

1 **Estuarine-deltaic controls on coastal carbon burial in the western Ganges-Brahmaputra**
2 **delta over the last 5,000 years**

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We welcome your feedback on this research; feel free to contact the authors via email (Rory.P.Flood@gmail.com, Georgina.Milne@afbini.gov.uk, G.Swindles@qub.ac.uk, I.Barr@mmu.ac.uk, J.Orford@qub.ac.uk). This is a non-peer-reviewed pre-print. Subsequent Revisions of this manuscript may include revisions based on feedback and the peer-review process. If accepted in a peer-reviewed journal, the final version of this manuscript will be available via the "Peer-reviewed Publication DOI" link on the EarthArXiv page for this paper.

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36 Keywords: Ganges-Brahmaputra delta; carbon; carbon burial; sedimentation; Sundarbans;
37 beta regression.

38

39 **Abstract**

40 The Ganges–Brahmaputra fluvial system drains the Himalayas and is one of the largest
41 sources of terrestrial biosphere carbon to the ocean. It represents a major continental reservoir
42 of CO₂ associated with c. 1–2 billion tons of sediment transported each year. Shallow coastal
43 environments receive substantial inputs of terrestrial carbon (900 Tg C yr⁻¹), with
44 allochthonous carbon capture on connected floodplains. Vegetated coastal ecosystems play
45 a dominant role in the sequestration of carbon and operate as highly efficient carbon sinks.
46 Mangrove sediments are subject to intense carbon-fixing processes that have a potentially

47 high impact on the global carbon budget. The Sundarbans is the largest tidal mangrove forest
48 in the world (10,200 km² in area) and is located on the marine-terrestrial boundary of the
49 Ganges-Brahmaputra delta and the Bay of Bengal, in West Bengal (India) and Bangladesh.
50 Estimates of sedimentation on the tidal delta plain of the Ganges-Brahmaputra delta reveal
51 mean rates of ~1.1 cm yr⁻¹ with accretion understood to approximately equal the regional rate
52 of sea-level rise of ~1.0 cm yr⁻¹. In this study, the properties of sediments from the western
53 Ganges-Brahmaputra delta are used to investigate controls on coastal carbon burial over the
54 past 5,000 years. Our main findings are: (1) Beta regression of aluminium and silica ratio data
55 is a robust method of estimating total organic carbon in sediment from the Indian Sundarbans;
56 (2) the estimated rate of sediment deposition over last 5,000 years is between 1.0 and 2.5 mm
57 yr⁻¹, with uncertainty surrounding the reworked origins of sediment; and (3) temporal variation
58 of total organic carbon accumulation through the last 5,000 years is generated by varying
59 sedimentary depositional processes. The delivery and burial of total organic carbon is
60 predicated on the continual supply of sediment to the Sundarbans, which future management
61 strategies may need to consider given changing rates of deposition.

62

63 **1. Introduction**

64 The world's oceans absorb atmospheric carbon dioxide (CO₂) at a rate comparable to that of
65 terrestrial ecosystems (Stocker et al., 2013; Watanabe and Kuwae, 2015). Coastal systems
66 are the interface between the land and the ocean and can act as carbon reactors that store,
67 process, and emit organic carbon (OC) (Butman et al., 2016; Galy et al., 2015, 2007; Hotchkiss
68 et al., 2015; Luisetti et al., 2019; Scott and Wohl, 2017; Sutfin et al., 2016). There is now
69 widespread recognition (Luisetti et al., 2019) that coastal environments themselves (e.g.,
70 mangroves, saltmarshes, and seagrass ecosystems) represent major globally-important
71 carbon sinks and stores (i.e., "Blue Carbon").

72 In estuarine coastal systems, the rate of OC burial is estimated to be higher (238 Tg C
73 yr⁻¹) than in the open ocean (6 Tg C yr⁻¹) (Nellemann et al., 2009; Watanabe and Kuwae,
74 2015). Shallow coastal environments also receive a substantial input of terrestrial carbon (C)

