Carbon cycle models suggest that past warming events in the Arctic may have caused large-scale permafrost thaw and carbon remobilization, thus affecting atmospheric CO$_2$ levels. However, observational records are sparse, preventing spatially extensive and time-continuous reconstructions of permafrost carbon release during the late Pleistocene and early Holocene. Using carbon isotopes and biomarkers, we demonstrate that the three most recent warming events recorded in Greenland ice cores—(i) Dansgaard-Oeschger event 3 (~28 ka B.P.), (ii) Bølling-Allerød (14.7 to 12.9 ka B.P.), and (iii) early Holocene (~11.7 ka B.P.)—caused massive remobilization and carbon degradation from permafrost across northeast Siberia. This amplified permafrost carbon release by one order of magnitude, particularly during the last deglaciation when global sea-level rise caused rapid flooding of the land area thereafter constituting the vast East Siberian Arctic Shelf. Demonstration of past warming-induced release of permafrost carbon provides a benchmark for the sensitivity of these large carbon pools to changing climate.

**INTRODUCTION**

Anthropogenic climate change has, over recent decades, resulted in rapid warming of the Arctic region (1). One consequence is thawing of permafrost (1), which currently holds a stock of organic carbon (OC) estimated to be 1300 Pg (2), about twice the inventory of atmospheric CO$_2$. Permafrost thaw includes gradual changes, such as deepening of the seasonally thawed surface soil (i.e., active layer) and loss of permafrost extent, as well as rapid thaw by thermal landscape collapse, thermokarst, and erosion of coastlines and river banks (3, 4). By exposing previously freeze-locked OC to microbial degradation, these processes may cause a permafrost carbon–climate feedback that reinforces anthropogenic emissions of greenhouse gases (e.g., CO$_2$ and CH$_4$) (1). Anticipating the strength and timing of this nonlinear carbon cycle response is a major challenge in climate change science. Past warming events may provide our best opportunity to learn about the response behavior of the massive freeze-locked permafrost OC pool to a warming climate.

The last deglaciation—here defined as the time period between the Last Glacial Maximum [LGM; 26 to 20 thousand years before the present (ka B.P.)] (5) and the early Holocene climate optimum (EH; 10 to 8 ka B.P.)—marked a period of profound climate change. This period witnessed rapid warming by ~3.5°C in the Northern Hemisphere (6), the demise of Arctic ice sheets resulting in a global sea-level rise of 134 m (5), and a rise of atmospheric CO$_2$ concentrations by 80 ppmv (parts per million by volume) (7). It is also suggested to have caused broad reorganization of OC pools in Arctic permafrost (8). This particularly affected the ice- and OC-rich Ice Complex deposits (ICD; also referred to as Yedoma), which developed during the Pleistocene in nonglaciated Arctic regions (9). During the LGM global sea-level low stand of −134 m, up to 40-m-thick ICD accumulated in northeastern Siberia likely over large areas that now make up the world’s largest conventional shelf—the East Siberian Arctic Shelf (ESAS) (8). The postglacial sea-level rise and flooding of what became the ESAS is thought to have removed about 220 to 260 Pg of OC (8, 10), leaving 400 Pg of total above-sea ICD-OC in near-coastal areas in Siberia and Alaska (Fig. 1) (10).

During the last deglaciation, large-scale permafrost thawing, including thaw of inland permafrost (11, 12) and flooding with resultant thermo-erosion of ICD on the then-exposed ESAS (13), is suggested to have liberated CO$_2$ by biodegradation of the thawed organic matter and thus have contributed to an increase in atmospheric CO$_2$ concentrations by 80 ppmv or 200 Pg carbon (14–16). Another important source of CO$_2$ was outgassing of deep ocean water by changing oceanic circulation at that time (7). However, atmospheric archives suggest that the deglacial rise in CO$_2$ was caused by $^{13}$C- and $^{14}$C-depleted carbon, and oceanic CO$_2$ alone may not fully explain this isotopic anomaly (15). Hence, inundation of the Arctic Ocean shelves and degradation of permafrost OC, followed by emissions of $^{14}$C-depleted CO$_2$, may also have contributed to part of the rise in atmospheric CO$_2$ (14). A few spatially limited and noncontinuous observational studies have confirmed with radiocarbon dating that the deglacial sea-level rise remobilized old OC, likely due to coastal retreat of permafrost (13, 17). Two studies from the northeast Pacific suggest that this also happened outside the Arctic Ocean during shelf inundation in the Bering Sea and the Sea of Okhotsk (Fig. 1) (14, 18). There is, however, no single continuous archive that explores the remobilization of OC from Arctic permafrost during the last deglaciation, leaving both the magnitude and the onset and end of this hypothetically massive permafrost OC remobilization largely unconstrained.

