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## 1 Channel cross-section heterogeneity of particulate organic carbon 2 transport in the Huanghe

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12 **Abstract.** The Huanghe (Yellow River), one of the largest turbid river systems in the world, has long been recognized  
13 as a major contributor of suspended particulate matter (SPM) to the ocean. However, over the last few decades, the  
14 SPM export flux of the Huanghe has decreased over 90% due to the high management, impacting the global export of  
15 particulate organic carbon (POC). To better constrain sources and modes of transport of POC beyond the previously  
16 investigated transportation of POC near the channel surface, SPM samples were for the first time collected over a  
17 whole channel cross-section in the lower Huanghe. Riverine SPM samples were analyzed for particle size and major  
18 element contents, as well as for POC content and dual carbon isotopes (<sup>13</sup>C and <sup>14</sup>C). The results show clear vertical  
19 and lateral heterogeneity of SPM physical and chemical characteristics within the river cross-section, with for example  
20 finer SPM carrying more POC with higher <sup>14</sup>C activity near the surface of the right bank. Notably, we discuss how  
21 bank erosion in the alluvial plain is likely to generate lateral heterogeneity in POC composition. The Huanghe POC  
22 is millennial-aged (4,020 ± 500 radiocarbon years), dominated by organic carbon (OC) from the biosphere, while the  
23 lithospheric fraction reaches up to ca. 33%. The mobilization of aged and refractory OC, including radiocarbon-dead  
24 biospheric OC, from deeper soil horizons of the loess-paleosol sequence through erosion in the Chinese Loess Plateau  
25 is an important mechanism contributing to fluvial POC in the Huanghe drainage basin. Altogether, anthropogenic  
26 activities can drastically change the compositions and transport dynamics of fluvial POC, consequentially altering the  
27 feedback of the source-to-sink trajectory of a river system to regional and global carbon cycles.



## 28 **1 Introduction**

29 Rivers are the main conveyor of rock and soil debris eroded from the continents to the ocean. Along with inorganic  
30 material, river sediments host particulate organic carbon (POC) derived mainly from three major sources: 1) recently  
31 photosynthesized OC of the biosphere, 2) aged and altered OC from soils, and 3) ancient OC contained in sedimentary  
32 rocks (Blair et al., 2010). The net effect of riverine POC transport on the carbon cycle and thus on the evolution of  
33 Earth's climate depends on POC provenance and fate. The effective sedimentary burial of POC derived from the  
34 terrestrial biosphere (biospheric OC,  $OC_{bio}$ ) represents a net, long-term sink of atmospheric  $CO_2$  (Galy et al., 2007,  
35 Bouchez et al., 2014; Hilton et al., 2015), whereas the oxidation of POC derived from continental rocks (petrogenic  
36 OC,  $OC_{petro}$ ) acts as a net, long-term source of  $CO_2$  to the atmosphere (Hilton et al., 2014). The erosion and burial of  
37  $OC_{petro}$  escaping from oxidation has no net effect on the long-term carbon cycle (Galy et al., 2008a; Bouchez et al.,  
38 2010; Hilton et al., 2011; Horan et al., 2019). In addition, the reactive nature of  $OC_{bio}$  might also result in short-term  
39  $CO_2$  emission during transport from both river channels and recently-deposited sediments (Mayorga et al., 2005; Galy  
40 and Eglinton, 2011; Blair and Aller, 2012).

41 Globally, rivers transport a total POC flux of *ca.* 200 Tg C/year, consisting of  $157^{+74}_{-50}$  Tg C/year of  $OC_{bio}$  and  $43^{+61}_{-25}$   
42 Tg C/year of  $OC_{petro}$  (Galy et al., 2015; Ludwig et al., 1996). Source-to-sink processes controlling the origin and fate  
43 of riverine POC are prominently river-specific, suggesting that the impact of POC on regional and global carbon  
44 cycles might significantly vary both spatially and temporally (Blair and Aller, 2012). It is thus crucial to understand  
45 the mechanisms controlling the POC export by large rivers that integrate vast portions of the land surface, and quantify  
46 the differing sources of carbon exported by those large river systems.

47 The Huanghe (Yellow River) is a highly turbid river system that exports over 85% of its OC as particulate matter,  
48 with efficient deposition and preservation in the ocean (Cauwet and Mackenzie, 1993; Bianchi, 2011; Zhang et al.,  
49 2013; Ran et al., 2013). The Huanghe has been highly managed over the last few decades through water and soil  
50 conservation measures as well as reservoir construction, leading to a decrease of nearly 90% of its sediment load  
51 (Wang and Fu et al., 2016; Wang et al., 2007; Milliman et al., 1987) and a significant decrease in its POC delivery to  
52 the ocean (Zhang et al., 2013). Reservoir construction dramatically affects the transport and fate of both sediment load  
53 and POC in large rivers (Syvitski et al., 2005; Li et al., 2015). The estimated POC flux of the Huanghe is thought to  
54 have shifted from 4.5 Tg C/yr in the 1980s (Cauwet and Mackenzie, 1993) to 0.34-0.58 Tg C/yr nowadays (Tao et al.,  
55 2018) in response to both anthropogenic influence (Hu et al., 2015; Tao et al., 2018; Yu et al., 2019a) and natural  
56 variability of the regional hydrological cycle (Qu et al., 2020). These large-scale perturbations have likely modified  
57 the OC input from the different terrestrial pools as well as the fate of exported POC that was previously reaching  
58 deposition centers in the ocean and that now remains stuck on land. Those alterations of the carbon cycle remain to  
59 be addressed.

60 Over the last decade, POC transport in the Huanghe has been investigated for 1) determination and quantification of  
61 POC sources, based on bulk or molecular carbon isotopic composition (Tao et al., 2015; Yu et al., 2019b; Ge et al.,  
62 2020; Qu et al., 2020); 2) temporal and spatial variations in POC export and distribution among different size fractions  
63 (Ran et al., 2013; Wang 2012; 2016; Yu et al., 2019a, b, Qu et al., 2020); 3) impact of anthropogenic activities (Hu et



64 al., 2015; Tao et al., 2018; Yu et al., 2019a); and 4) burial efficiency and preservation in the ocean (Sun et al., 2018;  
65 Tao et al., 2016; Ge et al., 2020). However, all these previous studies rely on suspended sediment samples collected  
66 near the channel surface or at a single, intermediate depth in the river channel, further assuming a homogeneous  
67 distribution of suspended sediment characteristics in the water column, both vertically and laterally. It is now well  
68 recognized that suspended sediments present physical, mineralogical, chemical, and isotopic heterogeneities across  
69 river cross-sections due to hydrodynamic sorting and tributary mixing (Galy et al., 2008b; Garzanti et al., 2010;  
70 Bouchez et al., 2010, 2011a). This is also true for POC, whose age and composition vary within cross-sections (*e.g.*,  
71 Bouchez et al., 2014; Baronas et al., 2020) and between sediment size fractions separated in the laboratory (Yu et al.,  
72 2019b; Ge et al., 2020). Such heterogeneity warrants a re-evaluation of POC transport in the Huanghe, accounting for  
73 the variability in suspended sediment characteristics over the channel cross-section.

74 In this study, we take advantage of in-river hydrodynamic sorting to access the full range of suspended sediment size  
75 fractions by collecting suspended particulate matter (SPM) samples along several river depth profiles distributed  
76 across a channel transect (*e.g.*, Bouchez et al., 2014; Freymond et al., 2018; Baronas et al., 2020). We apply this  
77 sampling scheme to a cross-section of the Huanghe located 200 km upstream from the river mouth and report SPM  
78 OC content, stable isotope composition, and radiocarbon activity as well as total nitrogen, major element composition  
79 (aluminum and silicon), and particle size distribution. Based on these novel samples and data sets, this study aims at  
80 1) determining the controls on POC content in the Huanghe; 2) tracing and quantifying the sources of riverine POC  
81 in the Huanghe; and 3) providing depth-integrated estimates of POC fluxes in the most turbid large river system.

## 82 **2 Study area**

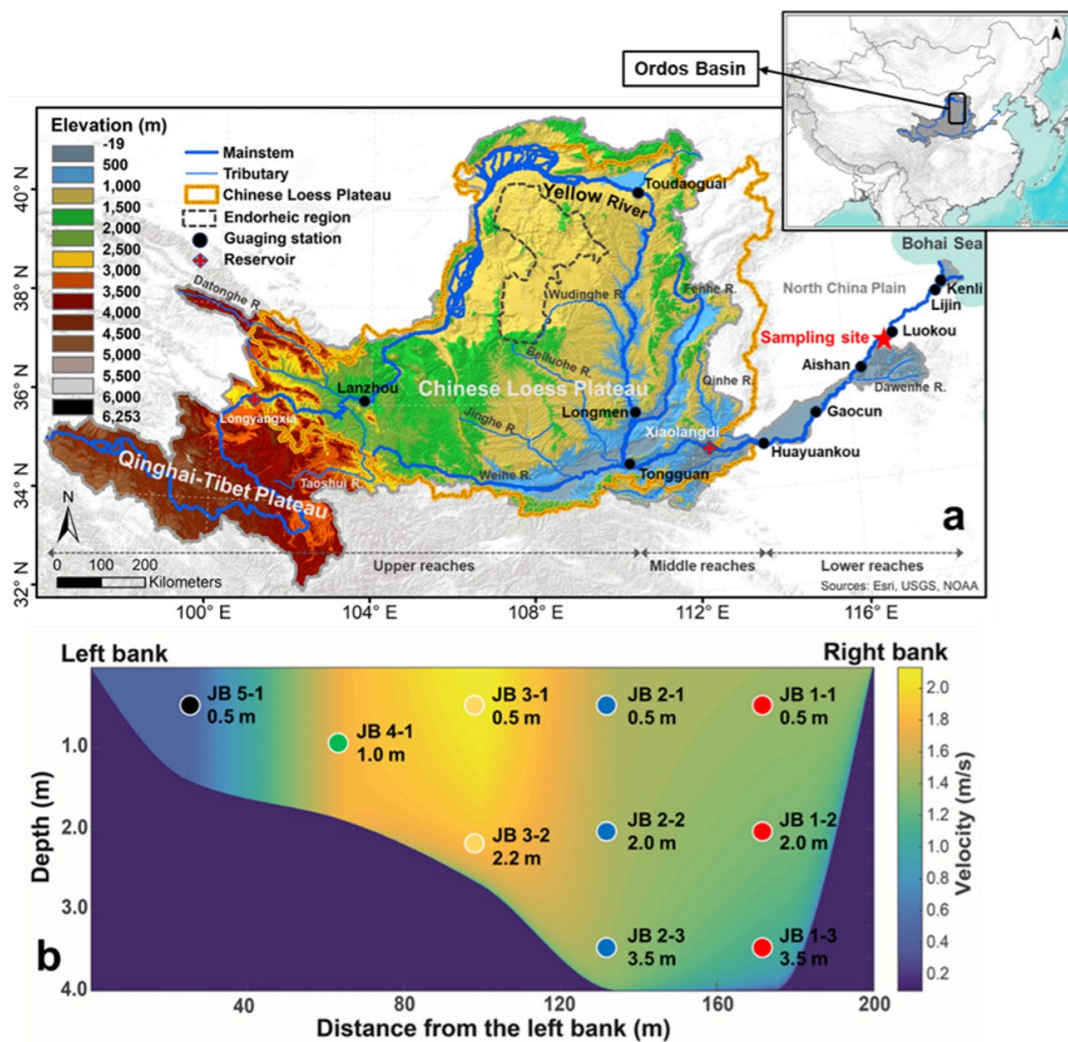
83 The Huanghe originates from the north-eastern Qinghai-Tibet Plateau (QTP) and runs through the Chinese Loess  
84 Plateau (CLP) and the North China Plain (NCP) to the Bohai Sea (Figure 1a). It is 5,464 km long and drains a basin  
85 area of  $79.5 \times 10^4$  km<sup>2</sup>. The Huanghe drainage basin can be subdivided into three main geomorphic units: 1) the high-  
86 relief upper reaches spanning from the source region (elevation of 4,500 m) to the city of Toudaoguai (located 3,472  
87 km downstream at an elevation of 1,000 m); 2) the middle reaches with a channel length of 1,206 km, ending at  
88 Huayankou (elevation of 110 m) draining landscapes characterized by relatively gentle slopes; and 3) the lower  
89 reaches where the river flows eastwards across a fluvial plain over a length of 786 km. These three sections drain  
90 53.8%, 43.3%, and 2.9% of the whole Huanghe basin area, respectively (Wang et al., 2007; YRCC, 2016). Most  
91 second-order tributaries drain the CLP region and feed the main channel in the middle reaches, the Dawenhe River  
92 being the only tributary of the lower reaches, with negligible water and sediment supply due to upstream trapping in  
93 lakes and reservoirs. It is worth noting that more than 50% of the water discharge at the Huanghe's mouth comes from  
94 the QTP, whereas over 90% of the sediment load originates from the CLP (Wang et al., 2010, 2017; Pan et al., 2016).  
95 The CLP is thus the principal source area of sediment to the Huanghe (Shi and Shao, 2000; Guo et al., 2002; Wang  
96 and Fu et al., 2016).

