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1 **Erosion of organic carbon in the Arctic as a geological carbon dioxide sink**

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16 **Soils of the northern high latitudes store carbon over millennial timescales ( $10^3$  yrs) and**  
17 **contain approximately double the carbon stock of the atmosphere<sup>1-3</sup>. Warming and**  
18 **associated permafrost thaw can expose soil organic carbon and result in mineralisation**  
19 **and carbon dioxide (CO<sub>2</sub>) release<sup>4-6</sup>. However, some of this soil organic carbon may be**  
20 **eroded and transferred to rivers<sup>7-9</sup>. If it escapes degradation during river transport and**  
21 **is buried in marine sediments, then it can contribute to a longer-term ( $>10^4$  yrs),**  
22 **geological CO<sub>2</sub> sink<sup>8-10</sup>. Despite this recognition, the erosional flux and fate of**  
23 **particulate organic carbon (POC) in large rivers at high latitudes remains poorly**  
24 **constrained. Here, we quantify POC source in the Mackenzie River, the main sediment**  
25 **supplier to the Arctic Ocean<sup>11,12</sup> and assess its flux and fate. We combine measurements**

26 **of radiocarbon, stable carbon isotopes and element ratios to correct for rock-derived**  
27 **POC<sup>10,13,14</sup>. Our samples reveal that the eroded biospheric POC has resided in the basin**  
28 **for millennia, with a mean radiocarbon age of 5800±800 yr, much older than large**  
29 **tropical rivers<sup>13,14</sup>. Based on the measured biospheric POC content and variability in**  
30 **annual sediment yield<sup>15</sup>, we calculate a biospheric POC flux of 2.2<sup>+1.3</sup><sub>-0.9</sub> TgC yr<sup>-1</sup> from**  
31 **the Mackenzie River, three times the CO<sub>2</sub> drawdown by silicate weathering<sup>16</sup>. Offshore**  
32 **we find evidence for efficient terrestrial organic carbon burial over the Holocene,**  
33 **suggesting that erosion of organic carbon-rich, high latitude soils may result in a**  
34 **significant geological CO<sub>2</sub> sink.**

35       Photosynthesis and the production of organic carbon by the terrestrial biosphere  
36 (OC<sub>biosphere</sub>) is a major pathway of atmospheric carbon dioxide (CO<sub>2</sub>) drawdown. Over  
37 millennial timescales (10<sup>3</sup> yrs), some OC<sub>biosphere</sub> escapes oxidation and contributes to a  
38 transient CO<sub>2</sub> sink in soil<sup>2,3,17</sup>. Longer-term CO<sub>2</sub> drawdown (>10<sup>4</sup> yrs) can be achieved if  
39 OC<sub>biosphere</sub> is eroded, transferred by rivers and buried in sedimentary basins<sup>9,10,18,19</sup>. Burial of  
40 OC<sub>biosphere</sub> represents a major geological CO<sub>2</sub> sink (and O<sub>2</sub> source) alongside the chemical  
41 weathering of silicate minerals by carbonic acid, coupled to carbonate precipitation<sup>16,19</sup>.  
42 These fluxes negate CO<sub>2</sub> emissions from the solid Earth<sup>20</sup> and from oxidation of rock-derived  
43 OC<sup>21</sup>, contributing to the long-term regulation of global climate<sup>19,20</sup>. Physical erosion is  
44 thought to play a significant role in this OC<sub>biosphere</sub> transfer because it controls the rate of  
45 biospheric particulate organic carbon (POC<sub>biosphere</sub>) export by rivers<sup>22</sup> and influences sediment  
46 accumulation and the efficiency of OC burial<sup>10,18,23</sup>.

47       In the northern high latitudes, large amounts of OC<sub>biosphere</sub> are stored in soil<sup>1,2</sup>. The  
48 upper three meters of soil in the region of northern circumpolar permafrost are estimated to  
49 contain 1035±150 PgC, approximately double the carbon dioxide (CO<sub>2</sub>) content of the pre-  
50 industrial atmosphere<sup>17</sup>. Many of these soils accumulated during the retreat of large

51 continental ice sheets following the Last Glacial Maximum, with a peak expansion between  
52 12,000 and 8,000 cal. yr BP<sup>24</sup> and the OC<sub>biosphere</sub> can be thousands of years old<sup>8</sup>. This vast  
53 carbon reservoir is located in a region sensitive to environmental change over glacial-  
54 interglacial timescales<sup>24</sup> and to warming over the coming century<sup>3</sup>. Much focus has been  
55 placed on its potential to become a CO<sub>2</sub> source<sup>3-6,8</sup>. However, geological CO<sub>2</sub> drawdown by  
56 POC<sub>biosphere</sub> erosion at high latitudes has remained poorly constrained<sup>9</sup>.

57 Here we sample POC carried by the major rivers in the Mackenzie Basin and  
58 investigate its fate using an offshore sediment core extending over the Holocene (Extended  
59 Data Fig. 1). The Mackenzie River is the largest source of sediment to the Arctic Ocean<sup>11,12,15</sup>  
60 and erosion of mountainous topography in the basin results in a high sediment discharge,  
61 similar to the combined total of 16 Eurasian rivers draining to the Arctic<sup>11,15</sup>. We collected  
62 river depth profiles to characterise POC across the range of grain sizes carried by large  
63 rivers<sup>13,14,25</sup> at the main conduit for sediment export to the Arctic Ocean in the Mackenzie  
64 Delta, at key points on the Mackenzie River and from its major tributaries (Extended Data  
65 Fig. 1). To investigate temporal variability of POC composition, river depth profiles were  
66 collected shortly after ice-break up at high/rising stage (June 2011) and during falling stage  
67 (September 2010), while river surface and bank samples were collected in June 2009. To  
68 correct for rock-derived, ‘petrogenic’ POC (POC<sub>petro</sub>), likely to be important in the  
69 Mackenzie Basin<sup>7,25</sup>, we combine measurements of radiocarbon (<sup>14</sup>C, reported as the ‘fraction  
70 modern’ F<sub>mod</sub>), total OC content ([OC<sub>total</sub>], %), stable isotopes of OC (δ<sup>13</sup>C<sub>org</sub>), nitrogen to OC  
71 ratio (N/OC<sub>total</sub>) and aluminium to OC ratio (Al/OC<sub>total</sub>), which allow us to assess the age and  
72 concentration of POC<sub>biosphere</sub> (Methods)<sup>7,10,13,14,22</sup>. Published surface samples from the  
73 Mackenzie River<sup>4,7</sup> (n=5) have <sup>14</sup>C-ages between 6,010 yr and 10,000 yr but the <sup>14</sup>C-depletion  
74 caused by POC<sub>petro</sub> versus aged POC<sub>biosphere</sub> has not been assessed. We also examine the

75 hydrodynamic behaviour of POC, using the aluminium to silicon ratio (Al/Si) ratio as a proxy  
76 of sediment grain size<sup>25</sup>.