Here, we present the first continuous record of permafrost OC remobilization from the Siberian-Arctic continental margin from before the LGM (27 ka B.P.) to the preindustrial. Carbon isotopes ($^{14}$C and $^{13}$C) and terrigenous biomarkers [lignin phenols; high-molecular weight (HMW) n-alkanes and n-alkanoic acids] were analyzed in the marine sediment core SWERUS-L2-31-PC1 (below...
study provides the first evidence for permafrost carbon export.

The mixing model thus includes two terrigenous end members, contributions to the terrestrial OC that was released. Accordingly, the dual-isotope source apportionment (~3.9 g m⁻² year⁻¹) is supported by terrigenous biomarkers, which were consistently higher in this period compared to the subsequent LGM (lignin phenols >10, HMW n-alkanes ~4, HMW n-alkanoic acids ~8; Fig. 2).

A provenance proxy based on lignin phenols provides additional information about the source of the terrigenous OC. The low ratio of syringyl over vanillyl phenols (S/V) indicates a strong contribution of gymnosperm lignin tissues (24) in sediments deposited between 27 and 26 ka B.P. (S/V, 0.41 ± 0.04; n = 11). This resembles the fingerprint of end members from more southern biomes hosting gymnosperm forests, similar to contemporary suspended fluvial material in the Lena river (~0.4) that drains these forests in central Siberia (Fig. 3) (25). Hence, comparatively shallow active layer material mobilized in central Siberia and transported by rivers is the most likely source of the large amounts of OC deposited on the southern Lomonosov Ridge between 27 and 26 ka B.P. The limited contribution from coastal thermo-erosion of permafrost at that time is consistent with global sea-level regression during the buildup to the LGM (5).

Biomarker degradation proxies suggest that terrigenous OC deposited 27 to 26 ka B.P. was less degraded than terrigenous OC deposited during the LGM, which indicates rapid remobilization and fluvial transport of the organic matter. This is shown by the ratio of 3,5-dihydrobenzoic acid relative to vanillyl (3,5-Bd/V), reflecting the removal of the more vulnerable vanillyl with increasing degradation of OC starting from 27 ka B.P. (0.6 to 1.6; fig. S2) toward the LGM (>5), as well as by the preferential decomposition of HMW n-alkanoic acids relative to HMW n-alkanes (1.5 versus 0.4 to 0.7). Taken together, the degradation pattern reveals transport of little-degraded plant material from distant sources, likely located in central Siberia, which suggests a period of accelerated discharge or a pulse of terrigenous OC to the Arctic Ocean.

Dansgaard-Oeschger (DO) events represent periods of abrupt warming and strong hydrological change during the last glacial period (26). While DO events are known to affect the global climate and carbon cycle, only a few records are able to attribute environmental change in Siberia to the effects of DO events. A study from Lake Baikal suggested that profound changes in precipitation triggered soil erosion events in the lake catchment synchronous to DO and Heinrich events in the North Atlantic region (27). It is likely that the terrigenous OC peak in 31-PC resulted from surface permafrost thawing in response to abrupt climate warming and increased precipitation during the DO-3 event peaking around 28 ka B.P. (Fig. 2) (26). There is greater uncertainty around this event compared to the two more recent warming events detailed below.
The 31-PC record demonstrates a minimum in terrigenous OC transport relative to the LGM (28) disrupted major transport pathways of terrigenous OC to the Arctic Ocean. Previous research stresses that the LGM probably caused large sea-ice expansion with periods of year-round coverage (19, 29) and perhaps even partial ice-shelf coverage in some parts of the Arctic (30). A more extensive permanent sea ice cover during the LGM is consistent with low OC accumulation rates in sediments along and off the Siberian continental margin (19, 29).

Despite the cold glacial climate, the predominance of gymnosperm lignin tissues suggests that Arctic rivers likely remained an important gateway for terrigenous OC to reach sediments on the Siberian continental margin. The ratio of S/V indicates higher contribution of gymnosperm lignin tissues (24) during the LGM (0.37 ± 0.12; n = 7) compared to the DO-3 event (0.41 ± 0.04; n = 11; Fig. 3) and even the Holocene (0.70 ± 0.37; n = 28).

