0.5%, 1.0% and 10% dilutions of Duvannyi Yar water with Kolyma River water and East Siberian Sea water (also filtered through pre-combusted 0.7 µm GF/F, no inoculum added [*Holmes et al.*, 2008; *Mann et al.*, 2012]). The dilutions were distributed into 40 mL pre-combusted glass vials (in triplicate). All vials, along with the control samples (filtered Kolyma River, East Siberian Sea and Duvannyi Yar water) were positioned on a shaker table and left to incubate at room temperature (ca. 20 °C). This is circa 4° warmer than the average July surface water temperature of Kolyma River (16.3 °C, SD = 0.66, n = 27; www.thepolarisproject.org/data/, http://arcticgreatrivers.org/data.html). The incubations were kept in dark, oxygenated conditions (capped loosely) for 14 and 28 days, to determine the bioavailable DOC (BDOC). At T = 14 and T = 28 days, the bottles were acidified to pH 2 and OC loss was measured directly from the vials. No flocculation was observed during the incubations or prior to analyses.

3. Results and Discussion

[6] In July 2010, Yedoma thaw streams of Duvannyi Yar (n = 6) carried exceptional amounts of suspended sediments **T1** (552 ± 69 g/L; mean ± SD; Table 1), with high dissolved OC (DOC) and particulate OC (POC) concentrations (196 ± 71 mg/L and 7970 ± 100 mg/L, respectively; Table 1). The following summer, a composite stream sample taken in July 2011 from four thaw streams (see section 2) contained 155 mg/L DOC. Studies on Arctic river DOC generally report modern ages [*Neff et al.*, 2006; *Guo and Macdonald*, 2006; *Raymond et al.*, 2007]. Duvannyi Yar stream waters, by contrast, contained DOC far older than ever previously reported in natural waters anywhere (2010: −946 ± 25‰ or 24,100 ± 3900 years, 2011: −933‰ or 21,700 years; Table 1). Ages of POC were comparable (2010: −953 ± 36‰ or 27,500 ± 8200 years; Table 1) to DOC ages. Stable C isotopic values of DOC and POC pools were also similar (DOC −25.7 ± 0.28‰, POC −25.1 ± 0.31‰; Table 1), further confirming a common source, and resembling Yedoma soil δ^{13}C values (−26.3‰, SD = 0.67‰, n = 374) [*Schirrmeister et al.*, 2011; *Vonk et al.*, 2012]. Finally, deuterium and oxygen isotope analyses on the 2011 composite stream water (−246‰ and −32.4‰, respectively) were similar to a local ice wedge sample (−256‰ and −33.4‰, respectively) and to previously reported Yedoma ice wedge values [*Meyer et al.*, 2002], indicating that the thaw streams originated predominantly in Pleistocene deposits, and not from recent precipitation (rain δ^{18}O −16.3‰, δ^{2}H ca. −101‰; snow δ^{18}O −26.2‰, δ^{2}H ca. −199‰) or river water (δ^{18}O ca. −22‰; δ^{2}H −175‰) [*Meyer et al.*, 2002; *Welp et al.*, 2005].

[7] Our biolability study focused on DOC instead of POC, because it is the most important intermediary in the global C cycle [*Battin et al.*, 2008]. Only low-molecular weight

Table 1. Bulk Geochemical Parameters of Duvannyi Yar Thaw Streams in 2010 and 2011