75 (900 Tg C yr⁻¹), with allochthonous carbon capture on floodplains (Regnier et al., 2013).
76 Vegetated coastal ecosystems play a key role in the sequestration of carbon particularly in
77 sea-grass meadows, mangrove forests, and tidal salt marshes as highly efficient C sinks
78 (Alongi, 2012; Bouillon et al., 2008; Bu et al., 2015; Chmura et al., 2003; Duarte et al., 2010,
79 2005; Iacono et al., 2008; Kennedy et al., 2010; Laffoley et al., 2009; Luisetti et al., 2019;
80 Mcleod et al., 2011; Nellemann et al., 2009). In such vegetated coastal ecosystems, long-term
81 Holocene (c. 8 – 10 ka years) rates of C accumulation in sediments range between 18 and
82 1713 g C m⁻² yr⁻¹ (Mcleod et al., 2011). In comparison long-term Holocene rates of C
83 accumulation in soils of temperate, tropical, and boreal forests range only between 0.7 and
84 13.1 g C m⁻² yr⁻¹ (Mcleod et al., 2011), emphasising the importance coastal ecosystems may
85 have for the sensitivity of contemporary and future C sequestration and the Earth's C cycle
86 more generally.

87 The Ganges–Brahmaputra (G–B) system drains the Himalayas and is one of the
88 largest sources of terrestrial biospheric carbon to the ocean. It therefore represents a major
89 continental reservoir of CO₂ (Galy et al., 2008; Galy and Eglinton, 2011). Approximately 1–2
90 billion tons of sediment are transported each year by the G–B fluvial system from the
91 Himalayas to the Bay of Bengal, with final deposition in the Bengal Fan (Galy et al., 2008).
92 Fluvial sedimentary characterisations of OC flux have the global significance of modern river
93 sources (Aucour et al., 2006; France-Lanord and Derry, 1997; Galy et al., 2008; Subramanian
94 and Ittekkot, 1991). In the Bengal Fan, modern burial flux of recent OC generated by Himalayan
95 erosion is $3.1 \pm 0.3 \times 10^{11}$ mol yr⁻¹ (Galy et al., 2008, 2007). This flux of OC represents c. 15%
96 of the global OC flux (Galy et al., 2008, 2007). OC burial in the Himalayan system is extremely
97 efficient, with nearly 100% of the OC exported by the G–B fluvial system buried in Bengal Fan
98 sediments (Galy et al., 2008). Thus, the G–B fluvial system plays a significant role in the
99 effective transport and burial of OC, which has major implications for atmospheric CO₂
100 sequestration (Galy et al., 2008).

101 Deltaic wetlands consist of one of the most dynamic landscapes and are under threat
102 by relative sea-level rise (RSLR) and decreases in fluvial sediment supply (Wagner et al.,

2017; Yang et al., 2020). The role of wetlands, particularly tidal wetlands, in carbon sequestration is well recognised with the possibility of incorporating carbon credits for tidal wetland restoration into the development of both voluntary and regulatory carbon markets for atmospheric CO₂ emissions reductions (Callaway et al., 2012; Crooks et al., 2010; Freedman et al., 2009; Hansen, 2009; Laffoley et al., 2009). Restoration of wetlands and ecosystem function could be tied with carbon credits in order to bury carbon (Callaway et al., 2012; Emmett-Mattox et al., 2011). The viable function of a carbon credit system would be primarily on the basis that approximately 100 years' worth of carbon could be buried, without the potential of carbon decomposition and return to the atmosphere (Callaway et al., 2012; Crooks et al., 2010). Determination of TOC burial in wetlands, particularly where restoration efforts have taken place, need baseline estimates of TOC to judge the efficacy of restoration and TOC burial (*cf.*, Callaway et al., 2012). The Indian Sundarbans offers a unique site to study burial of TOC over centennial to millennial timescales. The primary aim of this study is to quantify TOC in the Indian Sundarbans and examine the relationship between sedimentary processes in the accumulation of TOC.

118

119 **2. Materials and methods**

120 *2.1. Study Site*

121 The Sundarbans, split between India and Bangladesh (Fig. 1), is the largest tidal mangrove forest in the world at 10,200 km². It is located on the marine-terrestrial boundary of the G-B delta and the Bay of Bengal (Dutta et al., 2017). The Sundarbans was established as a UNESCO world heritage reserve site, with 60% of the reserved forest located in Bangladesh, 124 and 40% in India (Dutta et al., 2017). The Indian Sundarbans Biosphere Reserve (SBR), an area of c. 9,600 km², involves 1,800 km² of estuarine channels and 3,600 km² of reclaimed areas (Dutta et al., 2017). The Indian Sundarbans is dominated by a series of north-south oriented estuarine tidal channels (i.e., the Mooriganga, Saptamukhi, Thakuran, Matla, Bidya, 129 Gosaba, and Haribhanga), which define an archipelago of 102 islands, with 54 having been reclaimed for human settlement and the remainder uninhabited (Dutta et al., 2017). The overall