Biomarker degradation proxies further suggest more extensive degradation of terrigenous OC during the LGM. The 3.5-Bd/V record shows that terrigenous OC deposited during the LGM was strongly degraded (>5; n = 12; fig. S2), which is also supported by the depletion of HMW n-alkanoic acids relative to HMW n-alkanes (0.4 to 0.8 in the LGM versus 1.5 in the DO-3 pulse). Disregarding possible depth uncertainties due to local deviation from the global eustatic mean, the paleo-shoreline was likely less than 100 km from the 31-PC location during the global LGM sea-level low stand. Therefore, the long transport time of terrigenous OC to the 31-PC location, indicated by the degradation state, supports river transport from more southern locations during the LGM. Taken together, the LGM marks a minimum in OC accumulation with most of the OC delivered from distant terrigenous sources, likely with Arctic rivers being an important transport pathway at that time.

**Early deglaciation and carbon remobilization during the Bølling-Allerød warming**

The early deglaciation, including the Heinrich stadial 1 (HS-1; also referred to as the Older Dryas), kept permafrost OC pools in a freeze-locked state. Although the HS-1 exhibits the first increase in atmospheric CO₂ after the LGM at 17.5 k.a. B.P., results from 31-PC suggest that low terrigenous OC fluxes persisted from the LGM to the HS-1. Our results are largely in line with low accumulation rates of HMW n-alkanes in sediment archives from the Bering Sea and the Sea of Okhotsk for HS-1 (fig. S5) (14, 18). This suggests that despite a rise in global sea level by ~35 m (5) between 20 and 14.7 k.a. B.P., cold glacial conditions with steady LGM-like temperatures in the Arctic (6) protected permafrost carbon from substantial thaw during this period.

The Bølling-Allerød (BA; 14.7 to 12.9 k.a. B.P.), a period of rapid atmospheric warming in the Northern Hemisphere (6), saw the first substantial increase in terrigenous OC transport relative to the LGM and HS-1. The accumulation rates of HMW n-alkanes and HMW
n-alkanoic acids deposited between 14.7 and 12.9 ka B.P. were twice as large as those recorded before 14.7 ka B.P. (Fig. 2), while fluxes of lignin phenols remained at LGM and HS-1 levels. Furthermore, predepositional $^{14}$C-OC ages (10.1 ± 4.9 ka; $n = 5$) doubled compared to the organic matter deposited during the HS-1 and LGM (4.5 ± 5.3 ka; $n = 7$), pointing to the release of relatively older OC. Previous studies of sediments in the Sea of Okhotsk (14) and the Bering Sea (18) reported compound-specific $^{14}$C ages of terrigenous biomarkers (~10 ka old at the time of deposition) similar to the $^{14}$C-OC ages observed in this study, suggesting widespread release of strongly preaged terrigenous matter during the BA warm event.

Dual carbon isotope–based source apportionment reveals that the BA warming by nearly 1°C in the Northern Hemisphere (Fig. 4) (6) triggered the remobilization of permafrost OC mainly from ICD, accounting for 64 ± 6% ($n = 5$) of the OC after 14.7 ka B.P. compared to lower and more variable amounts (38 ± 29%; $n = 10$) during DO-3, LGM, and HS-1. The flux of permafrost carbon from ICD to 31-PC more than tripled from the LGM and HS-1 baseline (0.3 ± 0.1 g m$^{-2}$ year$^{-1}$) to the BA (1.1 ± 0.1 g m$^{-2}$ year$^{-1}$). This is in line with reconstructed fluxes of ICD-OC to the Chukchi Sea (Figs. 1 and 4), which were three times higher during the late BA (13.1 to 12.9 ka B.P.) than in the Holocene (13). The synchronous observational records in the Chukchi Sea and on the southern Lomonosov Ridge (31-PC) together represent a wide drainage footprint (ESAS) and suggest that thawing and remobilization of OC from ICD was a large-scale phenomenon across the entire ESAS coastline and drainage basin.