97 The Huanghe drainage basin is mostly underlain by the North China craton, and is bounded by several mountain belts.  
98 The watershed encompasses 46% of sedimentary rock outcrops (mainly siliciclastic rocks with minor carbonates), and  
99 about 45% of unconsolidated sediments (mainly Quaternary loess deposits). The remaining outcrops include



100 metamorphic, plutonic, and volcanic rocks formed from the Archean to the Tertiary (Figure S1). Although river  
101 incision is strong in the QTP, a substantial part of the corresponding eroded material is not effectively transferred to  
102 the lower reaches due to deposition in the CLP and the western Mu-Us desert, a situation that has prevailed since at  
103 least the middle Pleistocene (Nie et al., 2015; Licht et al., 2016; Pan et al., 2016). In addition, recent anthropogenic  
104 disturbance such as constructions of large dams in the upper reaches has profoundly modified the export of solid  
105 materials from the basin (Wang et al., 2007). The Huanghe then flows through the CLP that has acted as the major  
106 supplier of sediment to the system since at least the Calabrian Pleistocene (Stevens et al., 2013; Bird et al., 2015).  
107 There, an easily erodible loess-paleosol formation has accumulated since 2.58 Ma (Guo et al., 2002), over a thickness  
108 ranging from a few meters to more than 500 m, with an average of 100 m. This loess-paleosol formation and underlying  
109 Cretaceous sedimentary rocks are actively incised by the main stem and its tributaries (Shi and Shao, 2000; Guo et  
110 al., 2002; Wang and Fu et al., 2016). Notably, the Ordos Basin underlying the CLP is rich in oil and gas (Guo et al.,  
111 2014). In the lower reaches, the river drains Quaternary fluvial deposits and sedimentary rocks.

112 The Huanghe drainage basin encompasses the entire arid and semi-arid region of northern China in the upper and  
113 middle reaches, and is characterized by more humid climate conditions in the lower reaches. Annual average  
114 precipitation (over the period 1950 - 2000) in the upper, middle and lower reaches regions is 368 mm, 530 mm, and  
115 670 mm, respectively (Wang et al., 2007). As a result of the East Asian summer and winter monsoon circulations, the  
116 rainy season (June to September) contributes 85% of the annual precipitation (Wang et al., 2007). During the rainy  
117 season, frequent storm events lead to concentrated flows (relatively high discharge) in vulnerable gully-hill systems,  
118 the dominant regional geomorphic landscape, and actively participates in soil erosion in the CLP (Shi and Shao, 2000;  
119 He et al., 2004; Qu et al., 2020). The present-day (2002 to 2016) suspended sediment flux delivered by the Huanghe  
120 to the sea is about 0.12 Gt/yr, which implies a decrease of nearly 90% in sediment export compared to the widely cited  
121 estimate of 1.08 Gt/yr (average value between 1950 to 1980, Milliman and Farnsworth, 2011). This massive decrease  
122 in sediment export mostly results from human perturbations, including soil conservation practices in the CLP and  
123 retention in large reservoirs, rather than from climatic variations such as the decreasing precipitation observed in the  
124 region over the last decades (Wang et al., 2007; Ran et al., 2013; Wang and Fu et al., 2016). A scheme for water and  
125 sediment regulation (WSR) has been implemented through the construction of the Xiaolangdi Reservoir since 2002,  
126 aiming to mitigate water and sediment imbalances in the lower reaches. This regulation has resulted in a modification  
127 of the flux of sediment delivered to the lower reaches and estuary, making the Huanghe a highly human-regulated  
128 river system. However, no WSR was implemented in 2016, the year of our sampling campaign, suggesting that the  
129 collected SPM samples are not significantly affected by retention in dams, and thus are representative of the fluvial  
130 transport of terrestrial materials eroded from the CLP.



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Figure 1: (a) Elevation map of the Huanghe drainage basin showing the main reservoirs and gauging stations along the main stem as well as our sampling site (36.75°N, 117.02°E, near the Luokou gauging station); and (b) channel cross-section of suspended particulate matter (SPM) samples and modeled velocity distribution based on the "law of the wall", using the point velocity data measured by a current velocity meter attached to the sampler.

### 137 3 Sampling and analytical methods

#### 138 3.1 Sampling strategy

139 Detailed sampling of a cross-section of the Huanghe was carried out on the 17<sup>th</sup> of July, 2016, during the flood season  
140 (Figure S2). Samples were collected along five depth profiles near the Luokou hydrological station, 250 km upstream  
141 from the river mouth (Figure 1). This sampling strategy allows for accessing the full range of suspended sediment  
142 particle size (Bouchez et al., 2014). The cross-section is 200-meters wide at the surface and 4 meters deep at most





143 (Figure 1). As in previous studies, we used a home-made, 10-liter, point-sediment horizontal Niskin-type sampler  
144 attached to a current velocity meter, to collect river water samples and measure the water velocity simultaneously.  
145 Subsequently, two water samples were collected at the surface near the right bank in May and June 2017 before the  
146 flooding season, to retrieve fine suspended particulate matter. For each sample, approximately 30 liters of river water  
147 were collected and were then filtered through pre-weighed 0.22- $\mu\text{m}$  porosity cellulose acetate membrane filters within  
148 24 hours. After rinsing the filters with filtered water, all sediment samples were transferred into centrifuge tubes and  
149 freeze-dried before weighing and analysis. A bed sediment sample was collected on an exposed, recently flooded  
150 sediment bar of the riverbed.

### 151 3.2 Physical and geochemical analysis

152 Apart from a 50-mg aliquot of SPM samples preserved for particle size analysis, samples were finely ground using an  
153 agate mortar and pestle prior to chemical and isotopic analyses. The particle size distribution of the unground aliquots  
154 was measured using a Laser Diffraction Particle Size Analyzer (Beckman Coulter LS-12 320) at the École Normale  
155 Supérieure (ENS), Paris, France. Before analysis, unground SPM aliquots were dispersed in deionized water and then  
156 in sodium hexametaphosphate in an ultrasonic bath. For each sample, we measured three replicates and report the  
157 average median particle size ( $D_{50}$ ,  $\mu\text{m}$ ) with an uncertainty better than 2% (Table 1). The chemical composition of  
158 SPM samples was measured on ground aliquots at the Centre de Recherches Pétrographiques et Géochimiques  
159 (CRPG), Vandoeuvre-lès-Nancy, France, using inductively coupled plasma atomic emission spectroscopy (ICP-OES)  
160 for major elements with typical uncertainties of 3% (Carignan et al., 2001).

161 For particulate organic carbon content (POC%, wt.), stable carbon isotope  $\delta^{13}\text{C}$  (in ‰<sub>VPDB</sub>, *i.e.*, in ‰ relative to Vienna  
162 Pee Dee Belemnite) and radiocarbon isotope  $\Delta^{14}\text{C}$  (expressed as fraction modern, Fm), ground homogenized samples  
163 were fumigated using 12M HCl fumes in a closed Teflon tank at 60 °C for 48 hours to remove the carbonate fraction,  
164 and were then dried under vacuum prior to analysis. Total nitrogen content (TN%, wt.) was measured on non-acidified  
165 samples (Komada et al., 2008). Triplicate analysis on POC% and  $\delta^{13}\text{C}$  of POC (acidified aliquots) as well as TN%  
166 (non-acidified aliquots) were carried out on an Organic Elemental Analyzer (OEA) coupled with Isotope Ratio Mass  
167 Spectrometry (IRMS, Thermo Scientific Flash 2000) under continuous flow mode at Géosciences Paris Saclay  
168 (GEOPS), Orsay, France. Uncertainties on POC%,  $\delta^{13}\text{C}$ , and TN%, based on replicate measurements ( $1\sigma$ ,  $n=3$ ), are  
169 lower than 0.02%, 0.06‰, and 0.02‰, respectively. The  $^{14}\text{C}$  activity of POC was measured on a new compact  
170 accelerator mass spectrometry (AMS), *ECHO*MICADAS (Hatté et al., 2016), using a gas ion source interface system  
171 at the Laboratoire des Sciences du Climat et de l'Environnement (LSCE), Gif-sur-Yvette, France, with an absolute  
172 uncertainty of max  $\pm 0.5\%$ .