Sampling date	#	δ^{18}O (‰)	δ^{2}D (‰)	DOC (mg/L)	DOC δ^{13}C (‰)	DOC Δ^{14}C (‰)	DOC ^{14}C age (years)	POC (mg/L)	POC δ^{13}C (‰)	POC Δ^{14}C (‰)	POC ^{14}C age (years)	SPM g/L
2010												
27 Jul 2010	A	—	—	154	−25.6	−974 ± 1.2	29400 ± 380	8580 ± 530	−24.9 ± 0.04	−988 ± 0.73	35800 ± 500	608 ± 53
27 Jul 2010	B	—	—	336	−26.1	−951 ± 1.3	24200 ± 230	8480 ± 590	−25.7 ± 0.03	−992 ± 0.77	38300 ± 730	609 ± 16
27 Jul 2010	C	—	—	194	−25.9	−911 ± 0.76	19350 ± 70	9240 ± 440	−25.2 ± 0.05	−920 ± 1.0	20200 ± 100	628 ± 16
27 Jul 2010	D	—	—	176	−26.0	−970 ± 0.49	28000 ± 130	6700 ± 820	−25.1 ± 0.1	−974 ± 0.79	29210 ± 240	494 ± 13
27 Jul 2010	E	—	—	161	−25.3	−926 ± 1.1	20900 ± 130	6920 ± 300	−24.8 ± 0.08	−939 ± 0.78	22380 ± 100	491 ± 6.7
27 Jul 2010	F	—	—	154	−25.7	−943 ± 1.2	22900 ± 170	7880 ± 2000	−25.1	−907 ± 1.2	19070 ± 100	484 ± 9.5
2011												
22 Jul 2011	Comp	−32.4 ± 0.04	−246 ± 0.65	155 ± 2.4	−25.47	−933 ± 0.62	21700 ± 85	—	−25.2 ± 0.21	—	—	—

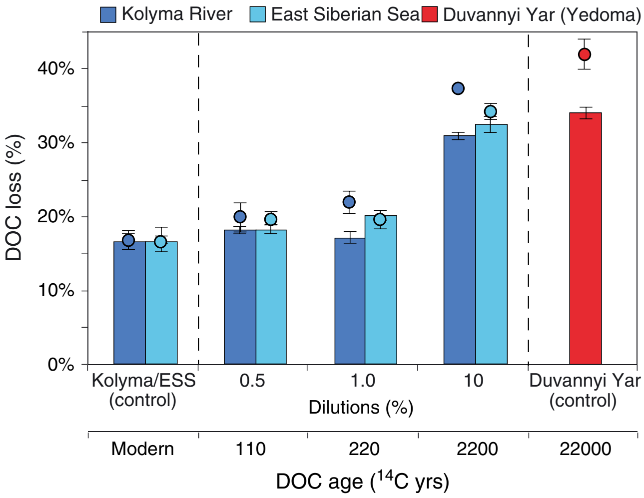

Figure 3. Dissolved organic carbon loss (%) after 14 days (bars) and 28 days (points) dark incubations at 20 °C for Kolyma River (light blue), East Siberian Sea (dark blue), Yedoma streams (red), and three dilutions of Yedoma water with Kolyma River or East Siberian Sea water. The ^{14}C age of DOC (years) (*x*-axis) represents the proportion of Duvannyi Yar (Yedoma) stream water added to Kolyma River or East Siberian Sea water. Standard deviations represent errors from triplicate experiments.

dissolved compounds can be transported through microbial cell membranes, and are thus readily available for microbial metabolism [*Battin et al.*, 2008]. Thaw streams originating in Pleistocene Yedoma deposits lost 34 ± 0.8% of their DOC after 14 days and 41 ± 3% after 28 days incubation (see section 2 and Figure 3). With an initial DOC concentration of 155 mg/L, these losses convert to a remarkable decrease in DOC concentration of 52 mg/L in 14 days, or 63 mg/L in 28 days. Additionally, samples of Kolyma River and East Siberian Sea water (salinity 26) that were spiked with Yedoma-DOC showed an increase in DOC loss with increasing Yedoma additions (Figure 3), from ca. 18% loss for a 0.5% Yedoma addition to ca. 32% loss for a 10% Yedoma addition after 14 days. Higher percent OC losses were observed in both fresh and saline waters with increasing additions of Yedoma OC (Figure 3) suggesting that Yedoma OC may enhance, or even prime, remineralization of contemporary DOC [*Bianchi*, 2011]. We propose that the high biolability of this > 21,000 year old DOC can be explained by the following: (i) a lack of pre-processing of DOC prior to thaw [e.g., *Dutta et al.*, 2006], (ii) the presence of more low-molecular-weight compounds or fewer aromatics [*Waldrop et al.*, 2010], (iii) nutrient availability, and (iv) an abrupt spike in the activity of Yedoma's intrinsic bacteria, having survived for thousands of years [*Rivkina et al.*, 1998], but now exposed to temperatures above zero and availability of (liquid) water. Previous reports on Arctic stream or river biolability report percent DOC losses up to 42% (3 months at 20 °C; Alaska) [*Holmes et al.*, 2008], up to 35% (40 days at 25 °C; Alaska) [*Balcarczyk et al.*, 2009], up to 53% for under-ice waters (28 days, 15 °C; Alaska) [*Wickland et al.*, 2012], up to 28% (97 days, temperature not given; W-Siberia) [*Kawahigashi et al.*, 2004], and up to 20% (28 days, room temperature; Kolyma River) [*Mann et al.*, 2011]. Variable incubation conditions make direct comparisons challenging, yet a 34% loss of ancient (> 21,000 ^{14}C years) DOC after only 14 days is among the highest ever reported for pristine, non-glacier-fed, rivers or streams.