131 morphology of the Sundarbans indicates a west to east continuum of tidal, mixed tidal-fluvial,
132 and predominantly fluvial sedimentary processes operate (Flood et al., 2018; Rogers et al.,
133 2013). The overall G-B delta is divided into two key systems, the older fluvially-abandoned
134 part in the west, now tidally-dominated (the focus of this study), and the eastern, fluvially-
135 dominated system, with associated fluvially-driven shoreline progradation following the joining
136 of the Ganges and Brahmaputra Rivers (Allison, 1998a; Flood et al., 2018; Rogers et al.,
137 2013). The western extent of the delta, that underlies the present day Indian Sundarbans, was
138 fluvially abandoned prior to c. 5000 cal years BP, with the migration of the Ganges River
139 eastward towards its present position (Flood et al., 2018; Goodbred and Kuehl, 2000; Rogers
140 et al., 2013; Sarkar et al., 2009). Since this eastern migration of the Ganges, the Indian
141 Sundarbans is considered to have been principally estuary-tidal dominated.

142

143 *2.2. Sediment coring*

144 The study site is shown in Fig. 1, with an inlay map showing the extent of the West Bengal
145 Sundarbans. The primary surface sedimentary unit under investigation for TOC is the thin mud
146 facies (TMF); understood to be a Late Holocene (c. 5000 cal years BP to present) capping
147 unit of the Quaternary stratigraphy (Goodbred and Kuehl, 2000). The TMF consists of
148 overbank deposits of the modern and recent floodplain system, found in floodplain
149 environments and is absent near active fluvial channels (Goodbred and Kuehl, 2000). Coring
150 of the TMF sediment was conducted using a motor driven percussion coring device, with
151 latitude, longitude and elevation recorded with a differential GPS. Shallow percussion coring
152 was conducted at Lothian, Gplot, Dhanchi (2010), and Dhanchi-2, Bonnie Camp, and
153 Sajnekhali (2011) sites (Fig. 1). A detailed summary of these sites is provided in Flood (2014)
154 and Flood et al. (2018, 2016).

155 *2.3. ED-XRF geochemistry of the Sundarbans*

156 Data acquisition using energy dispersive – X-ray fluorescence spectrometry (ED-XRF) was
157 undertaken following the approach outlined by Flood et al. (2018, 2016), using a Bruker S1
158 TURBO SD portable X-ray fluorescence (PXRF) spectrometer (Bruker Corporation,
159 Massachusetts, USA) consisting of a 10 mm X-Flash® SDD Peltier-cooled detector with a 4-
160 W X-ray tube with an Ag target and a maximum voltage of 40 kV. Analysis was performed on
161 discrete samples collected from the Lothian, Gplot, Dhanchi, Dhanchi-2, Bonnie Camp, and
162 Sajnekhali Island cores. Precision and accuracy of the preparation and the instrumental
163 performance of the PXRF was checked using the international reference samples and a
164 summary of these is provided by (Flood et al., 2016). Al and Si values were log-normal (ln)
165 transformed for the purposes of TOC prediction using binomial logistic and beta regression
166 model coefficients.

167

168 *2.4. Development of TOC prediction for the Ganges-Brahmaputra Delta: modelling*
169 *approaches used*

170 There is a monotonic and simultaneous relationship between sediment properties and OC in
171 the G-B as identified by Galy et al. (Galy et al., 2015, 2011, 2008, 2007) and is understood to
172 result from OC content that is (1) mainly controlled by sediment properties, and/or (2) affected
173 by mixing and sorting processes, akin to mixing and sorting of detrital grains. Two species
174 modes of OC have been identified in the sediments of the G-B river and delta: (1)
175 organomineral associations; and (2) free organic particles. Within suspended sediments of the
176 G-B system, there is a tendency for segregation of free organic particles by size, in a similar
177 manner to detrital mineral grains; with fine grained sediments hosting mainly tiny free organic
178 particles, and coarse sediments hosting coarse organic debris. The most significant
179 relationship between sediment properties and TOC has been found by Galy et al. (2007), who
180 identified a high degree of correlation between TOC and the aluminium to silica (Al/Si) ratio
181 (Supplementary Figure B1). The Al/Si ratio represents the bulk mineralogical composition of

182 the sediments being a proxy for sediment size; low Al/Si values indicate high proportions of
183 quartz and low TOC, while high Al/Si indicate high proportions of micas and clay minerals, and
184 higher TOC (Galy et al., 2011, 2007).