Biomarker degradation proxies suggest a high degree of decomposition of the OC released by thawing ICD during the BA (Fig. S2). The 3,5-Bd/V ratio (~8.1) remains in the same range of degradation during HS-1 (~8.8) and an even stronger depletion of HMW n-alkanoic acids relative to HMW n-alkanes during the BA (0.2 to 0.5) when compared with the HS-1 (0.6 and 0.7). The high degree of decomposition indicates a predominance of thaw and remobilization of OC from inland ICD on today’s ESAS by thermokarst, thermo-erosion, and, possibly, riverbank erosion (9), with degradation both on site and during long-distance transport to the 31-PC location. It appears that only a smaller amount of the OC was remobilized by coastal erosion resulting from global sea-level rise (~27 m) and inundation of about 10% of the total ESAS during the BA (5, 31). Irrespective of the exact mobilization mechanism, the (i) highly enhanced liberation of old OC from ICD permafrost and (ii) molecular evidence for extensive degradation support the hypothesis that ICD thawing during the BA contributed to the concurrent rise of atmospheric CO$_2$ and its isotopic changes (13C, 14C) (Fig. 4) (14, 15). However, the exact amount of OC that was stored as ICD on exposed Arctic shelves is still challenging to estimate (8), and it is not yet possible to quantitatively constrain the magnitude of the fraction that got converted to CO$_2$ after thaw. In summary, the BA warming at 14.7 ka B.P. activated the thawing and degradation of old Pleistocene ICD permafrost that covered the large “dry land” ESAS and thereby released large amounts of OC into active cycling.

**Massive permafrost carbon remobilization during the Early Holocene**

The most striking observational evidence for a large remobilization of permafrost OC is during the EH. Isotope-traced OC and biomarker fluxes show a sudden and strong increase in deposition of released permafrost OC, arriving after long lateral transport on the Lomonosov Ridge between 10 and 7.6 ka B.P. The PF-C flux from ICD was, on average, 2.7 ± 0.6 g m$^{-2}$ year$^{-1}$ (10 to 9 ka B.P.; $n = 4$), which is roughly one order of magnitude higher than the flux during the LGM and HS-1 (0.3 ± 0.1 g m$^{-2}$ year$^{-1}$) and two to three times higher than during the BA (1.1 ± 0.1 g m$^{-2}$ year$^{-1}$). This is despite the fact that the 31-PC location was much further from land during the EH than during the earlier periods (text S2 and fig. S5), which likely changed the depositional dynamics and thus sedimentation rates as large parts of the ESAS were flooded during the EH. Source apportionment shows that the release of permafrost OC

**Fig. 3. Lignin source diagnostics of terrigenous material in the 31-PC (80 samples) compared with previously studied sediment cores from the Siberian Shelf 4-PC (13), GC-58 (17), and PC-23 (11), as well as coastal ICD, AL permafrost (32), and Lena river particulate OC (25).** The size of the 31-PC circles is proportional to the magnitude of the respective lignin flux (0.01 to 6.6 mg year$^{-1}$ m$^{-2}$). The abundance ratio of syringyl over vanillyl (S/V) indicates contributions of angiosperm compared to gymnosperm plants, and the ratio between cinnamyl and vanillyl (C/V) distinguishes between woody (e.g., stems) and nonwoody (e.g., leaves) plant tissues (24).

Martens et al., Sci. Adv. 2020; 6 : eabb6546 16 October 2020
Martens et al., Sci. Adv. 2020; 6 : eabb6546     16 October 2020

**DISCUSSION**

This new time-continuous record from the Siberian continental slope provides insights into the release of previously inactive OC from permafrost across northeastern Siberia over the last 27 ka. Characterization of OC using carbon isotopes ($\delta^{13}C$ and $\Delta^{14}C$) and terrestrial biomarkers reveals that during stadial conditions (LGM, HS-1), the Siberian continental margin received low inputs of fluvially transported terrigenous OC. By contrast, three pulses of permafrost OC release coincide with the three last largest warming events recorded in Greenland ice cores (26). Abrupt climate warming during the DO-3 event appears to have caused a large remobilization of relatively young OC from thawing of predominantly permafrost active layer with subsequent fluvial transport to the Arctic Ocean. Warming and global sea-level rise during the deglaciation

from active layer thawing was a minor input to sediments on the Siberian continental margin during the EH (0.7 ± 0.4 g m$^{-2}$ year$^{-1}$). The lignin-based source fingerprint [ratios of C/V (cinnamyl/vanillyl) and S/V] reveals that OC remobilized during the EH contained notably more angiosperm tissues (S/V, 0.53 ± 0.11; $n = 15$) than during the LGM, suggesting a more tundra-like OC source similar to organic material stored in ICD (Fig. 3) (32).