[8] The conventional way to calculate and globally-upscale greenhouse gas release fluxes from thawing permafrost is typically based on direct CO_2 and CH_4 emission measurements from thawing tundra soils [*Dorrepaal et al.*, 2009]. Here, we show that a substantial portion of greenhouse gases may be generated during stream, river and shelf transport. One should note here that we only examined release of Yedoma OC in the dissolved phase; the dynamics of POC, with fluxes that are likely orders of magnitude higher (Table 1), and that may breakdown (also photochemically) to release substantial amounts of DOC, remain undetermined. Our results further indicate that differences in the reactivity of contemporary versus old OC, and associated fluxes need to be accounted for in future studies. The strong degradation of Yedoma-DOC in the stream network could also explain why significant seasonal DOC aging in the Kolyma River main-stem has not been observed [*Neff et al.*, 2006]. Increased contributions of permafrost-derived C may not be easy to detect at the mouth of large Arctic rivers as it is removed rapidly over short incubation times likely comparable to water residence times within the headwaters (~3–7 days from Duvannyi Yar to river mouth, assuming average river velocities of 0.5–1.5 m/s, *Holmes et al.*, [2012]). Furthermore, the flux of terrestrial-derived OC to the Arctic Ocean, estimated from measurement at the river mouth, may misrepresent the actual mobilization and turnover of the DOC in the watershed, as processing of DOC within the catchment, prior to arrival at the river mouth is not included in these estimates. Similar to recent studies that revealed the presence of biolabile ancient OC being liberated from glaciers [*Hood et al.*, 2009], our data also contrasts to the prevailing paradigm of age versus reactivity of OC.

4. Conclusions

[9] We show here that Yedoma DOC, with > 21,000 ^{14}C years the oldest ever reported, is highly biolabile upon release, both in fluvial (i.e., Kolyma River) and in coastal environments (i.e., East Siberian Sea) (Figure 3). Estimates of the exact contribution of Yedoma OC in Arctic C cycling are improving [*Vonk et al.*, 2012], but are still subject to considerable uncertainties, e.g., due to a lack of adequate spatial coverage of Yedoma deposits [*Romanovskii*, 1993], and accurate estimates of delta and riverbank erosion. Ongoing Arctic climate warming is expected to further increase the thermal exposure, thaw and erosion of Yedoma, particularly along the extensive East Siberian Arctic coastline, where Yedoma deposits are ubiquitously present and increasingly exposed to wave fetch and storms due to recent reductions in sea ice [*IPCC*, 2007; *Stroeve et al.*, 2007]. Yedoma OC, hosting ca. 25% of the total belowground soil OC [*Zimov et al.*, 2006; *Tarnocai et al.*, 2009] is very old, yet highly biologically reactive upon mobilization. The high biolability of this material seems likely to amplify the effect of the PCF scenario in the Arctic. Furthermore, examining permafrost degradation through ^{14}C-DOC measurements at river mouths may not be representative for the actual mobilization and turnover of permafrost C, as extensive processing of permafrost derived-C *within* the watershed may be masking the river mouth signal. It is apparent that further studies are

needed to address the magnitude of the flux of thawing Yedoma OC, but also, importantly, to incorporate the reactivity of this material in regional and global C budgets.

[10] **Acknowledgments.** We would like to acknowledge financial support from the US-NSF (Polaris Project #1044610 and #1203885 to R.G.M.S.), Russian Foundation for Basic Research (RFBR) and the Dutch-NSF (J. E. Vonk, Rubicon # 825.10.022). Irka Hajdas and Lukas Wacker (ETH-Zürich, Laboratory of Ion Beam Physics) are kindly thanked for their assistance with ^{14}C analyses on POC.

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