### 173 3.3 POC source apportionment

174 To quantify the contribution and associated uncertainties of various sources to POC transported in the Huanghe, a  
175 Bayesian Markov chain Monte Carlo based on a three-end member mixing scheme (burn-in of 1,000 steps, data  
176 thinning of 100, and iterations of 5,000,000) was adopted (Andersson et al., 2015). This approach assumes that the  
177 uncertainty on each end member contribution can be represented by a normal distribution. The mixing model was



178 constructed on the dual stable and radioactive isotope of the riverine POC pool ( $\delta^{13}\text{C}$  and  $\Delta^{14}\text{C}$ ) and of the three  
179 potential source pools (section 5.2) by the following equations:

$$180 \quad \text{Isotope\_ratio}_{\text{sample}} = \sum_{\text{source}} (f_{\text{source}} * \text{Isotope\_ratio}_{\text{source}})$$

$$181 \quad \sum_{\text{source}} f_{\text{source}} = 1$$

182 where  $\text{Isotope\_ratio}_{\text{sample}}$  is either the  $\delta^{13}\text{C}$  or  $\Delta^{14}\text{C}$  value of the sample,  $\text{Isotope\_ratio}_{\text{source}}$  is either the  $\delta^{13}\text{C}$  or  
183  $\Delta^{14}\text{C}$  value of different possible sources of POC and  $f_{\text{source}}$  the relative contribution of each source of POC.

### 184 3.4 Depth-integrated fluxes

185 Instantaneous depth-integrated fluxes of SPM and POC sources were calculated for the cross-section using a method  
186 developed by Bouchez et al. (2011a, b). This method is based on the systematic variation of SPM concentration in the  
187 water column (Figure 2) applying a Rouse-based model (Rouse, 1937). We first constructed a bathymetric profile of  
188 the river cross-section based on the depth information collected in the field and then modeled the velocity distribution  
189 across the transect (Figure 1b) through fits of the so-called "law of the wall" to water velocity measured at the location  
190 of each sample within the cross-section using a current meter. Afterward, the concentration of total SPM and various  
191 particle size fractions could be estimated by applying the so-called Rouse model (Rouse, 1937) to each particle size  
192 fraction separately (Bouchez et al., 2011a), resulting in a map of the particle size distribution in the river cross-section  
193 (Figure S3). The aluminum to silicon ratio (Al/Si mass ratio) is inversely related to the particle size of river SPM in  
194 the Ganges-Brahmaputra, the Amazon, and the Mackenzie Rivers (Galy et al., 2007; Bouchez et al., 2014; Hilton et  
195 al., 2015). Such a linear relationship between D50 and Al/Si was also observed in our dataset, allowing for computing  
196 the spatial distribution of POC content in the cross-section, based on the linear relationship between POC and Al/Si  
197 (Figure 3). Finally, combining modeled water velocity, SPM concentration, and POC distribution we calculated a  
198 depth-integrated, instantaneous POC flux for the whole river channel (Figure S3, detail in supplementary material).

## 199 4 Results

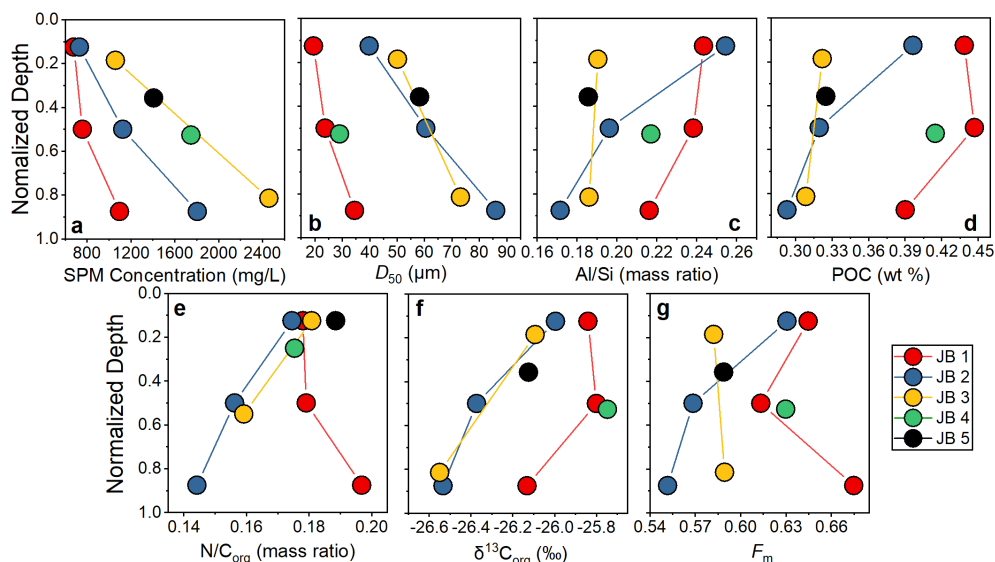
200 We report the first isotopic dataset of POC samples collected along several depth profiles distributed over a cross-  
201 section of the Huanghe (Table 1). SPM concentrations range from 679 to 2,459 mg/L (avg. 1,286 mg/L) and show an  
202 obvious increase from the surface to the bottom and from the right bank to the left bank (Figure 1b and 2a). The  
203 surface SPM concentration (*i.e.*, samples collected 0.5 m below the surface) decreases laterally as the water column  
204 deepens. The range of measured Huanghe SPM D50, *i.e.* the median particle size (19.5-86.0  $\mu\text{m}$ , Figure 2b) agrees  
205 with that (16.6-120.1  $\mu\text{m}$ ) of SPM collected at Lijin during the same flooding season by Moodie et al. (2022). The  
206 finest SPM is transported on the right bank and at the surface, while the coarsest SPM is found at the bottom of the  
207 middle profile (sample JB 2-3). Two types of depth profiles can be distinguished at Luokou based on particle size  
208 distributions (Figure S4) and the relationship between D50 and water depth (Figure 2b). On the one hand, the JB 1  
209 and JB 4 profiles show a well-marked, bi-modal distribution of particle size (Figure S4) together with relatively low  
210 and consistent D50 (Figure 2b). On the other hand, the JB 2, JB 3, and JB 5 profiles show a more unimodal distribution





211 of particle size (Figure S4) and a unique D50 - sampling depth relationship (Figure 2b). Interestingly, these two groups  
212 can also be distinguished in terms of relationships between POC% and  $\delta^{13}\text{C}$  with water depth (Figures 2d and 2f). As  
213 expected, the Al/Si ratio is well-related to the particle size, with ratios of the middle profile varying from 0.17 (sample  
214 JB 2-3) to 0.26 (sample JB 2-1), covering the full range of Al/Si measured in the whole cross-section (avg,  $0.21 \pm$   
215  $0.03$ , Figure 2c). The relatively low Al/Si ratios are comparable to that of the middle Huanghe (Qu et al., 2020) and  
216 other large turbid river systems such as the Ganges-Brahmaputra (Galy et al., 2008b), Salween, and Irrawaddy (Tipper  
217 et al., 2021).

218 SPM in the Huanghe is characterized by low TN and POC content (wt.%), ranging from 0.04% to 0.08% (avg.,  $0.06$   
219  $\pm 0.01\%$ ) and from 0.29% to 0.42% (avg.,  $0.37 \pm 0.06\%$ ), respectively (Figure 2d; Table 1). POC content generally  
220 decreases from the surface to the river bed, with quantitative differences from one profile to another (Figure 2d).  
221 Notably, the JB 1 profile shows the highest POC% and TN%. In addition, the ratio of TN% to POC%:  $\text{N}/\text{C}_{\text{org}}$  increases  
222 with depth in the JB 1 profile (from top to bottom), while it decreases in the JB 2 and JB 3 profiles (Figure 2e). The  
223  $\delta^{13}\text{C}$  of POC varies over a narrow range from  $-26.55\%$  to  $-25.75\%$  (avg.,  $-26.12 \pm 0.29\%$ , Figure 2f) and becomes  
224 lighter with depth, showing that fine SPM has higher  $\delta^{13}\text{C}$  than coarse SPM. These values are lower than those  
225 previously reported for other Huanghe sampling sites upstream:  $-24.7 \pm 0.4\%$  at Toudaoguai,  $-24.9 \pm 0.6\%$  at  
226 Longmen, and  $-23.8 \pm 0.6\%$  at Lijin (Qu et al., 2020, Hu et al., 2015; Tao et al., 2015; Yu et al., 2019a; Ge et al.,  
227 2020). The radiocarbon activity of POC of the Huanghe at Luokou is relatively low (Figure 2g), with  $F_m$  ranging  
228 from 0.552 ( $\Delta^{14}\text{C} = -453\%$ ; sample JB2-3) to 0.675 ( $\Delta^{14}\text{C} = -331\%$ ; sample JB1-3), spanning from 3,160 to 4,780  
229  $^{14}\text{C}$  yr, and the average value is  $0.607 \pm 0.038$  ( $\Delta^{14}\text{C} = -412\%$ ). This range of radiocarbon activity is consistent with  
230 published values for POC collected at the river surface downstream of Toudaoguai (Qu et al., 2020). All the POC  
231 radiocarbon activity data reported so far for the Huanghe are comparable to mean values for Arctic large rivers ( $\Delta^{14}\text{C}$   
232  $= -397\%$ , ca. 4,480  $^{14}\text{C}$  yrs, Ke et al., 2022), revealing the multimillennial-age nature of POC transported by the  
233 Huanghe. The elemental and isotopic signatures of the two fine SPM samples HH 17.05 and HH 17.06 (on average  
234  $\text{POC}\% = 1.07\%$ ,  $\delta^{13}\text{C} = -25.67\%$ ,  $F_m = 0.720$ ; and  $\text{Al}/\text{Si} = 0.37$ ) are significantly different from those of the depth  
235 profile samples (Table 1). The bedload sample has a comparatively low POC% (0.21%),  $\delta^{13}\text{C}$  ( $-27.35\%$ ),  $F_m$  (0.099),  
236 and Al/Si ratio (0.17).