77 We find that river POC is <sup>14</sup>C-depleted throughout the Mackenzie Basin (Extended  
78 Data Table 1).  $F_{\text{mod}}$  values range between 0.28 (<sup>14</sup>C age = 10,106±42 yr) and 0.63 (<sup>14</sup>C age =  
79 3675±36 yr) in the suspended load (n=27) and  $F_{\text{mod}} = 0.12$  (<sup>14</sup>C age = 17,002±84 yr) to 0.16  
80 (<sup>14</sup>C age = 14,601±64 yr) in the river bed materials (n=4). To investigate the cause of this  
81 <sup>14</sup>C-depletion, we examine the N/OC<sub>total</sub> ratio. Degradation of organic matter in soils can  
82 increase the relative N abundance<sup>6,26</sup>, differentiating degraded POC<sub>biosphere</sub> (high N/OC<sub>total</sub>)  
83 from young, fresh POC<sub>biosphere</sub> (low N/OC<sub>total</sub>). Suspended load samples display a negative  
84 relationship between N/OC<sub>total</sub> and  $F_{\text{mod}}$  (Fig. 1), similar to measurements from a peat core in  
85 the Mackenzie Basin<sup>26</sup> away from permafrost. There, N/OC<sub>total</sub> ratios increased with <sup>14</sup>C age  
86 (1,250 yr to 10,200 yr) and soil depth (0m to 3m). In contrast, river bed materials have lower  
87  $F_{\text{mod}}$  values and a relatively restricted range of N/OC<sub>total</sub> values and are distinct from  
88 suspended load (Fig. 1). A dominance of POC<sub>petro</sub> in bed materials<sup>10,14</sup> with a N/OC<sub>total</sub> ratio  
89 of ~0.07 can explain their composition.

90 Together, the  $F_{\text{mod}}$  and N/OC<sub>total</sub> values suggest that POC in the Mackenzie River is a  
91 mixture of POC<sub>petro</sub> and POC<sub>biosphere</sub>, itself varying in <sup>14</sup>C age from ‘modern’ to ~8000 yrs  
92 (Fig. 1). The  $\delta^{13}\text{C}_{\text{org}}$  values and Al/OC<sub>total</sub> ratios support this inference (Extended Data Fig. 2).  
93 Using an end member mixing analysis<sup>10,13</sup> we quantify POC<sub>petro</sub> content of sediments  
94 (Methods) and find that suspended load at the Mackenzie River Delta is dominated by  
95 POC<sub>biosphere</sub> (~70-90% of the total POC). Having corrected for POC<sub>petro</sub>, we investigate the  
96 source of POC<sub>biosphere</sub> by estimating its average <sup>14</sup>C age. This varies from 3030±150 yr to  
97 7900±400 yr (Extended Data Fig. 3) with an average <sup>14</sup>C age of POC<sub>biosphere</sub> = 5800±800 yr  
98 (±2SE) in suspended sediments of the Mackenzie River Delta. These values are older than  
99 estimates of POC<sub>biosphere</sub> age from the Amazon River (1120-2750 yr)<sup>14</sup> and Ganges River

100 (1600-2960 yr)<sup>13</sup>. The ages reflect mixing of young, fresh POC<sub>biosphere</sub> present in each of these  
101 large river basins, with an older POC<sub>biosphere</sub> in the Mackenzie Basin (Fig. 1), likely to be  
102 peats which expanded between 9,000 yr and 8,000 yr (<sup>14</sup>C-age)<sup>24</sup>. POC<sub>biosphere</sub> can be eroded  
103 by slumping and landsliding on river banks, across deep soil profiles<sup>4,7</sup>. Sections of the  
104 landscape which have discontinuous permafrost and those undergoing permafrost  
105 degradation<sup>27</sup> may be important sources of aged POC<sub>biosphere</sub>, in addition to river banks which  
106 are undercut during peak water discharge following ice-break up<sup>15</sup>. Our samples suggest that  
107 erosion and fluvial transfer of millennial-aged POC<sub>biosphere</sub> is extensive in the Mackenzie  
108 Basin.

109         Once in the river, POC<sub>biosphere</sub> is sorted with river depth, revealed by the Al/Si ratio  
110 (Fig. 2b) a proxy for grain size<sup>25</sup>. In bed materials with low Al/Si, POC<sub>petro</sub> dominates (Fig. 1)  
111 and leads to low F<sub>mod</sub> values (Fig. 2c). Just above the river bed, during the two sampling  
112 campaigns coarse suspended sediments (low Al/Si) hosted the youngest, least degraded  
113 POC<sub>biosphere</sub> (low N/C) leading to a significant contrast in <sup>14</sup>C age from the bed materials.  
114 Towards the river surface, older, more degraded POC<sub>biosphere</sub> appears to dominate, and is  
115 transported with fine sediment and clays (high Al/Si)<sup>25</sup>. The significant contribution of  
116 degraded, very old POC<sub>biosphere</sub> (>5000 yrs) contrasts with large tropical rivers where organic  
117 matter turnover in terrestrial ecosystems is more rapid (Fig. 2c)<sup>13,14</sup>.