Our results of enhanced input of remobilized OC for the Younger Dryas (YD)—EH warming expands upon an earlier report on this (11). Tesi and coworkers (11) showed that the dominant source of the large OC input to their sampling location in the Laptev Sea was fluvially transported material from the permafrost active layer (Fig. 1). This previous study built on source apportionments in core PC-23 (Fig. 1), which is located near the mouth of the paleo-Lena river. It is likely that OC transported to this location showed a particularly strong contribution of river-transported material from inland Siberia that may not be representative for most of the ESAS. An increasing number of studies with a broader regional footprint (East Siberian Sea; Chukchi Sea) (13, 17), as well as the present study, now indicate that deglacial ESAS sediments and its large OC input are dominated by remobilized ICD permafrost, which was released by EH warming and sea-level rise.

Permafrost OC remobilization during the EH caused extensive underway degradation. The OC remobilized during the EH was, however, somewhat less degraded than in the earlier periods, which is consistent with a massive pulse-like remobilization of OC. A comparison of the travel distance for terrigenous OC with modern cross-shelf degradation trends in the Laptev Sea (33) shows that the 3,5-Bd/V ratio in 31-PC for the EH interval between 10 and 7.6 ka B.P. (0.4 to 0.5) agrees with the 3,5-Bd/V ratio of ~0.5 at a cross-shelf transport distance of about 400 km or more (33). The modern system relationship between transport distance and the 3,5-Bd/V degradation status is consistent with the EH period observations as the offshore distance of the core site increased from about 200 to 400 km between 11 and 7.6 ka B.P. (5, 31). However, other degradation status proxies such as the ratio of HMW n-alkanics over HMW n-alkanes (0.7 ± 0.3) are even somewhat lower than expected based on the comparison above (acids/alkanes > 1.9) (33). Given the extent of degradation indicated by the lipid biomarkers, it seems that large part of the initially remobilized terrigenous OC during the EH was degraded during cross-shelf transport, implying release of $^{14}C$-depleted CO$_2$ to the ocean with subsequent venting to the atmosphere.

The large global sea-level rise of ~44 m (5) and the inundation of the ESAS displaced the coast line by about 200 km during the EH (31, 34). Considering the large offshore distance of the 31-PC location by the time of the EH, resulting in longer cross-shelf transport times, we expect a substantial time gap between permafrost remobilization and final (re-)deposition of the OC on the Lomonosov Ridge. We estimate (text S2 and fig. S5) that cross-shelf transport may delay the seaward transport of terrigenous OC by 0.6 to 2.7 ka for material released during the EH. In contrast, cross-shelf transport times during earlier periods of permafrost OC remobilization (BA and DO-3) were much shorter (below 0.2 ka; fig. S5). Accordingly, OC deposited at the 31-PC location between 10 and 7.6 ka B.P. was likely released from land in the time window 12.5 to 8.2 ka B.P., based on the two cross-shelf transport scenarios (text S2). This period thus coincides with the EH onset around 11.7 ka B.P. (Fig. 4) when northern hemispheric temperatures increased by more than 1°C (6), and massive permafrost OC release to Laptev Sea sediments has been demonstrated (11). The EH warming likely resulted in large-scale permafrost thawing and net emissions of $^{14}C$-depleted CO$_2$ to the atmosphere.
(BA and EH) caused remobilization of much older OC from ICD permafrost on the broad aerially exposed Siberian shelf, via thermokarst formation, riverbank erosion, and coastal thermo-erosion. This occurred in two stages: (i) during the BA (14.7 to 12.9 ka B.P.) where reconstructed fluxes of terrigenous biomarkers and OC source fractions reveal permafrost OC release two to three times higher than during the LGM and (ii) an even larger release during the EH (centered around ~11.7 ka B.P.), with fluxes one order of magnitude higher than during stadial conditions (LGM, HS-1). Biomarker degradation proxies suggest strong underway OC degradation throughout the 31-PC, which implies that massive OC release during these three warming pulses (DO-3, BA, and EH) not only transferred large amounts of old OC from terrestrial to aquatic systems but also caused enhanced OC degradation with resulting emissions of $^{14}$C-depleted CO$_2$ to the atmosphere.