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**Figure 2: Variation of physical and chemical parameters in the river cross-section, shown as a function of sampling depth normalized to total depth of the water column at the location of the considered depth-profile, for the Luokou cross-section on the Huanghe (June 16, 2017). (a) SPM concentration; (b) particle size distribution (shown as D50); (c) Al/Si mass ratio; (d) POC content (weight %); (e) N/C<sub>org</sub> mass ratio; (f) stable carbon isotope ratio  $\delta^{13}\text{C}_{\text{org}}$  (‰); (g) radiocarbon activity  $F_m$ .**

242 **Table 1: SPM characteristics and POC properties of the river-cross-section sampling.**

Sample ID	Type	Depth (m)	SPM mg/L	POC (%)	SD	$\delta^{13}\text{C}_{\text{org}}$ (‰)	SD	$F_m$	$\Delta^{14}\text{C}$ (‰)	SD	$^{14}\text{C}$ age	TN (%)	N/C <sub>org</sub>	Al/Si	D50 $\mu\text{m}$
JB 1-1	SPM	0.5	679	0.44	0.02	-25.84	0.03	0.645	-360	7	3527	0.078	0.178	0.243	19.5
JB 1-2	SPM	2	757	0.45	0.01	-25.80	0.06	0.613	-392	11	3929	0.080	0.179	0.238	23.7
JB 1-3	SPM	3.5	1095	0.39	0.01	-26.13	0.04	0.675	-331	7	3161	0.077	0.197	0.216	34.5
JB 2-1	SPM	0.5	730	0.40	0.03	-26.00	0.01	0.631	-374	7	3703	0.069	0.175	0.255	39.8
JB 2-2	SPM	2	1124	0.32	0.01	-26.37	0.06	0.569	-436	13	4537	0.050	0.156	0.196	60.4
JB 2-3	SPM	3.5	1806	0.29	0.01	-26.53	0.06	0.552	-453	38	4779	0.042	0.144	0.172	86.0
JB 3-1	SPM	0.5	1058	0.32	0.02	-26.09	0.03	0.582	-422	12	4346	0.058	0.181	0.190	50.1
JB 3-2	SPM	2.2	2459	0.31	0.02	-26.55	0.04	0.589	-415	11	4247	0.049	0.159	0.186	73.1
JB 4-1	SPM	1	1747	0.41	0.02	-25.75	0.06	0.630	-375	8	3714	0.073	0.175	0.217	29.0
JB 5-1	SPM	0.5	1406	0.32	0.01	-26.12	0.05	0.589	-416	11	4256	0.061	0.188	0.186	58.2
HH 17.05	SPM	0	83	0.92	0.00	-25.73	0.14	0.711	-295	19	2740	0.184	0.200	0.358	5.2
HH 17.06	SPM	0	54	1.21	0.01	-25.60	0.07	0.729	-277	25	2539	0.261	0.215	0.377	4.3
HH	Bedload			0.21	0.03	-27.35	0.05	0.099	-901	7	18539	0.019	0.087	0.175	44.4

## 243 5 Discussion

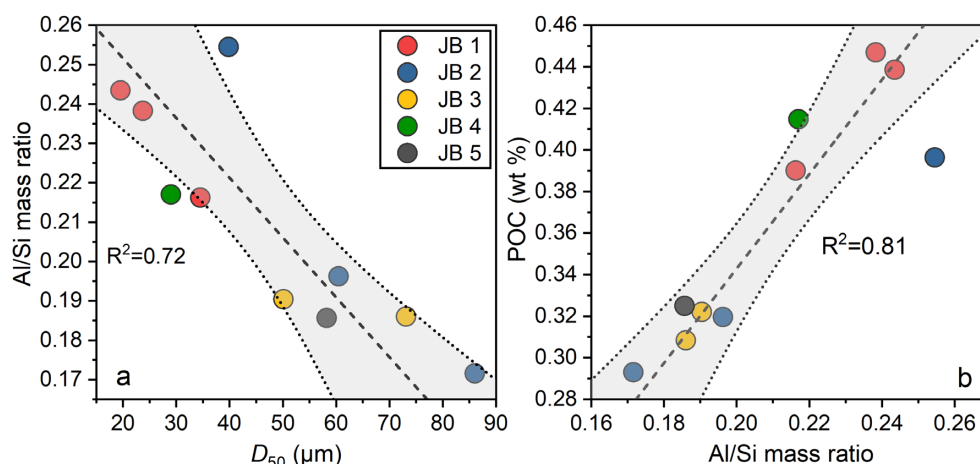
244 We observe significant heterogeneities of elemental and isotopic carbon composition as well as inorganic chemistry  
 245 over the studied river cross-section. The possible mechanisms behind these variations are assessed hereafter. Then,  
 246 sources of riverine POC are determined and quantified, confirming that erosion of the loess-paleosol sequence of the  
 247 CLP is a major source of aged and refractory biospheric OC to the Huanghe. Finally, we assess the POC load and its  
 248 variability over the transect profile, inferring the importance of the supply of POC from the river bottom in the  
 249 Huanghe.



250 **5.1 Transportation mode of POC in the Huanghe**

251 **5.1.1 POC loading and its controls**

252 The Huanghe is characterized by a high SPM load with relatively low POC% (avg.  $0.37 \pm 0.06\%$ ). In the Luokou  
 253 cross-section, POC content generally increases with decreasing particle size (Figure S5), with the two clay-sized  
 254 ("HH") samples showing the largest POC content (Table 1). Consistently, the Al/Si ratio of Huanghe sediments, which  
 255 varies as an inverse linear function of the median particle sizes  $D_{50}$  ( $R^2=0.72$ , Figure 3a), positively correlates with  
 256 POC% ( $R^2=0.81$ , Figure 3b), a pattern observed globally (Galy et al., 2008b; Bouchez et al., 2014; Hilton et al., 2015).  
 257 This pattern is consistent with POC variability in the Huanghe reported for manually separated size fractions of  
 258 sediments (Yu et al., 2019b).

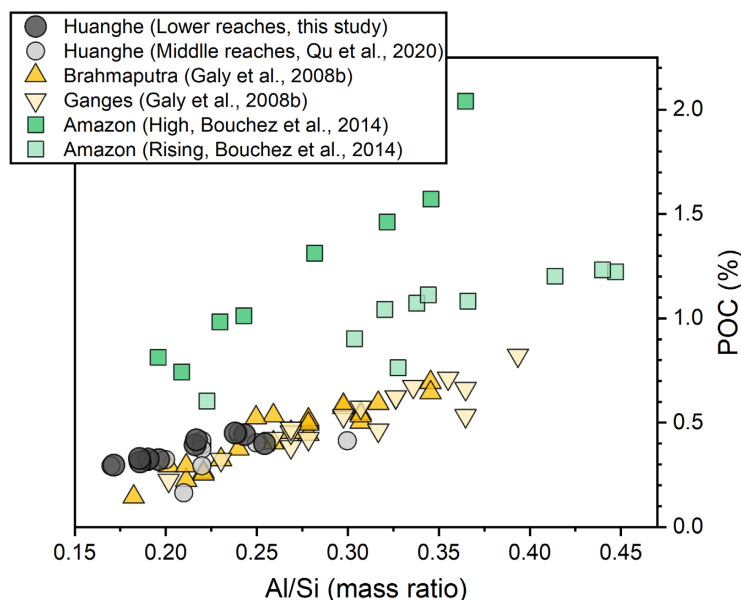


259  
 260 **Figure 3: Relationships between (a) particle size  $D_{50}$  and Al/Si mass ratio; (b) Al/Si mass ratio and POC content for the**  
 261 **Luokou cross-section on the Huanghe (July 17, 2016). The grey shade represents the 95% confidence area of the linear best-**  
 262 **fit (black dashed line), the upper and lower bound are marked by grey dotted lines.**

263 The first reason for the low POC content of Huanghe sediments is therefore their relatively low values of Al/Si  
 264 compared to other systems - a feature than can be related to the quartz-rich, OC-poor nature of the loess-paleosol  
 265 formations of the CLP (Jahn et al., 2021; Huang and Ren, 2006; He et al., 2006; Ning et al., 2006; Wang and Fu,  
 266 2016). However, Huanghe sediments are relatively poor in POC, even considering their low Al/Si, compared to other  
 267 rivers globally. To that effect, the so-called "POC loading" can be characterized by the slope described by sediment  
 268 data in an Al/Si-POC diagram (Galy et al., 2008b; Figure 4). For a given Al/Si ratio, the POC% in the Luokou cross-  
 269 section is similar to that of the middle Huanghe (Qu et al., 2020), indicating the relatively invariant transport mode of  
 270 POC between the middle and lower reaches. Previous studies have shown that the positive relationship between POC%  
 271 and Al/Si can be partially explained by OC adsorption onto the mineral surface (Curry et al., 2007; Galy et al., 2008b;  
 272 Blair and Aller, 2012; Bouchez et al., 2014; Qu et al., 2020). In the loess-paleosol deposits acting as a source of  
 273 sediments to the Huanghe, OC is mostly preserved and stabilized by forming organo-aggregates with kaolinite and  
 274 through adsorption onto iron oxides (Wang et al., 2013).



275 However, POC loading in the Huanghe is small compared to that of the Amazon (Bouchez et al., 2014), but similar  
 276 to that of the Ganges-Brahmaputra system (Galy et al., 2008b). While many factors could influence POC loading  
 277 across these catchments, we note that another similarity between the Huanghe and Ganges-Brahmaputra fluvial  
 278 systems is their millennial-aged  $OC_{bio}$  (Galy et al., 2007; Tao et al., 2016). This is in stark contrast with the Amazon,  
 279 where younger  $OC_{bio}$  ages have been reported (Bouchez et al., 2014). Given that younger  $OC_{bio}$  recently  
 280 photosynthesized in terrestrial or aquatic ecosystems can be readily oxidized within catchments (Mayorga et al., 2005),  
 281 the relatively low POC loading observed at the mouth of the Ganges-Brahmaputra and the Huanghe (Figure 4) could  
 282 be related to the predominance of refractory, aged  $OC_{bio}$  and  $OC_{petro}$  in those systems, while the Amazon sediments  
 283 would still contain significant amount of younger, more labile  $OC_{bio}$ .



284 **Figure 4: "POC loading" of river SPM of large rivers. The POC loading is estimated from the slope of the relationship**  
 285 **between POC content and the Al/Si ratio of each fluvial system (Galy et al., 2008b). All SPM samples were collected along**  
 286 **depth profiles except for the middle Huanghe (Qu et al., 2020).**  
 287

288 In detail, and as explained in more detail below (Section 5.1.2) we also observe a significantly different POC loading  
 289 between the JB 1 and JB 2 depth profiles at the Luokou station (Figure 3). This difference in POC loading in the cross-  
 290 section of the Huanghe might indicate the delivery of recent  $OC_{bio}$ , specifically near the right bank (the closest to the  
 291 JB 1 profile) for the Luokou site, a scenario which is supported by the comparatively younger age of POC in profile  
 292 JB 1. Consistently with this interpretation, temporally variable POC loading at a given site has been reported for the  
 293 Amazon (Bouchez et al., 2014), where higher POC loading during the high-water stage compared to the rising water  
 294 stage has been attributed to the erosion of discrete organic debris from riverbanks.