118         To assess how erosion in the Mackenzie River may lead to long-term CO<sub>2</sub> drawdown,  
119 we estimate POC<sub>biosphere</sub> discharge. River depth profiles collected at high and falling stage  
120 suggest that the [OC<sub>total</sub>] of the suspended sediment load did not vary systematically with  
121 sediment grain size (Extended Data Fig. 4). Future work should seek to assess temporal  
122 variability in POC content and composition. Our samples suggest that changes in grain size  
123 with water discharge (Fig. 2b) could be important in setting the variability of POC<sub>biosphere</sub> age  
124 carried by the river (Fig. 2c). The [OC<sub>total</sub>] values at the Delta were 1.6±0.5% (n=8, ±1σ),

125 which were similar to the mean measured in the Mackenzie Delta in June-July 1987 of  
126  $1.4 \pm 0.2$  (n=10)<sup>12</sup>. While our sample set is modest in size, it helps us to better constrain the  
127 range of POC contents in suspended load of the Mackenzie River. In addition, our end  
128 member mixing analysis allows us to provide the first estimates of [OC<sub>biosphere</sub>], which varies  
129 between  $0.7 \pm 0.1\%$  and  $2.4 \pm 0.2\%$ . To estimate POC<sub>biosphere</sub> discharge, we use the most  
130 complete dataset of annual sediment discharge to the Mackenzie Delta (1974-1994)<sup>15</sup>, which  
131 ranged from 81 Tg yr<sup>-1</sup> to 224 Tg yr<sup>-1</sup>. A Monte Carlo approach is used to account for the  
132 modest sample size by using the full measured variability in both [OC<sub>biosphere</sub>] and sediment  
133 discharge (Methods). We estimate POC<sub>biosphere</sub> discharge =  $2.2^{+1.3}_{-0.9}$  TgC yr<sup>-1</sup> which is  
134 sustainable over  $10^3$ - $10^4$  years, depleting the soil carbon stock by  $\sim 0.006\%$  yr<sup>-1</sup> (Methods).  
135 We estimate the POC<sub>petro</sub> discharge =  $0.4^{+0.1}_{-0.1}$ . These estimates do not account for ice covered  
136 conditions when  $<10\%$  of the annual sediment discharge is conveyed<sup>12</sup>. Nevertheless, our  
137 estimate of POC<sub>biosphere</sub> discharge is greater than the combined POC discharge of  $\sim 1.9$  TgC yr<sup>-1</sup>  
138 <sup>1</sup> by the major Eurasian Arctic Rivers (Ob, Yenisei, Lena, Indigirka and Koyma)<sup>11,27</sup> which  
139 cover  $\sim 8.6 \times 10^6$  km<sup>2</sup>. Based on the available measurements, the Mackenzie River dominates  
140 the input of POC<sub>biosphere</sub> to the Arctic Ocean.

141 The mobilisation of millennial-aged POC<sub>biosphere</sub> from soils at high latitudes has been  
142 viewed as a short-term source to the atmosphere if decomposition releases greenhouse gases  
143 (CH<sub>4</sub> and CO<sub>2</sub>)<sup>2-6,8</sup>. However, if POC<sub>biosphere</sub> escapes oxidation during river transport and is  
144 buried offshore, erosion acts as a long-term CO<sub>2</sub> sink<sup>10,18,20</sup>. Offshore, aged POC<sub>biosphere</sub> from  
145 the Mackenzie River (Fig. 1) can explain the <sup>14</sup>C depletion and  $\delta^{13}$ C of bulk organic matter,  
146 and old <sup>14</sup>C ages of terrestrial plant wax compounds (up to 20,000 yr) in surface sediments of  
147 the Beaufort Sea<sup>7,28,29</sup>. We provide new evidence that terrestrial POC is buried efficiently  
148 offshore and accumulates in sediments over  $10^4$  years. Benthic foraminifera <sup>14</sup>C ages in a  
149 borehole located at the head of the Mackenzie Trough (MTW01) indicates that 21m of

150 sediment have accumulated since  $9183_{-156}^{+125}$  cal. yr BP, suggesting a high sedimentation rate  
151 during the Holocene  $2.7 \pm 0.1$  m ka<sup>-1</sup> (Extended Data Table 2, Methods). These marine  
152 sediments have [OC<sub>total</sub>] values similar to those measured in the Mackenzie River in both the  
153 <63µm (1.5% to 1.7%) and >63µm (1.1% to 1.4%) size fractions (Fig. 3). Their N/OC<sub>total</sub> and  
154  $\delta^{13}\text{C}_{\text{org}}$  values suggest that they are dominated by terrestrial POC with minor marine OC  
155 addition (Extended Data Fig. 5). We use the change in OC<sub>total</sub>/Al ratios offshore to estimate  
156 OC burial efficiencies have been  $65 \pm 27\%$  or more over the Holocene at this site (Methods).  
157 Rapid sediment accumulation and low temperature are likely to promote high POC burial  
158 efficiency<sup>18,23,28</sup>. Also, the fluvial transport dynamics of POC<sub>biosphere</sub> may promote burial (Fig.  
159 2c). The oldest, most-degraded POC<sub>biosphere</sub> is transported with clays<sup>25</sup>, whose association  
160 with organic matter may enhance burial efficiency<sup>18</sup>, while the youngest, least-degraded  
161 POC<sub>biosphere</sub> is carried near the river bed at the highest sediment concentrations. Our findings  
162 suggest that erosion and riverine transfer at high latitudes can lead to the long-term  
163 preservation of terrestrial POC in marine sediments (Fig. 3).

164 Erosion of high latitude soils and riverine export of POC<sub>biosphere</sub> may represent an  
165 important geological CO<sub>2</sub> sink. Our estimate of the modern day POC<sub>biosphere</sub> discharge of  
166  $2.2_{-0.9}^{+1.3}$  TgC yr<sup>-1</sup> in the Mackenzie River may be refined by additional temporal sampling.  
167 However, it is three times the modern rates of CO<sub>2</sub> drawdown by weathering of silicate  
168 minerals by carbonic acid in the Mackenzie River<sup>16</sup>, at  $\sim 0.7$  TgC yr<sup>-1</sup>. Preservation of POC  
169 offshore (Fig. 3) suggests that erosion of high latitude soils, riverine POC<sub>biosphere</sub> transport and  
170 export to the ocean acts as the largest geological CO<sub>2</sub> sink operating in the Mackenzie Basin.  
171 It is important to note that these longer-term fluxes are lower than estimates of greenhouse  
172 gas emissions from high latitude soils in permafrost zones due to projected warming over the  
173 coming century<sup>3,5,6,30</sup>. While these fluxes remain uncertain, recent work<sup>30</sup> has proposed  
174 emissions of  $\sim 1\text{--}2$  PgC yr<sup>-1</sup> which equate to a yield of  $\sim 70$  tC km<sup>-2</sup> yr<sup>-1</sup> over  $17.8 \times 10^8$  km<sup>2</sup> of