The results from this study on large-scale OC remobilization from permafrost are consistent with a growing set of observational records from the Arctic Ocean and provide support for modeling studies that simulated large injections of CO$_2$ into the atmosphere during deglaciation (14–16). This demonstrates that Arctic warming by only a few degrees may suffice to abruptly activate large-scale permafrost thawing, indicating a sensitive trigger for a threshold-like permafrost climate change feedback.

**MATERIALS AND METHODS**

**Study area and large-scale carbon sources**

The Siberian continental margin is the northern margin of the Eurasian continent and recipient of large terrestrial export of sediments and OC (Fig. 1). Taken together, the shelf area of the Laptev, East Siberian, and Russian Chukchi Seas constitutes the world’s largest continental shelf area, i.e., the ESAS, where about half of the area is shallower than 50 m depth (31). This region today receives terrigenous OC both from erosion of permafrost coastlines and from riverine transport of permafrost material from continental Siberia (20), of which OC from thermo-erosion of ICD exceeds OC released from the permafrost active layer and riverine transport by a factor of 2 (20, 35). Within the boundaries of the shelf, terrigenous OC input strongly influences the ESAS water chemistry (36) and dominates the composition of ESAS surface sediment OC (32, 33). Beyond the shelf edge, marine planktonic OC sources become a larger contributor to the sediment OC pool (33).

**Sampling**

The sediment core SWERUS-L2-31-PC1 was collected on 2014-09-15 in 1120 m water depth (79°54.89′N, 143°14.01′E) during the Swedish-Russian-U.S. Investigation of Climate, Cryosphere, and Carbon interactions in the East Siberian Arctic Ocean (SWERUS-C3) program onboard the Swedish icebreaker *Oden*. Basic physical (bulk density and magnetic susceptibility) and geochemical analyses (x-ray fluorescence spectroscopy) were performed shipboard using a Multi-Sensor Core Logger (Geotek, UK) and onshore using an ITRAX core scanner (Cox Analytical, Sweden), respectively. Subsampling for OC characterization (OC, δ$^{13}$C, Δ$^{14}$C, and molecular biomarkers) at 10-cm resolution was carried out at Stockholm University.

**Age-depth model of core 31-PC**

The sedimentation history of the 8-m-long core 31-PC encompasses the last 27 ka (fig. S1) and builds on stratigraphic correlation to Greenland ice-core records within the constraints of radiocarbon dating, using a novel Bayesian probabilistic alignment method (19). Accordingly, this environmental record starts at the onset of early Marine Isotope Stage (MIS-2), which includes the entire LGM (26.5 to 20 ka B.P.) (5), the early deglaciation including the HS-1 (late MIS-2; 20 to 14.7 ka B.P.), the BA warm interval (late MIS-2; 14.7 to 12.9 ka B.P.), the YD cold interval (late MIS-2; 12.9 to 11.7 ka B.P.), the rapid warming transition into the EH, and then the entire Holocene (MIS-1; 11.7 ka B.P. to today). Details about the age model construction are presented in a previous study (19).

**Carbon isotope analysis**

Stable carbon isotope (δ$^{13}$C) ratios are a powerful tool to distinguish between terrigenous and marine sources of OC. Ratios around −27‰ typically characterize terrigenous biomass in marine sediment OC, whereas phytoplankton usually distributes around −21‰. For the current study, we analyzed 80 freeze-dried and homogenized bulk sediment samples for δ$^{13}$C and OC. For this purpose, ~10 mg of sediment was weighed in silver capsules and acidified with 3 M HCl to remove carbonates. Analyses were performed using a Carlo Erba NC2500 elemental analyzer coupled to an isotope-ratio mass spectrometer (Finnigan DeltaV Advantage) in the Department of Geological Sciences, Stockholm University. The OC concentrations were previously published in a paper describing the 31-PC chronology (19).