295 Variable POC loading amongst large catchments has implications for evaluating the likelihood of POC preservation  
 296 in estuaries. The Ganges-Brahmaputra system delivers relatively old, refractory  $OC_{bio}$  to the Bengal Fan with an almost  
 297 complete burial efficiency (Galy et al., 2007). Given the observed similarity in POC loading and age, we can thus  
 298 expect a similar, efficient preservation for the Huanghe offshore depositional system. In addition to the low reactivity



299 of the POC transported by the Huanghe, the high sediment accumulation rates in the Huanghe coastal domain might  
300 further inhibit OC oxidation (Blair and Aller, 2012). Consequently, the case of the Huanghe differs drastically from  
301 that of the Amazon, where higher POC loading is observed, with a larger contribution of young, labile OC<sub>bio</sub> either as  
302 discrete organic matter or associated with mineral surfaces, leading to low POC burial efficiency in the ocean  
303 (Bouchez et al., 2014; Blair and Aller, 2012).

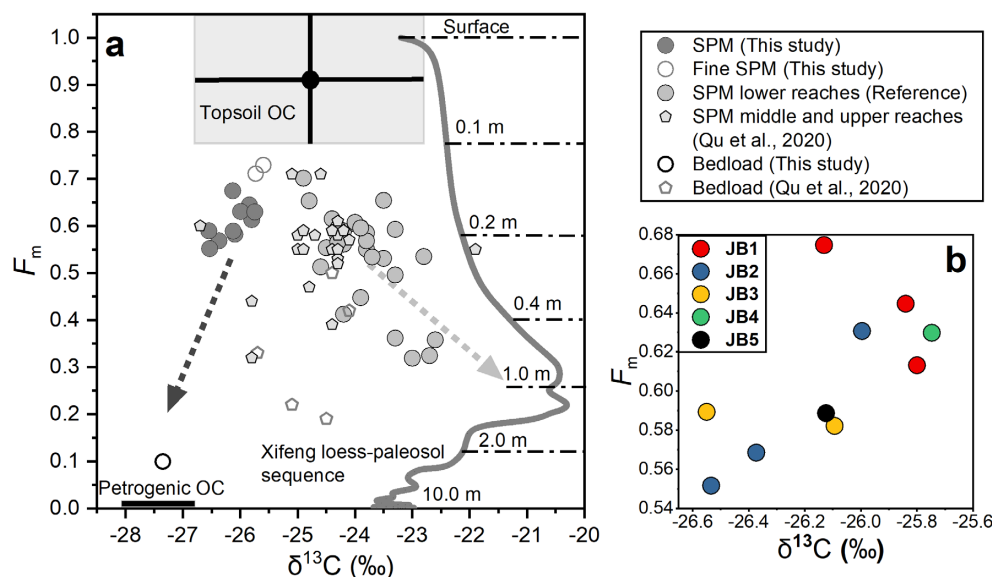
### 304 **5.1.2 Chemical heterogeneity within the transect**

305 There is clear lateral and vertical variability of POC content and SPM inorganic chemistry across the Luokou cross-  
306 section of the Huanghe. For each vertical depth profile, clay-rich fine particles are transported near the channel surface,  
307 and quartz-rich coarse particles flow near the river bottom. Accordingly, the Al/Si ratio, POC content and POC  
308 radiocarbon activity generally decrease with depth. Elemental (POC%) and isotopic POC signatures (<sup>13</sup>C and <sup>14</sup>C) are  
309 inversely related to the particle size (D<sub>50</sub>; Figure S5). These patterns are observed in other large fluvial systems, e.g.,  
310 Ganges and Brahmaputra, Amazon and Mackenzie (Galy et al., 2008b; Bouchez et al., 2014, Hilton et al., 2015),  
311 showing that hydrodynamic sorting is the primary control on suspended sediment OC content, segregating inorganic  
312 and organic material according to particle size (Bouchez et al., 2011a, 2014).

313 At the Luokou sampling site, lateral variability at the channel surface shows that POC-rich fine particles are  
314 preferentially transported near the right bank (Figure 2 and Figure S3). The channel geometry thus needs to be  
315 examined as a potential factor to produce such lateral heterogeneity, in particular through bank erosion and bedload  
316 resuspension.

317 Bank erosion can be a significant mechanism for the delivery of sediments to river systems (Guo et al, 2007). Bank  
318 erosion at Luokou would make OC from the lower Huanghe alluvial plain a potential source of POC in the lower  
319 reaches of the Huanghe. This mechanism provides a possible explanation for the opposite trends displayed by samples  
320 from the JB 1 and JB 2 profiles in the F<sub>m</sub> vs. δ<sup>13</sup>C space (Figure 5). The youngest POC was found at the bottom of  
321 the JB 1 profile. Meanwhile, the JB 1 samples have comparatively higher N/C<sub>org</sub> ratios and N%, consistent with the  
322 input of discrete organic matter (e.g., plant debris) from the bank in addition to rock-derived detrital clastic material  
323 in the coarse fractions (> 32 μm, Yu et al., 2019b). Such input would also provide an explanation for the higher POC  
324 loading of the JB 1 profile (**Section 5.1.1**).

325 Resuspension of bed sediments is also a possible mechanism to explain the lateral heterogeneity in POC content in  
326 the study cross-section of the Huanghe. Indeed, scouring of channel bed sediment at high water flow may also shift  
327 POC to more negative radiocarbon and stable isotope signatures. Our sample set collected in July 2016 during flooding  
328 (water flow velocity up to 2.1 m/s, Figure 1) supports this scenario. Indeed, the increase in D<sub>50</sub> of surface SPM  
329 samples from right to the left bank, that is with total channel depth decrease, is consistent with coarse sediment  
330 resuspension from the bed. Such a scenario is also supported by the three-fold increase in sediment flux observed  
331 along the course of the Huanghe from Huayuankou to Lijin in July 2016, despite a four-fold decrease in water  
332 discharge (Figure S2).



333  
 334 **Figure 5: (a)  $^{14}C$  activity (expressed as  $F_m$ ) vs.  $\delta^{13}C$  for a compilation of POC data collected over the 2011-2016 period in**  
 335 **the lower Huanghe, including samples from this study and previous studies at Huayuankou, Lijin, and Kenli (Hu et al.,**  
 336 **2015; Tao et al., 2015; Yu et al., 2019a; and Ge et al., 2020); SPM and bedload collected by Qu et al., 2020 at Toudaoguai**  
 337 **(most downstream location of the upper reaches) and Longmen in the middle reaches (Table S1). The grey curve**  
 338 **corresponds to  $\delta^{13}C_{org}$  of the top 10 m of the Xifeng loess-paleosol (Ning et al., 2006), and the corresponding  $F_m$  was**  
 339 **calculated from  $^{10}Be$ -derived ages following ‘Age =  $-8033 \cdot \ln(F_m)$ ’. The soil depth is marked above the dot-dash line (Zhou**  
 340 **et al., 2010). Topsoil OC represents OC from the upper 10 cm of the loess-paleosol sequence with standard deviation marked**  
 341 **with black lines. (b)  $^{14}C$  activity (expressed as  $F_m$ ) vs.  $\delta^{13}C$  diagram for the Huanghe sediment samples collected in this**  
 342 **study at the Luokou cross-section.**

## 343 5.2 POC provenance in the Huanghe: the significance of loess-paleosol-derived OC

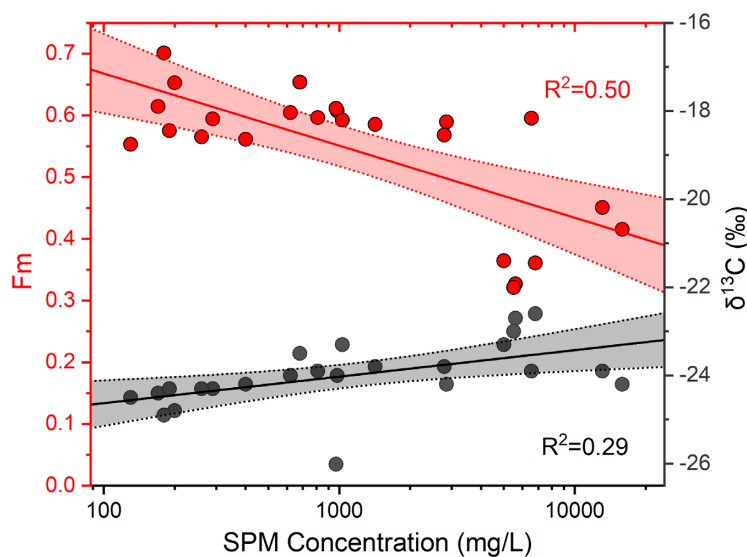
### 344 5.2.1 Physical erosion of the loess-paleosol sequence

345 Over decennial to centennial time scales, the POC export of the Huanghe is mainly controlled by erosion of the CLP.  
 346 Throughout the Quaternary, the erosion rate in the Huanghe basin has been mainly driven by climate shifts until human  
 347 activities started and profoundly impacted sediment fluxes in the mid-Holocene (He et al., 2006). To determine the  
 348 contributions of the various terrestrial OC components to Huanghe POC, we compiled published POC carbon isotope  
 349 data for sediments collected in the lower reaches from 2011 to 2016 (Figures 5 and 6). Even though the Huanghe has  
 350 experienced a 90% decrease in annual sediment load since the 1950s (Wang et al., 2015), the radiocarbon ages of  
 351 Huanghe POC are considerably old (avg  $5,100 \pm 1,700$   $^{14}C$  yr,  $n=29$ ), with a minor fraction of modern  
 352 photosynthesized  $OC_{bio}$  (Tao et al., 2015; Yu et al., 2019a, b). This relatively  $^{14}C$ -depleted POC suggests the significant  
 353 contribution of OC originated from deep soil horizons within the catchment. Given the fact that loess is easily erodible,  
 354 the negative trend between  $^{13}C$  and  $F_m$  of POC for sediment samples collected in the Huanghe over the 2011-2016  
 355 period (Figure 5a) suggests that deep horizons of loess-paleosol formations are a plausible source for the  $^{14}C$ -depleted  
 356 end member. Besides, the preferential erosion of recently photosynthesized and possibly degraded  $OC_{bio}$  from the  
 357 overlying topsoils ( $< 10$  cm) most likely contributes to riverine POC (Tao et al., 2015).