175 soils in permafrost zones. This estimate of accelerated release of CO<sub>2</sub> due to anthropogenic  
176 warming<sup>30</sup> is more rapid than the natural geological drawdown fluxes, of which we estimate  
177 POC<sub>biosphere</sub> ~ 2–5 tC km<sup>-2</sup> yr<sup>-1</sup> for the Mackenzie Basin (Methods). Over longer-time periods,  
178 we postulate that this geological CO<sub>2</sub> sink may be sensitive to climate conditions in the  
179 Arctic. The carbon transfer can operate when high latitudes host significant POC<sub>biosphere</sub>  
180 stocks in soil, and while rivers can erode and transfer sediments to the Arctic Ocean. Over the  
181 last 1Ma, the POC<sub>biosphere</sub> transfer was likely to have been enhanced during interglacials<sup>24</sup>  
182 (Fig. 3), whereas during glacial conditions, lower soil POC<sub>biosphere</sub> stocks and extensive ice-  
183 sheet coverage suggest that POC<sub>biosphere</sub> erosion may have been suppressed. We propose that  
184 erosion of terrestrial POC<sub>biosphere</sub> by large rivers draining the Arctic could play an important  
185 role in long-term CO<sub>2</sub> drawdown<sup>19,20</sup>, coupling the carbon cycle to climatic conditions at high  
186 latitudes.

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273 V.G. and M.D. designed the fieldwork and collected the river samples. M.O. and H.C.

274 collected sediment and carbonate data from the offshore borehole. R.G.H., V.G., M.D., C.B.  
275 and D.G. processed the samples and carried out the geochemical analyses. R.G.H. wrote the  
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## 281 **Figure Legends**

### 282 **Figure 1: Source of particulate organic carbon (POC) in the Mackenzie River Basin.**

283 Radiocarbon activity of POC ( $F_{\text{mod}}$ ) versus the nitrogen to organic carbon ratio ( $N/OC_{\text{total}}$ ) of  
284 sediments from the Mackenzie River (circles) at the delta (black), Tsiigehtchic (grey) and  
285 Norman Wells (white) and major tributaries the Liard (diamond), Peel (dark blue square) and  
286 Arctic Red (light blue square). River depth profiles collected in 2010 and 2011 suspended  
287 load (filled symbols), river bed materials (open symbols) and sieved bank samples (2009) are  
288 shown with analytical errors (2 s.d.) as grey lines if larger than points. The dashed line shows  
289 the compositions expected by mixing rock-derived, petrogenic POC ( $POC_{\text{petro}}$ ) and biospheric  
290 POC ( $POC_{\text{biosphere}}$ ). Solid green line is the trend from a peat core in western Canada<sup>26</sup>.

### 291 **Figure 2: Transport of particulate organic carbon (POC) in the Mackenzie River. a.**

292 River depth profile collection from the Mackenzie River Delta during falling stage, with  
293 Acoustic Doppler Current Profiler data to determine channel geometry, water velocity and  
294 water discharge ( $\text{m}^3 \text{s}^{-1}$ ). **b.** Aluminium to Silicon ratio ( $Al/Si$ , molar), a proxy for sediment  
295 grain size<sup>25</sup>, with water depth normalised to maximum depth. Coarser materials are carried  
296 throughout the profile during high stage. **c.** Radiocarbon activity of POC ( $F_{\text{mod}}$ ) versus  $Al/Si$   
297 for the Mackenzie Basin (this study, symbols as Fig. 1), Amazon River<sup>14</sup>, and Ganges

298 River<sup>10,13</sup>. River suspended load (filled) and river bed materials (open) are distinguished with  
299 analytical errors (2 s.d.) shown as grey lines if larger than points.

300 **Figure 3: Fate of particulate organic carbon offshore.** **a.** Organic carbon concentration  
301 ( $[OC_{total}]$ , %) of suspended sediments in the Mackenzie River Delta (n=8) where solid line  
302 and grey box show the mean  $\pm$  standard error, whiskers show  $\pm$  standard deviation and the  
303 circles indicate the minimum and maximum values. **b.**  $[OC]_{total}$  in sediments  $<63\mu m$  and  
304  $>63\mu m$  from core MTW01 in the Mackenzie Trough (Extended Data Fig. 1) for depths dated  
305 by the <sup>14</sup>C activity of mixed benthic foraminifera (Methods), where whiskers show analytical  
306 error if large than the point size.

## 307 **Methods**

308 **River sample collection and preparation:** River depth-profiles from September 2010 and  
309 June 2011 (Extended Data Table 1) were used to collect the full range of erosion products  
310 and POC in large river systems, taking advantage of the hydrodynamic sorting of  
311 particles<sup>10,13,14,25</sup>. At each sampling site (Fig. 1) channel depth, water velocity and  
312 instantaneous water discharge were measured by two or more transects with an Acoustic  
313 Doppler Current Profiler (ADCP Rio Grande 600 kHz) before each depth profile was  
314 collected at a single point ( $\pm 10m$ ) in the middle of the channel. On the boat, each sample (~7-  
315 8L) was evacuated into a clean bucket and stored in sterilised plastic bags and the procedure  
316 repeated depending upon the total water depth. Each bag was weighed to determine the  
317 sampled volume, then the entire sample was filtered within 24 hrs through pre-cleaned Teflon  
318 filter units through 90 mm diameter 0.2 $\mu m$  PES filters<sup>13,25</sup>. Suspended sediment was  
319 immediately rinsed from the filter using filtered river water into clean amber-glass vials and  
320 kept cool. River bed materials were collected at the base of the depth transects from the boat,  
321 using a metal bucket as a dredge, and decanted to a sterile bag. River bank deposits (June

322 2009) were collected from fresh deposits close to the channel (Extended Data Table 3) and  
323 sieved at 250  $\mu\text{m}$  and 63  $\mu\text{m}$  to investigate the sorting of POC<sup>31</sup>. All sediments were freeze  
324 dried upon return to laboratories within 2 weeks, weighed and homogenised in an agate  
325 grinder.

326 **Offshore borehole sampling and benthic foraminifera sample preparation:** Benthic  
327 foraminifera-containing marine sediment samples were obtained from the upper 22 m  
328 Holocene sequence of an 85.1 m MTW01 borehole<sup>32</sup> located at 69° 20' 53" N, 137° 59' 13"  
329 in 45 m water depth in the Mackenzie Trough (Extended Data Fig. 1). Drilled by the  
330 Geological Survey of Canada in 1984, the core is currently archived at the GSC-Atlantic core  
331 repository. To isolate foraminifera, sediment samples were disaggregated over a <38 $\mu\text{m}$  mesh  
332 sieve using deionized water. Based on down-hole microfossils counts, 4 samples were  
333 selected with sufficient specimens for radiocarbon dating.