Radiocarbon ages ($^{14}$C) of OC provide additional information about the source contributions of different terrestrial OC pools given the large age offset between more contemporary permafrost soils and strongly pre-aged permafrost deposits from the Late Pleistocene. A subset of 27 samples was sent to the U.S. National Ocean Science Accelerator Mass Spectrometry (NOSAMS) facility at Woods Hole Oceanographic Institution (Woods Hole, MA, USA) for $^{14}$C analysis. To remove carbonates, the sediments were treated with HCl vapor at NOSAMS. To calculate the predepositional $^{14}$C activity of the organic matter, we accounted for the $^{14}$C decay of the OC after the year of deposition (Yd) provided by the age-depth model (19), following Eq. 1

$$\Delta^{14}C = (Fm \times e^{\lambda(1950-Yd)} - 1) \times 1000$$

where Fm is the fraction modern provided by the AMS and $\lambda$ is the decay constant, i.e., 1/(true mean life) of radiocarbon (1/8267). Accordingly, the predepositional ages represent the age gap between fixation of the atmospheric $^{12}$C-CO$_2$ and deposition at the core location.

**Specific mineral surface area of the sediment samples**

For sediment surface area (SA) analysis, an aliquot of about 0.7 g of freeze-dried sediment was heated to 400°C for 12 hours to combust all organic matter. After cooling for at least 10 hours, the samples were repeatedly washed with 50 ml of Milli-Q water and pelletized in a centrifuge for 10 min at 8000 rpm for removal of water, followed by 48 hours of freeze drying for complete water removal. Before SA analysis, the samples were degassed under N$_2$ purge at 200°C for at least 2 hours. Specific SA was finally measured by six pressure point measurements on a Micromeritics Gemini VII SA and Porosity analyzer. The instrument performance was monitored regularly using two reference materials (black carbon and TiO$_2$) provided by Micromeritics.
Molecular biomarker analysis: CuO products and lignin phenols

We used lignin phenols as one of several means to trace terrigenous OC in marine sediments (24). A CuO oxidation protocol (32) using a microwave system was used to biopolymer breakup. Depending on the total OC content, 1 to 1.8 g of freeze-dried sediment were loaded into Teflon tubes, mixed with 300 mg of cupric oxide (CuO) and 50 mg of ammonium iron (II) sulfate hexahydrate [(NH₄)₂Fe(SO₄)₂·6 H₂O], and suspended in N₂-purged 2 M NaOH solution. The extraction program used an UltraWave Milestone 215 microwave digestion system using a program of 90-min duration at 130°C. After extraction, an internal recovery standard (ethyl-vanillin, cinnamic acid) was added and samples were acidified to pH 1 using concentrated HCl. Afterward, phenols were extracted from the water phase using ethyl acetate (EtOAc) and residual water was removed from the supernatant using anhydrous sodium sulfate (Na₂SO₄). The EtOAc was evaporated in a CentriVap (Christ RVC 2-25) at 60°C for 1 hour and redissolved in pyridine.

For analysis, the extracts were derivatized using bis-trimethylsilyl trifluoroacetamide (BSTFA) and 1% trimethylchlorosilane (TMCS) to silylate exchangeable hydrogen atoms and then analyzed in single ion monitoring (SIM) mode on a gas chromatograph (GC) with mass spectrometer (GC-MS detector (GC-MS 7820A, Agilent Technologies USA) using a DB1-MS column (30 m x 0.25 mm; 0.25 μm film thickness). The GC program had an initial temperature of 50°C and a subsequent heating ramp of 10°C per minute until 300°C. Individual compound concentrations were calculated using a seven-point calibration of external quantification standards of long-chained n-alkanes and n-alkanoic acids. The recoveries of the internal standards were 92 ± 12% for the nonpolar fractions and 57 ± 20% for the acid fractions. As for lignin phenols, this study reports concentrations of HMW n-alkanes (ΣC₃₃-C₃₃) and n-alkanoic acids (ΣC₂₄-C₃₂) as normalized to SA (ng m⁻²) and as flux rates (mg m⁻² year⁻¹). We also report the carbon preference index (CPI; table S3) as a degradation proxy of terrigenous OC for both compound classes [CPIₖᵣᵢₑ₄ for HMW n-alkanes; CPIₕᵢₐᵓᵂ for HMW n-alkanoic acids; (33)], which is based on the degree of the odd-over-even predominance of HMW n-alkane homologues and the even-over-odd predominance of HMW n-alkanoic acids. Both decrease toward 1 with degradation.