358 As such, variable contribution of aged and radiocarbon-free OC from deep horizons of loess-paleosol formations of  
 359 the CLP should have a significant impact on the elemental and isotopic signature of POC in the lower Huanghe.  
 360 Erosion of loess-paleosol can also explain the decreasing POC% with increasing SPM concentration at different sites  
 361 of the main channel (Ran et al., 2013; Qu et al., 2020), the negative relationship between SPM concentration and  
 362 corresponding POC Fm at Luokou (Figure 6), and the low POC loading of the Huanghe (Section 5.1.1), as the deep  
 363 horizons of the loess-paleosol sequences are OC-poor, and mostly host OC that is highly degraded and refractory (Liu  
 364 et al., 2012; Wang et al., 2013; Cheng et al., 2020). However, the slight increase of POC  $\delta^{13}\text{C}$  with increasing SPM  
 365 concentration might indicate a significant supply of soil OC from loess-paleosol shallower depth, as inferred from the  
 366  $\delta^{13}\text{C}$  variation within the Xifeng loess-paleosol sequence (Figure 5a).



367  
 368 **Figure 6:**  $^{14}\text{C}$  activity (expressed as Fm; red circles) and  $\delta^{13}\text{C}$  (black circles) of POC vs. SPM concentration for surface  
 369 samples from the Huanghe collected from 2011 to 2016 (average SPM concentration of surface samples in this study and  
 370 from Hu et al. (2015), Tao et al. (2015), Yu et al. (2019a), and Ge et al. (2020)). These paired dual carbon isotope data  
 371 corresponds to the group ‘SPM lower reaches’ in Figure 5. Straight lines correspond to best-fit logarithmic curves, and  
 372 shaded areas represent the 95% confidence interval.

373 The  $\text{N}/\text{C}_{\text{org}}$  ratio provides additional evidence for the significant contribution of loess-paleosol material to Huanghe  
 374 POC (Figure S6). Indeed, the  $\text{N}/\text{C}_{\text{org}}$  ratios of SPM collected in the lower reaches ranges from 0.10 to 0.23 (this study,  
 375 Ran et al., 2013; Yu et al., 2019a), whereas topsoils of the CLP are characterized by  $\text{N}/\text{C}_{\text{org}}$  lower than 0.14 (Liu and  
 376 Liu, 2017) and sedimentary rocks typically have very low  $\text{N}/\text{C}_{\text{org}}$  (Hilton et al., 2015). Soil OC input from the North  
 377 China Plain is also unlikely given its  $\text{N}/\text{C}_{\text{org}}$  of 0.10-0.13 (Shi et al., 2017). Therefore, all these possible sources cannot  
 378 explain the high  $\text{N}/\text{C}_{\text{org}}$  signatures of riverine SPM. In addition, the high turbidity of the Huanghe ( $> 600 \text{ mg/L}$ ) during  
 379 the sampling season is likely to inhibit *in-situ* primary production ( $\text{N}/\text{C}_{\text{org}} > 0.13$ ) (Zhang et al., 2013; Hu et al., 2015).  
 380 As a result, only soil OC from deep loess-paleosol horizons appears as a plausible supplier to downstream Huanghe  
 381 POC, given the high  $\text{N}/\text{C}_{\text{org}}$  ratios previously reported for various loess-paleosol sequences (Figure S6, Ning et al.,  
 382 2006).



383 Geomorphic processes in the CLP region support the erosion of deep soil horizons. There, gully erosion is thought to  
384 be responsible for more than 80% of the total sediment yield in the CLP (He et al., 2006). Gullies are densely  
385 distributed and cover about 42% of the total area of the CLP and up to 60% in hilly regions (Huang and Ren, 2006;  
386 He et al., 2006). Nowadays, the well-developed gully geomorphic system of the CLP is characterized by gullies with  
387 a depth of about 10 m on average and represents the most active vertical and regressive erosion of loess (Huang and  
388 Ren, 2006). This incision process erodes all types of unconsolidated materials, including the loess-paleosol sequence,  
389 underlying red clays, and colluvial deposits in the form of creeps, falls, and slides in the watershed (Zhu, 2012). All  
390 these observations suggest that gully erosion strongly impacts the composition of riverine POC. As gully erosion is  
391 sensitive to climate change and anthropogenic activities, soil dynamics in the Huanghe basin have been altered since  
392 the mid-Holocene. In recent years, soil and water conservation and environmental rehabilitation campaigns (Wang et  
393 al., 2007) largely contributed to the reduction of SPM export by the Huanghe with a transfer to the estuary of 10.6 Mt  
394 in 2016, which represents a few percent of the annual sediment flux measured in 2013 (172.8 Mt) and less than one  
395 percent of the flux of the 1950s (*ca.* 1,340 Mt; Wang et al., 2015). Such modifications should thus have drastically  
396 inhibited the POC export by the Huanghe.

### 397 **5.2.2 POC source determination and end member apportionment**

398 Considering the SPM geochemistry and the basin characteristics, three terrestrial sources can be identified as necessary  
399 to form the composition of the Huanghe POC at the Luokou cross-section. As discussed in **Section 5.2.1** and shown  
400 in Figure 5a, two of these sources are (a) topsoil-derived OC ( $OC_{ts}$ ) and (b) OC from deeper horizons of the loess-  
401 paleosol sequence ( $OC_{lps}$ ) excluding topsoil. In addition, at the Luokou cross-section, bed OC shows lower  $F_m$  and  
402  $\delta^{13}C$  values compared to that of SPM, suggesting a significant contribution of (c) rock-derived OC from erosion in the  
403 middle reaches ( $OC_{petro}$ ).

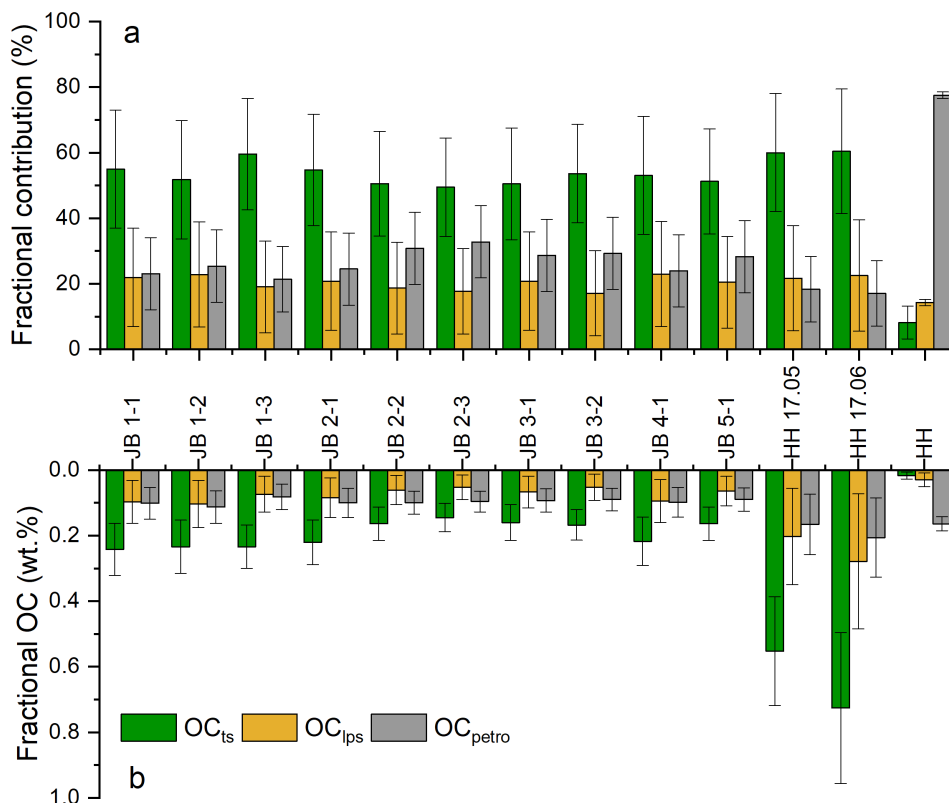
404 We adopted a Bayesian Monte-Carlo model to reconstruct source apportionment based on the mass balance of carbon  
405 isotopes ( $\delta^{13}C$  and  $\Delta^{14}C$ ) of our three defined end members (**Section 3.3, Appendix A**). Modeling results are shown  
406 in Figure 7 as relative contributions (Figure 7a) and weight percentage (Figure 7b) of  $OC_{ts}$ ,  $OC_{lps}$ , and  $OC_{petro}$  (Table  
407 S2). The contribution of  $OC_{petro}$  to total Huanghe POC at Luokou varies between 21% and 33% in the cross-section,  
408 which is higher than the contribution calculated for in the two fine SPM samples (17% on average) and much smaller  
409 than for the bed sediment sample ( $78 \pm 10\%$ ). The inferred  $OC_{petro}$  concentration in the sediment is remarkably uniform  
410 in the cross-section, representing  $0.10 \pm 0.01\%$  of SPM (Figure 7b). This result is consistent with the OC contents of  
411 midstream sedimentary rocks at  $0.09 \pm 0.08\%$  (Qu et al., 2020). In addition, these findings imply that  $OC_{petro}$   
412 concentration does not depend on particle size and confirm previous findings of  $OC_{petro}$  being present in a range of  
413 clastic particles or as discrete particles (Galy et al., 2008a; Bouchez et al., 2014). In other words, the rock-derived OC  
414 has a relatively invariant contribution with depth (Galy et al., 2008a; Bouchez et al., 2014), meaning that biospheric  
415 OC exerts a first-order control on POC content and isotopic variations throughout the cross-section.

416 At the study cross-section,  $OC_{ts}$  and  $OC_{lps}$  contribute 50%-60% and 17%-23% to the total POC, respectively (Figure  
417 7a). The sum of these two components can be considered as  $OC_{bio}$ , which varies between 67% and 77%. The  
418 corresponding  $OC_{bio}$  content of sediment is quite variable, ranging from 0.20% (sample JB 2-3) to 0.34% (sample JB



419 1-1), and generally decreases from the river surface to the bottom. Given the rather invariant  $OC_{petro}$  concentration in  
 420 the sediment, there are thus marked heterogeneities of POC provenance in the cross-section. For instance, POC  
 421 transported close to the right bank and in the finer SPM samples show a higher contribution from  $OC_{bio}$ . From the  
 422 knowledge of the relative contributions of  $OC_{ts}$  and  $OC_{lps}$  and their corresponding  $^{14}C$  activity, the  $F_m$  values for the  
 423 bulk  $OC_{bio}$  can be estimated based on mass balance. The modeled radiocarbon activity of  $OC_{bio}$  is relatively  
 424 homogeneous with an average  $\Delta^{14}C$  value of  $-178 \pm 11\text{‰}$  ( $1,510 \pm 110$   $^{14}C$  yr) whereas a greater variance is observed  
 425 in the bulk  $^{14}C$  content. In summary, our results support the first-order control of  $OC_{bio}$  abundance on POC content  
 426 and age in the Huanghe.