334 **Geochemical analyses:** For the river suspended sediments and core samples for organic  
335 carbon analyses, inorganic carbon was removed using a HCl fumigation technique to avoid  
336 loss of a component of POC which is known to occur during a HCl leach<sup>33</sup>. An adapted  
337 method to ensure full removal of detrital dolomite was used<sup>34</sup>. In summary, samples were  
338 placed in an evacuated desiccator containing ~50 ml 12N HCl in an oven at between 60 and  
339 65 °C for 60 to 72 hours. Sample were then transferred to another vacuum desiccator charged  
340 with indicating silica gel, pumped down again and dried to remove HCl fumes. River  
341 sediment samples were analysed for organic carbon concentration [ $\text{OC}_{\text{total}}$ ] on acidified  
342 aliquots and nitrogen concentration ([N], %) on non-acidified aliquots by combustion at  
343 1020°C in O<sub>2</sub> using a Costech elemental analyser (EA) in Durham. For river depth profile  
344 samples, acidified aliquots were prepared to graphite at the NERC Radiocarbon Facility of  
345 between 1-2 mg C for each sample and standard and <sup>14</sup>C was measured by Accelerator Mass  
346 Spectrometry at the Scottish Universities Environmental Research Centre and reported as

347 fraction modern ( $F_{\text{mod}}$ ) by standard protocol<sup>35</sup>. Process standards (96H humin) and  
348 background materials (bituminous coal) were taken through all stages of sample preparation  
349 and  $^{14}\text{C}$  analysis and were within  $2\sigma$  uncertainty of expected values. Stable isotopes of POC  
350 ( $\delta^{13}\text{C}_{\text{org}}$ ) were measured by dual-inlet isotope ratio mass spectrometer (IRMS) on an aliquot  
351 of the same  $\text{CO}_2$ . These measurements were consistent with  $\delta^{13}\text{C}_{\text{org}}$  measurements made by  
352 EA-IRMS normalised based on measured values standards ( $n=7$ ) spanning  $>30\text{‰}$  and long-  
353 term analytical precision of  $0.2\text{‰}$ . River bank samples from 2009 were analysed by similar  
354 procedures at the National Ocean Sciences Accelerator Mass Spectrometry Facility  
355 (NOSAMS) at Woods Hole Oceanographic Institution.

356 Mixed benthic foraminifera samples picked from the MTW01 core were analysed at  
357 NOSAMS for  $^{14}\text{C}$  analyses. Samples were rinsed and no pre-treatments were used. The  
358 samples were directly hydrolyzed with strong acid,  $\text{H}_3\text{PO}_4$ , to convert the carbon in the  
359 sample to  $\text{CO}_2$ . Calibration of the  $^{14}\text{C}$  dates was performed using CALIB (version 7.1)<sup>36</sup>. All  
360  $^{14}\text{C}$  dates were normalized to a  $\delta^{13}\text{C}$  of  $-25\text{‰}$  versus VPDB (<http://intcal.qub.ac.uk/calib/>).  
361 Foraminifera dates were calibrated using the MARINE13 dataset<sup>37</sup>, with a reservoir age  
362 correction ( $\Delta R$ ) of  $335\pm 85$  yrs (Extended Data Table 2). The  $\Delta R$  value is based on a recent  
363 re-analysis of ages from 24 living molluscs collected before 1956 from the northwestern  
364 Canadian Arctic Archipelago<sup>38</sup>. This calibration set does not include specimens from the  
365 Beaufort Sea and as such provides only a best available estimate for  $\Delta R$  in the Mackenzie  
366 Trough.

367 **End member mixing model:** The  $F_{\text{mod}}$ ,  $\text{N}/\text{OC}_{\text{total}}$  (Fig. 1),  $\delta^{13}\text{C}_{\text{org}}$  values and  $\text{Al}/\text{OC}_{\text{total}}$   
368 values (Extended Data Fig. 2) are consistent with a mixing of  $\text{POC}_{\text{petro}}$  and  $\text{POC}_{\text{biosphere}}$   
369 dominating the bulk geochemical composition of river POC. Autochthonous sources are not  
370 significant based on those measured values, which is consistent with the turbid nature of the  
371 Mackenzie River (mean suspended sediment concentration of  $\sim 300\text{-}400$  mg/L) meaning that



372 like other turbid river systems (e.g. the Ganges-Brahmaputra) light penetration is minimal. A  
373 mixture of  $\text{POC}_{\text{petro}}$  and  $\text{POC}_{\text{biosphere}}$  can be described by governing equations<sup>10,13,31</sup>:

374  $f_{\text{biosphere}} + f_{\text{petro}} = 1$  (Equation 1)

375  $f_{\text{biosphere}} \times \theta_{\text{biosphere}} + f_{\text{petro}} \times \theta_{\text{petro}} = \theta_{\text{sample}}$  (Equation 2)

376 where  $f_{\text{biosphere}}$  and  $f_{\text{petro}}$  are the fractions of POC derived from biospheric and petrogenic  
377 sources, respectively.  $\theta_{\text{sample}}$  is the measured composition (e.g.  $F_{\text{mod}}$ ) of a river POC sample,  
378 and  $\theta_{\text{biosphere}}$  and  $\theta_{\text{petro}}$  are the compositions of biospheric and petrogenic sources. To quantify  
379 the  $f_{\text{petro}}$  in each sample we use the aluminium (Al) to  $\text{OC}_{\text{total}}$  concentration ratio in river  
380 sediments. At each locality, a linear trend between  $F_{\text{mod}}$  and  $\text{Al}/\text{OC}_{\text{total}}$  (Extended Data Fig.  
381 2b) can be explained by a mixture of an Al-rich, OC-poor material (rock fragments  
382 containing  $\text{POC}_{\text{petro}}$ ) with Al-poor, OC-rich material (soils and vegetation debris as  
383  $\text{POC}_{\text{biosphere}}$ ). Taking advantage of the fact that the  $\text{POC}_{\text{petro}}$  has  $F_{\text{mod}} \sim 0$ , the intercept at  $F_{\text{mod}} =$   
384 0 gives an estimate of the  $\text{Al}/\text{OC}_{\text{total}}$  values and associated uncertainty of the sedimentary  
385 rock end member. To estimate the average concentration of  $\text{OC}_{\text{petro}}$  of bedrocks in each basin  
386 ( $[\text{OC}_{\text{petro}}]$ , %), we use the Al concentration of river bed materials as a proxy of for the Al  
387 concentration in the bedrocks<sup>25</sup> and the  $\text{Al}/\text{OC}_{\text{total}}$  value at  $F_{\text{mod}} \sim 0$ . Following previous work  
388 in large rivers, we then assume that the  $\text{OC}_{\text{petro}}$  is well mixed in the water column and has a  
389 relatively constant  $[\text{OC}_{\text{petro}}]$  (refs 10,13,14). This method may overestimate  $f_{\text{petro}}$  if  $\text{OC}_{\text{petro}}$  has  
390 been more extensively oxidised in fine grained weathering products carried in the suspended  
391 load<sup>21</sup>.  $f_{\text{petro}}$  is quantified using  $[\text{OC}_{\text{petro}}]$  and measured  $[\text{OC}_{\text{total}}]$ .