Statistical method for source apportionment of OC

This study uses a Bayesian statistical approach (21) to calculate the relative fractions derived from the major OC sources (i.e., ICD, permafrost active layer, and marine biomass) using the Δ¹³C-OC and Δ¹⁴C-OC values in core 31-PC (fig. S3). This method applies a dual-isotope mass balance with three end members in a Markov chain Monte Carlo simulation (21) using Matlab R2018a with 1,000,000 runs and a burn-in period of 10,000 runs per sample. The isotopic definition of the end members was based on an extensive literature survey of Δ¹³C and Δ¹⁴C values measured in potential OC sources following the approach used in a number of previous ESAS studies (11, 13, 20). Each end member consists of an isotopic mean ± SD for both Δ¹⁴C-OC and Δ¹³C-OC and thus accounts for the natural end member variability. Analytical errors are not included in the mixing model, as these are considered negligible compared to the uncertainties of the end member variability. For marine biomass, Δ¹⁴C-OC of −50 ± 12‰ (n = 5) and Δ¹³C-OC of −21.0 ± 2.6‰ (n = 31) was used, representing an open-marine Arctic environment (13). The permafrost active layer end member was based on active layer observations from Siberia (maximum of 1 m depth), with Δ¹⁴C-OC of −197.5 ± 148.3‰ (n = 60; mean ± SD) and Δ¹³C-OC of −26.4 ± 0.8‰ (n = 56; fig. S3) (22). The Δ¹³C-OC of the ICD end member was constrained on the basis of a previous review on ICD exposures in Siberia (−26.3 ± 0.7‰; n = 374) (23). For the Δ¹⁴C-OC of ICD, however, we needed to consider the temporal depth of the 31-PC record (i.e., 27 ka B.P.), which overlaps with the period of active ICD formation ca. 120 to 10 ka B.P. (23, 38), as well as the limits of radiocarbon dating that allow no consideration of deposits older than ca. 50 ka. We therefore used a previously published database.
of $\Delta^{14}$C-OC in Siberian ICD exposures (−962 ± 61% $n = 415$) (22) to assess the variability in ICD formation over time. On the basis of this assessment, this study describes the ICD end member assuming 9. N. N. Romanovskii, H.-W. Hubberten, A. V. Gavrilov, V. E. Tumskoy, G. S. Tipenko, et al.: eabb6546     16 October 2020

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SUPPLEMENTARY MATERIALS
Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/6/42/eabb6546/DC1

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Acknowledgments: We thank the crew of the Swedish icebreaker i/b Oden. We further thank C. Johansson and H. Siegmund for their help with core handling and OC analysis; N. Barrientos, G. West, and E. Sivertsson for their help in the laboratory; and A. Andersson and H. Holmstrand for their advice on source apportionment and analytical work. We also want to thank two anonymous reviewers and the editor who improved the manuscript with their suggestions.

Funding: The SWERUS-C3 program was supported by the Knut and Alice Wallenberg Foundation (KAW contract 2011.0027 to Ö.G. and M.J.) and the Swedish Research Council (VR contract 621-2013-5297 to Ö.G., 621-2012-1680 to M.J.). This study was also supported by the European Research Council (ERC AdG CC-TOP 695331 to Ö.G.), the VR Distinguished Professorship Program (contract 621-2017-01601 to Ö.G.), the Russian Government (grant 14, Z50.31.0012 to I.S.), and the Russian Science Foundation (grant 15-17-20032 to O.V.D.).

Author contributions: The SWERUS-C3 program and research concepts were developed by a coordinating team led by Ö.G., M.J., and I.S. B.W. and Ö.G. supervised the editing and writing of the paper. J.M., B.W., and Ö.G. lead the conceptual development of this study. J.M. analyzed the samples, performed source apportionments, and drafted and coordinated the manuscript. F.M. and M.O. provided the age model for 31-PC. M.O., M.J., I.S., O.V.D., and Ö.G. participated in the sampling during the SWERUS-C3 program. All authors were involved in the interpretation of the results and participated in the editing process of the manuscript.

Competing interests: The authors declare that they have no competing interests.

Data and materials availability: All data needed to evaluate the conclusions in the paper are present in the Supplementary Materials and are available on the Bolin Centre Database at https://bolin.su.se/data/martens-2020.

Submitted 9 March 2020
Accepted 4 September 2020
Published 16 October 2020
10.1126/sciadv.abb6546

Citation: J. Martens, B. Wild, F. Muschitiello, M. O’Regan, M. Jakobsson, I. Semiletov, O. V. Dudarev, O. Gustafsson, Remobilization of dormant carbon from Siberian-Arctic permafrost during three past warming events. Sci. Adv. 6, eabb6546 (2020).