427



428

429

430 **Figure 7: (a) Relative contributions of the three different sources of Huanghe POC, (b) fractional OC weight percentage**  
 431 **Huanghe POC at the Luokou cross-section, as inferred from a mixing model.  $OC_{ts}$  is the topsoil-derived OC,  $OC_{lps}$**   
 432 **represents the loess-paleosol sequence OC excluding topsoil, and  $OC_{petro}$  is the rock-derived OC eroded from the Huanghe**  
 433 **middle reaches.**

433

434

435

436

437

Applying the same mixing model to previously published Huanghe POC data (2011-2016, Table S1) shows (1)  
 dominance of the  $OC_{bio}$  contribution to POC, (2) variable relative mixing proportions of  $OC_{ts}$  and  $OC_{lps}$ ; (3) a wide  
 range of  $^{14}C$  age for  $OC_{bio}$  (from 1,040 to 8,050 yr). In particular,  $OC_{ts}$  and  $OC_{lps}$  contributed 20%-30% and 46%-68%  
 to POC collected in 2013 (Hu et al., 2015), leading to 75-89% of  $OC_{bio}$ . Yu et al. (2019a) estimated that  $OC_{bio}$   
 contributed 63%-81% to the lower Huanghe POC (2015-2016) using a different mixing model. Using their data in our



438 mixing model results in a slightly higher  $OC_{bio}$  contribution of 77%-87%, consisting of 39%-53% for  $OC_{is}$  and 30%-  
439 47% for  $OC_{lps}$ . The small difference in source contribution mainly results from the fact that old  $OC_{bio}$  from loess-  
440 paleosol sequences was not considered in Yu et al. (2019a), and from the different isotopic signatures chosen for the  
441 POC endmembers. However, both estimates ignore the possible presence of rock-derived OC in soils. In any case, our  
442 results suggest that the Huanghe transports more  $OC_{bio}$ -derived POC than previously thought, with more aged, soil-  
443 derived OC.

444 It is worth noticing that these calculations suggest that the  $OC_{lps}$  fraction in the Huanghe was significantly higher in  
445 2013 than in 2016. As most Huanghe sediments are derived from the CLP, higher physical erosion in the CLP should  
446 enhance supply of aged, refractory  $OC_{bio}$  to the river system. As a consequence, the decrease in sediment supply from  
447 the CLP initiated a few decades ago (Wang and Fu et al., 2016), which is likely to continue in the future, will probably  
448 lead to the reduction of the contribution of  $OC_{lps}$  to total POC export from the Huanghe. This might have an impact  
449 on the burial efficiency of riverine POC on the continental margins, as  $OC_{is}$  is more labile than  $OC_{lps}$ , and thus more  
450 prone to the remineralization process before burial. Moreover, decreasing erosion rate in the Huanghe basin will lead  
451 to decreasing sediment accumulation rate in the estuary, which potentially favors the oxidation of all POC components  
452 before burial (Blair and Aller, 2012). The time scale over which such effect could take place is yet unknown, as  
453 anthropogenic intervention is the primary reason for the sediment yield reduction, through afforestation and soil and  
454 water conservation measures in the CLP and reservoir operation in the middle reaches of the Huanghe. However, it is  
455 plausible that in response to decreased terrestrial physical erosion on the Loess Plateau over at least decadal timescales,  
456 an increased proportion of Huanghe POC will be oxidized before burial in the ocean, thereby leading to a weakened  
457 preservation efficiency for the terrestrial eroded POC.

### 458 **5.3 POC export by the Huanghe**

459 In the Huanghe, POC content varies both vertically and laterally throughout the cross-section (Figure 2). This spatial  
460 variability of both physical and chemical SPM characteristics must be considered when estimating integrated  
461 instantaneous POC concentration and flux (Section 3.4).

462 We calculate that (Text S1) at the sampling time (July 2016), the Huanghe at Luokou transported 1,075 kg/s of SPM  
463 for a water discharge of 731 m<sup>3</sup>/s, such that the spatially-integrated SPM concentration over the cross section ( $SPM_{int}$ )  
464 was 1,472 mg/L, a value relatively close to the straightforward average concentration of our 10 samples (1,286 mg/L).  
465 The Luokou gauging station records a monthly SPM load of 1,826 kg/s in July 2016, and the daily average SPM load  
466 of 1,096 kg/s for a daily average water discharge of 643 m<sup>3</sup>/s on the 16<sup>th</sup> and 17<sup>th</sup> of July 2016 (method: three water  
467 samples collected at 0.5 m below the channel surface across the transect profile, data available at  
468 <http://www.yrcc.gov.cn>). Even though the latter estimate neglects the vertical heterogeneity within this relatively  
469 shallow river (< 5.0 m), estimates give similar results.

470 We further obtain an instantaneous POC flux of 3.69 kg/s, corresponding to a cross-section integrated average POC  
471 content ( $POC_{int}\%$ ) of 0.34% when dividing this instantaneous POC flux by the instantaneous SPM load. Given the  
472 relatively homogenous distribution of  $OC_{petro}$ , the instantaneous flux of  $OC_{petro}$  was calculated by multiplying the  
473 average  $OC_{petro}$  content by the instantaneous cross-section integrated SPM flux, yielding  $1.07 \pm 0.11$  kg/s. The



474 instantaneous  $OC_{bio}$  flux was then calculated by subtracting the instantaneous flux of  $OC_{petro}$  from the instantaneous  
475 POC flux, yielding  $2.61 \pm 0.26$  kg/s. Assuming that our SPM samples are representative in terms of POC content  
476 exported in July 2016, and taking the SPM flux of the gauging station for July, then the estimated fluxes of POC,  
477  $OC_{bio}$ , and  $OC_{petro}$  for the flood period of July 2016 are 6.1, 4.3, and 1.8 kg/s, respectively. Taking  $POC_{int}$  content for  
478 estimating the annual POC flux yields a value of 1.1 kg/s consisting of 0.8 and 0.3 kg/s for  $OC_{bio}$  and  $OC_{petro}$  fluxes,  
479 respectively. Note that these numbers are lower-bound estimates because POC content in Huanghe SPM collected  
480 during flood periods is generally the lowest (Ran et al., 2013).

481 The above numbers present a sharp decrease compared to the estimated POC and  $OC_{bio}$  fluxes transported by the  
482 Huanghe over the period 2008 to 2013. Galy et al. (2015) estimated an  $OC_{petro}$  flux of 1.9 kg/s and an  $OC_{bio}$  flux of  
483 11.4 kg/s from 2008 to 2012 (SPM flux: 3,655 kg/s, YRCC 2016), while Tao et al. (2018) reported an  $OC_{petro}$  flux of  
484 5.8 kg/s and a similar  $OC_{bio}$  flux of 12.6 kg/s from June 2012 to May 2013 (SPM flux: 5,723 kg/s, YRCC 2016).

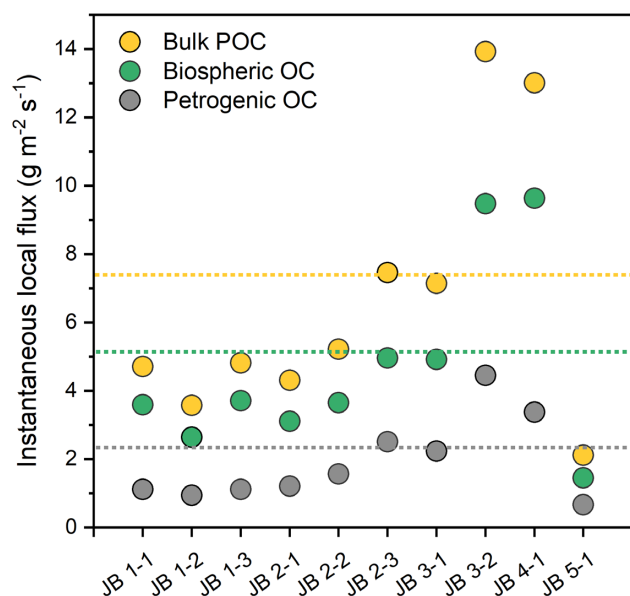
485 We first note that previous estimates of POC flux in the Huanghe might be biased as these estimates neglect the  
486 variability over the cross-section (*e.g.*, Hu et al., 2015; Ran et al., 2013; Tao et al., 2015), SPM samples analyzed so  
487 far for the Huanghe were generally collected within the first 0.5 m below the surface, meaning that previous POC  
488 estimates did not consider the observed vertical and lateral POC heterogeneities and have thus misestimated POC  
489 sources and fluxes. Those estimates were calculated by multiplying an individual surface POC content by the  
490 corresponding monthly or weekly suspended sediment load, as provided by hydrological stations. Such estimates can  
491 be problematic because POC content in SPM generally decreases from top to bottom (Figure 2), resulting in biased  
492 surface-based estimates of fluxes (Bouchez et al., 2014). Using our cross-section data, we can estimate the bias in  
493 POC flux estimates when a single sample is used for such flux estimates, by multiplying depth-integrated sediment  
494 flux by the POC content of each sample. Such calculation shows different POC fluxes ranging from  $-15\%$  to  $+30\%$   
495 compared to the depth-integrated estimate, which is mostly influenced by the variable POC content. Considering SPM  
496 collected at the channel surface, POC flux estimates using samples JB 1-1 and JB 2-1 are 28% and 15% higher,  
497 respectively, and are 6% and 5% lower using samples JB 3-1 and JB 5-1, respectively, than the depth-integrated  
498 estimate. This simple sensitivity analysis shows that channel surface sampling of SPM alone does not necessarily  
499 result in an overestimation of POC flux because of lateral heterogeneity, even though the POC content of SPM is  
500 generally higher at the surface than at the bottom (Figure 2). As a consequence, and although accurate estimation of  
501 POC fluxes requires grain-size variations to be accounted for, the corresponding bias cannot explain the large  
502 difference between our estimates of Huanghe POC export for the year 2016 and previous estimates for preceding  
503 years. The dramatic decrease in sediment load of the Huanghe (Wang and Fu et al., 2016) has most likely exerted a  
504 first-order control on the reduction in POC export from the Huanghe river system, and will probably continue to do  
505 so in the near future.