392 The mixing analysis returns a  $[\text{OC}_{\text{petro}}] = 0.12 \pm 0.03\%$  ( $\pm 2\sigma$ ) in the Liard River and  
393 Mackenzie River at Tsiigehtchic, higher values in the Peel River  $[\text{OC}_{\text{petro}}] = 0.63 \pm 0.30\%$ ,  
394 with the Mackenzie River at the delta with an intermediate value  $[\text{OC}_{\text{petro}}] = 0.29 \pm 0.05\%$ .  
395 This is consistent with the known presence of  $\text{POC}_{\text{petro}}$ -bearing sedimentary rocks in the

396 Mackenzie River Basin and high OC<sub>total</sub> contents of bedrocks in the upper Peel River Basin  
397 and Mackenzie mountains<sup>39</sup>. To quantify the average <sup>14</sup>C age of POC<sub>biosphere</sub> in each sample,  
398 Equations 1 and 2 can be solved for  $\theta_{biosphere}$ , using the  $f_{petro}$  and assumed unmeasurable above  
399 background <sup>14</sup>C content of POC<sub>petro</sub> ( $F_{mod} = 0$ ). The uncertainty mainly derives from that on  
400  $f_{petro}$  and [OC<sub>total</sub>] and has been propagated through the calculations.

401 To test if the mixing of POC<sub>biosphere</sub> and POC<sub>petro</sub> can describe the composition of the  
402 suspended load samples, we predict the  $\delta^{13}C_{org}$  measurements which were not used in the  
403 mixing analysis. The calculated  $f_{petro}$  values and end member values of  $-26.2 \pm 0.5\text{‰}$  for  
404 POC<sub>biosphere</sub> and  $-28.6 \pm 0.5\text{‰}$  for POC<sub>petro</sub> were used, informed by measurements of  
405 bedrocks<sup>39</sup> and vegetation and soil in the basin<sup>40</sup>. The mixing model (equation 2) can robustly  
406 predict the  $\delta^{13}C_{org}$  differences between the Peel and Liard rivers, and between suspended load  
407 and bed material  $\delta^{13}C_{org}$  values (Extended Data Fig. 2c), supporting a mixing control on the  
408 variables.

409 **Mackenzie River POC discharge:** To quantify the discharge of POC we need to account for  
410 the variability in suspended sediment discharge and the variability in the POC<sub>biosphere</sub> and  
411 POC<sub>petro</sub> content of sediments in the basin. We use the longest, most complete quantification  
412 of sediment flux by the Mackenzie River from 1974-1994<sup>15</sup>, which has an average  $127 \pm 40 \text{ Tg}$   
413  $\text{yr}^{-1}$  ( $\pm 1\sigma$ ). Annual sediment yield varied from  $81 \text{ Tg yr}^{-1}$  to  $224 \text{ Tg yr}^{-1}$ . While the POC  
414 samples were not collected at the same time period, our measurements of [OC<sub>total</sub>] at the  
415 Delta, mean =  $1.6 \pm 0.5\%$  ( $n=8$ ,  $\pm 1\sigma$ ) and do not vary systematically between falling and high  
416 stage (Extended Data Fig. 4) and are consistent with available data from samples<sup>12</sup> collected  
417 in 1987 ( $1.4 \pm 0.2$ ,  $n=10$ ). While future work should aim to constrain the variability in POC  
418 composition further, these observations suggest that temporal variability may be less  
419 important than the potential variability in [OC<sub>total</sub>] with depth at a given time, where we find  
420 [OC<sub>total</sub>] values can range from 1.0% to 2.7%. We use our measured range of [OC<sub>biosphere</sub>] and

421 [OC<sub>petro</sub>] values and the full range of annual sediment yields<sup>15</sup> to quantify POC<sub>biosphere</sub> and  
422 POC<sub>petro</sub> discharge and associated uncertainty using a Monte Carlo approach. Over 100,000  
423 simulations, we use a ‘flat’ probability for the range of values for both variables (i.e. equal  
424 probability of all measured values). This allows us to fully explore the range of estimates  
425 given the available measurements. Future work seeking to expand the number of [OC<sub>biosphere</sub>]  
426 measurements to assess its flux-weighted mean and variability, while assessing temporal  
427 variability in more detail, will allow POC discharge estimates and their uncertainty to be  
428 refined. POC<sub>biosphere</sub> ( $2.2^{+1.3}_{-0.9}$  TgC yr<sup>-1</sup>) and POC<sub>petro</sub> ( $0.4^{+0.1}_{-0.1}$ ) discharges are reported as the  
429 median (50%) ± 1 s.d. Over the sediment source areas of the Mackenzie (Downstream of the  
430 Great Slave Lake<sup>15</sup>) of 774,200 km<sup>2</sup>, these equate to yields of POC<sub>biosphere</sub> =  $2.9^{+1.7}_{-1.1}$  tC km<sup>-2</sup>  
431 yr<sup>-1</sup> and POC<sub>petro</sub> =  $0.6^{+0.2}_{-0.2}$  tC km<sup>-2</sup> yr<sup>-1</sup>. The total POC discharge is slightly higher than a  
432 previous estimate (2.1 TgC yr<sup>-1</sup>)<sup>12</sup> based on measurements of POC content made in 1987  
433 because we: i) account for higher POC<sub>biosphere</sub> concentrations which may occur in water-  
434 logged POC<sub>biosphere</sub> near the river bed (Figs. 2c, Extended Data Fig. 4); and ii) account for the  
435 potential for very high annual sediment discharge<sup>15</sup>. Based on estimates of soil carbon stock  
436 in the Mackenzie Basin<sup>2</sup> of  $\sim 50 \times 10^3$  tC km<sup>-2</sup> and the upstream sediment source area  
437 (downstream of the Great Slave Lake, 774,200 km<sup>2</sup>), the present rate of POC<sub>biosphere</sub> export  
438 represents a depletion of the soil carbon stock by  $\sim 0.006\%$  yr<sup>-1</sup>, which is sustainable over 10<sup>3</sup>-  
439 10<sup>4</sup> years.

440 **OC burial efficiency in MTW01:** To estimate the burial efficiency of terrestrial POC at the  
441 MTW01 site, we normalise the measured [OC<sub>total</sub>] concentrations (Fig. 3) by Al  
442 concentration, an immobile inorganic element hosted by major mineral phases. The OC<sub>total</sub>/Al  
443 normalization allows the effects of dilution to be distinguished from net OC gain (increased  
444 ratio) or OC loss (decreased ratio). The mean OC<sub>total</sub>/Al of the MTW01 samples was  
445  $0.17 \pm 0.02$  (g g<sup>-1</sup>, n=4, ±2SE). This is lower than the mean OC<sub>total</sub>/Al of the suspended load

446 samples from the Mackenzie River delta of  $0.26 \pm 0.10$  ( $\text{g g}^{-1}$ ,  $n=8$ ,  $\pm 2\text{SE}$ ). The decrease in the  
447 ratio offshore may suggest a higher relative proportion of  $\text{POC}_{\text{petro}}$  (Extended Data Fig. 2b),  
448 however this is not consistent with the less negative  $\delta^{13}\text{C}_{\text{org}}$  values (Extended Data Fig. 5).  
449 The decrease can therefore be interpreted in terms of OC loss, with the ratio of core to river  
450 samples ( $0.17 \pm 0.02 / 0.26 \pm 0.10$ ). Assuming that all the change in  $\text{OC}_{\text{total}}/\text{Al}$  is driven by OC  
451 loss, and taking into account the measurement variability in these values, we estimate that  
452  $65 \pm 27\%$  of the OC has been preserved. However, we note that the  $\text{OC}_{\text{total}}/\text{Al}$  ratios in the core  
453 are not statistically different from the river suspended load samples (one-way ANOVA,  
454  $P > 0.1$ ) which suggest the OC burial efficiency could be higher (i.e. 100%). In addition, if we  
455 use the  $\text{OC}_{\text{total}}/\text{Al}$  of finer river sediments carried near the channel surface which may be  
456 more easily conveyed offshore of  $0.20 \pm 0.04$  ( $\text{g g}^{-1}$ ,  $n=4$ ,  $\pm 2\text{SE}$ ), we calculate burial efficiency  
457 =  $85 \pm 20\%$ . Future work should seek to better constrain these burial efficiencies with  
458 additional terrestrial and marine samples. Nevertheless, despite the remaining uncertainty,  
459 these high burial efficiencies<sup>18</sup> are consistent with the high sedimentation rate and low  
460 temperature setting. The long-term burial of POC delivered to sites deeper in the Beaufort  
461 Sea<sup>28</sup> still remains to be assessed, in order to provide a complete picture of source to sink  
462 carbon transfers.

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## 491 **Extended Data**

492 Available by contacting the lead author ([r.g.hilton@durham.ac.uk](mailto:r.g.hilton@durham.ac.uk)) or on the online version of  
493 the paper: doi:10.1038/nature14653.