506 In the lower Huanghe, the POC content is very low and has small variance among different size fractions (Get et al.,  
507 2020), such that the POC flux is controlled by the SPM flux. In particular, it is worth noting that the Huanghe displays  
508 strong density stratification effects compared to other rivers (Moodie et al., 2022), with near-bed flow dominating the  
509 transport of SPM. In order to appraise how spatial and temporal variability in SPM flux could influence POC export,  
510 "local" POC loads can be calculated throughout the cross-section using the local water velocity, SPM concentration,



511 and POC content (Figure 8). In general, in the lower Huanghe more POC is transported near the riverbed and above  
 512 shallower bathymetry on the left side of the channel (except for profile JB 5). For instance, there is a nearly two-fold  
 513 increase in POC export from the surface to the bottom for the JB 2 and JB 3 profiles. The maximum local bulk POC  
 514 export (sample JB 3-2),  $OC_{bio}$  (sample JB 3-2), and  $OC_{petro}$  (sample JB 4-1) are over 6 times higher than the  
 515 corresponding minimum values (sample JB 5-1). This spatial pattern of POC load is almost the reverse of the POC%  
 516 variation over the cross-section, again stressing the importance of sediment river dynamics in POC delivery. From  
 517 these considerations, it could be anticipated that during the low-water season, when water velocity is slower, near-  
 518 bottom Huanghe SPM is deposited on the channel bed, withdrawing a significant fraction of the POC export, as shown  
 519 in other large rivers (Ke et al., 2022). This topic should be further examined in future research, in order to  
 520 systematically investigate the stratification of sediment and associated OC transport dynamics in lowland and high-  
 521 turbidity fluvial systems.

522



523

524 **Figure 8: Estimates of instantaneous “local” fluxes of Huanghe bulk POC,  $OC_{bio}$  and  $OC_{petro}$ , calculated for each sample of**  
 525 **the Luokou cross-section. The three dotted lines marked in orange, green, and grey represent the corresponding**  
 526 **instantaneous, cross-section integrated fluxes.**

527 Interestingly, anthropogenic activities may have antagonistic effects on POC export. Deforestation, agriculture, and  
 528 mining have considerably enhanced the sediment yield from the CLP since the mid-Holocene (He et al., 2006) while  
 529 the construction of large dams, soil and water conservation measures, and afforestation has considerably reduced the  
 530 sediment yield since the 1950s (Wang and Fu et al., 2016; Wang et al., 2007; Syvitski et al., 2005). Yet the Huanghe  
 531 exports substantial  $OC_{bio}$  and  $OC_{petro}$  with a significantly higher burial efficiency (~42% on average; Sun et al., 2018)  
 532 than other large fluvial systems entering passive continental margins, such as the Changjiang, Amazon, and  
 533 Mississippi (Blair and Aller, 2012). It is reported that aged soil OC is nearly fully preserved in continental margins  
 534 and that  $OC_{petro}$  has a *ca.* 70% burial efficiency (Tao et al., 2016). However, the contribution of the Huanghe OC burial





535 to the global C sink is likely to be lower in the future as the consequence of 1) sharp decrease in SPM and POC export  
536 due to weakened physical erosion in the CLP; 2) reduced sediment accumulation rate favoring OC remineralization  
537 in estuaries (Blair and Aller, 2012; Walling and Fan, 2003; Milliman and Farnsworth, 2011; Galy et al., 2015).

## 538 **6 Conclusions**

539 In this contribution, we present the first detailed study of particulate organic carbon (POC) over a complete river cross-  
540 section of the Huanghe, providing new perspectives on the transport mode, source, and instantaneous fluxes of POC  
541 in this highly turbid large river.

542 At the scale of a cross-section, physical and chemical properties of SPM are heterogeneous both vertically and  
543 laterally, a feature that is mainly controlled by bathymetry and hydrodynamic sorting. Resuspension of bed sediment  
544 and local erosion of the right bank together impact the suspended POC composition at the sampled location. This  
545 spatial heterogeneity shows that near-bottom SPM plays a dominant role in the delivery of OC<sub>bio</sub> (topsoil and deep  
546 soil OC combined) and OC<sub>petro</sub>. Despite a relatively shallow river channel (< 5.0 m) and narrow width (< 200 m), we  
547 show how the heterogeneity of POC transport over a cross-section needs to be considered in constraining POC  
548 transport mode and estimating POC fluxes.

549 Despite its millennial age, POC in the Huanghe is dominated by OC<sub>bio</sub> with a contribution of from 67% to 79%. OC<sub>petro</sub>  
550 content in SPM is relatively homogeneous (0.08% - 0.11%) over the cross-section, indicating that the variability in  
551 bulk POC age is mainly controlled by the variability in OC<sub>bio</sub> content, especially in the finest SPM fraction. OC<sub>bio</sub> ages  
552 deduced from the application of a mixing model to previously published data (record period 2011-2016) are highly  
553 variable, ranging from 1,040 to 8,050 <sup>14</sup>C yr. We interpret this feature as resulting from the erosion of deep horizons  
554 by gully systems in the loess-paleosol sequences containing <sup>14</sup>C-dead OC<sub>bio</sub>. Enhanced erosion of deep loess-paleosol  
555 horizons mobilizes aged and refractory OC to the ocean, with high burial efficiency on the passive margin. The erosion  
556 of loess-paleosol horizons is thus an efficient process of CO<sub>2</sub> burial. However, the construction of large dams has  
557 drastically affected the sediment load of the Huanghe system and retains substantial quantities of sediments that were  
558 previously exported to the ocean. Future work is needed to further quantify how these anthropogenic modifications  
559 alter POC composition and transport, by conducting comprehensive cross-section sampling campaigns over extended  
560 time series upstream and downstream from dams.

## 561 **Appendix A**

562 Fluvial POC delivered in the Huanghe POC could originate from three terrestrial sources. As topsoil typically contains  
563 recently photosynthesized OC<sub>bio</sub>, we used a  $\delta^{13}\text{C}$  value of  $-24.8 \pm 1.9\text{‰}$  (n=166) according to the subsurface soil OC  
564 values measured across the Huanghe basin (Rao et al., 2017). Over the sampled cross-section, the depleted <sup>13</sup>C values  
565 indicate the dominant and almost exclusive input of C3 plant-derived material to the Huanghe POC in the lower  
566 reaches. Based on <sup>14</sup>C (Liu et al., 2012) and <sup>10</sup>Be (Zhou et al., 2010) dating of < 10 cm-deep soil horizons in the  
567 Huanghe Basin, the average age of topsoil was chosen as being younger than 2,000 yr (*i.e.*,  $\Delta^{14}\text{C} > -220\text{‰}$ ). As the  
568 topsoil end member includes modern biospheric material ( $\Delta^{14}\text{C}$  around 40‰, Hua et al., 2013), we assigned a  $\Delta^{14}\text{C}$



569 value of  $-90 \pm 130\%$  ( $F_m = 0.91 \pm 0.13$ ) to this endmember. This range also includes the range of  $\Delta^{14}\text{C}$  values of pre-  
570 aged soil OC indicated by the long-chain  $n\text{-C}_{24+26+28}$  alkanols of the Huanghe POC reported by Tao et al. (2015) and  
571 Yu et al. (2019a). Their results show consistent POC  $\Delta^{14}\text{C}$  values in the lower reaches of  $-204 \pm 20\%$  ( $F_m = 0.80 \pm$   
572  $0.03$ ,  $n=7$ ) from June 2015 to May 2016 and  $-219 \pm 33\%$  ( $F_m = 0.79 \pm 0.04$ ,  $n=4$ ) at Kenli and of  $-198 \pm 15\%$  ( $F_m =$   
573  $0.81 \pm 0.02$ ,  $n=6$ ) from June 2015 to April 2016 at Huayuankou.

574 The second end member should be characterized by aged and refractory OC from the loess-paleosol sequence  
575 excluding topsoil (upper 10 m) of the CLP. Radiocarbon dating has an upper age limit of around 50,000 yr, age above  
576 which  $F_m$  is equal to 0. However, radiocarbon-free OC spanning from 50,000 to 100,000 yr must still be considered  
577 as  $\text{OC}_{\text{bio}}$  in the long-term carbon cycle. Here, we name this ignored OC as the “dormant” OC, without which the  $\text{OC}_{\text{bio}}$   
578 (*i.e.*, less than 100,000 yr old) would be underestimated to some extent because the radiocarbon-free OC would be  
579 misinterpreted as having a petrogenic origin. To consider this “dormant” OC, a  $\delta^{13}\text{C}$  values of  $-22.7 \pm 1.0\%$  and a  
580  $\Delta^{14}\text{C}$  values of  $-610 \pm 390\%$  ( $F_m, 0.39 \pm 0.39$ ) were adopted based on an average of values over the whole loess-  
581 paleosol sequence. Although radiocarbon-free (*i.e.*, older than 100,000 yr) OC overlaps with this endmember, such  
582 old soil organic carbon is probably not mobilized as modern gully erosion mainly concerns the upper 10 m of the  
583 loess-paleosol sequences, where soil  $\text{OC}_{\text{bio}}$  is assumed to be significantly younger comparatively (Figure 5).

584 Rock-derived OC from the QTP and the CLP, as well as kerogen from oil-gas fields from the Ordos Basin,  
585 were all considered to be possible contributors to the  $\text{OC}_{\text{petro}}$  endmember. The  $\delta^{13}\text{C}$  of  $\text{OC}_{\text{petro}}$  greatly varies between  
586 the QTP ( $-21.2 \pm 1.2\%$ ,  $n=11$ , Liu et al., 2007) and the CLP ( $-26.8 \pm 0.5\%$ ,  $n=8$ , Qu et al., 2020). However, most of  
587 the sediments eroded from the QTP are not transferred to the lower reaches as they remain trapped in the CLP and the  
588 western Mu Us desert (Nie et al., 2015; Licht et al., 2016; Pan et al., 2016). In addition, the construction of large dams  
589 in the upper reaches has considerably reduced the transfer of solid materials downstream (Wang et al., 2007).  
590 Therefore, rock-derived OC inherited from the denudation of the QTP region is not further considered. Kerogen from  
591 the oil-gas fields of the Ordos Basin in the CLP region (Figure 1) has  $\delta^{13}\text{C}$  values of  $-29.2 \pm 0.9\%$  ( $n=10$ , Guo et al.,  
592 2014). Taking these constraints together, we consider a  $\delta^{13}\text{C}$  value of  $-28.1 \pm 1.5\%$  for the  $\text{OC}_{\text{petro}}$  end member, and  
593 a  $\Delta^{14}\text{C}$  value of  $-1000\%$  ( $F_m = 0$ ) by definition.

#### 594 **Data availability**

595 All datasets are included in the paper and the supplementary materials.

#### 596 **Author contributions**

597 DC, JB, CQ, and YK conceptualized the study. DC, JB, YK, MM, and BC determined the methodology. HC and JC  
598 collected the sediment samples. YK, MM, and AN assisted with elemental and isotopic carbon analysis. DC and CQ  
599 supervised the work. KY performed data analysis and wrote the original draft, and all authors contributed to the review  
600 and editing of the paper.



601 **Competing interests**

602 We declare there is no competing interest.

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