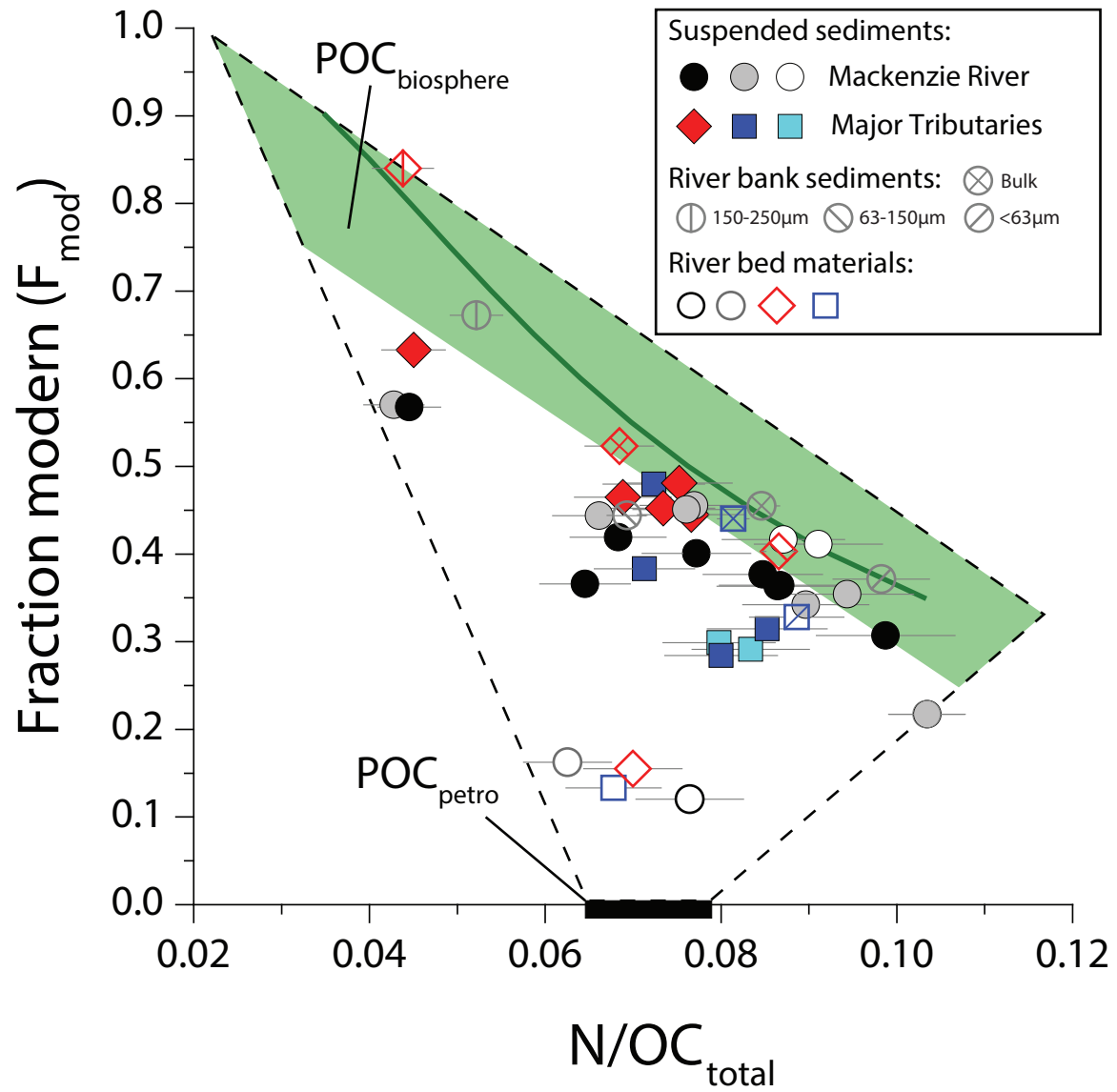


Figure 1

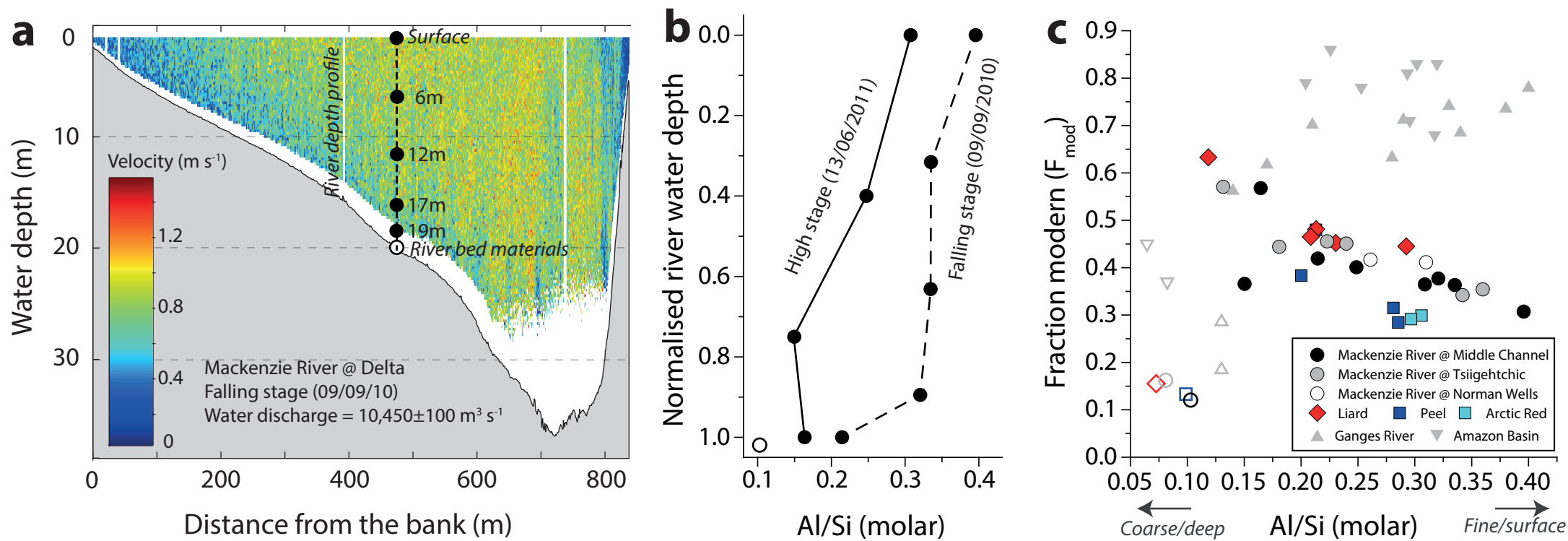


Figure 2

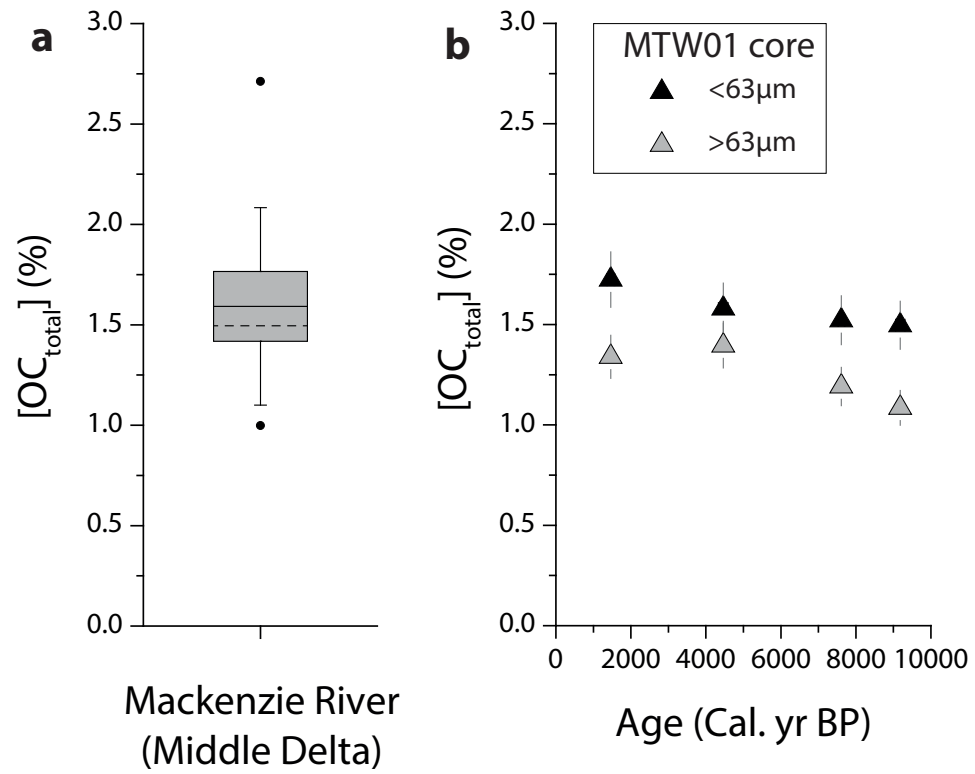


Figure 3