185

186 *2.5. Estimation of TOC from Al/Si – the beta regression modelling approach*

187 The ultimate aim of this component of the analysis was to use the previously published TOC
188 and Al/Si ratio data by Galy et al. (2007) as a calibration dataset to derive estimates of TOC
189 values for the six sites in Figure 1. The Galy et al. (2007) data were selected for this purpose
190 based on both location relative to potential sediment inputs from the Ganges-Brahmaputra-
191 Meghna river systems; along with these samples being retrieved from contemporaneous
192 sediments of the TMF unit. This was achieved by fitting the original model between Al/Si and
193 TOC from Galy et al. (2007), and using this model to “predict” TOC at our six sites of interest.

194 Linear regression models using ordinary least squares (OLS) and maximum likelihood
195 estimation (MLE) are a generally accepted approaches used to examine the relationship
196 between an independent (predictor) variable and a dependent (outcome / response) variable.
197 However, these models pose serious challenges and are found to be inappropriate where the
198 response variable is restricted to the interval (0, 1); i.e., where the response variable is
199 constrained by a unit sum (Douma and Weedon, 2019; Ferrari and Cribari-Neto, 2004; Weltje
200 and Tjallingii, 2008). As a result of this imposed constraint, OLS estimation may yield fitted
201 values for the variable of interest that exceed both lower and upper bounds, and in some cases
202 produce values greater than 1, and <0, which is especially profound in cases where
203 measurements are low (Ferrari and Cribari-Neto, 2004; Weltje and Tjallingii, 2008). As a result,
204 where there may be a high degree of goodness-of-fit (e.g., $R^2 = 0.99$), such linear relations
205 are unlikely to be applicable outside the range of variation of the samples on which they are
206 based (Weltje and Tjallingii, 2008).

207 A common solution to these challenges include transformation of the dependent
208 variable so that data assumes values in real Euclidean space (\mathbb{R}^n), and then to model the

209 mean of the transformed response as a linear predictor. A number of issues are encountered
210 with this transformation approach, principally the fact that the model parameters cannot be
211 easily interpreted in terms of the original response. A better approach requires the use of
212 generalised linear models (GLMs) which are mathematical extensions of linear models that
213 allow for outcome data to vary from normality constraints (e.g., outcome data can assume
214 binomial, Poisson, negative binomial, or gamma distribution) (Guisan et al., 2002). In this
215 instance, the binomial model was used to model outcomes which vary between 0 and 1, but
216 with the limitation that such models are usually more applicable for modelling fractional counts
217 e.g. 1/5, 2/5. TOC data, however, is continuous and thus represents a “true” proportion e.g.
218 “20%”, as opposed to a fractional proportion. Beta regression (Douma and Weedon, 2019;
219 Ferrari and Cribari-Neto, 2004) provides some additional flexibility to model these types of
220 data. Furthermore, measures of proportions are generally asymmetric, and inferences based
221 on the normality assumption can be misleading as a result (Ferrari and Cribari-Neto, 2004)
222 making the beta distribution more appropriate for these data. TOC was reported as a
223 percentage, but was easily converted into proportions (prop.) by dividing by 100 prior to
224 modelling. Whilst we fit the Galy et al. (2007) data using the binomial distribution and beta
225 distribution, the primary focus of this work are models fitted using the beta distribution, and
226 the majority of the results will refer to this. The binomial models are, however, presented for
227 the sake of comparison.

228 Binomial logistic regression was carried out using the ‘stats’ package in R Version 4.1.2
229 (Core Team, 2021), with a logistic link function and the binomial model family. Beta regression
230 was carried out using the ‘betareg’ package by (Cribari-Neto and Zeileis, 2010) in R Version
231 4.1.2 (Core Team, 2021). Linear regression of predicted TOC against depth, along with
232 graphical illustration of binomial logistic regression and beta regression results was carried
233 out using the ‘ggplot2’ package (Wickham, 2016) in R Version 4.1.2 (Core Team, 2021).

234 2.6. Grain size analysis

235 Given the importance of the Al/Si ratio for determination of TOC, and given the relationship
236 between sediment properties and/or mixing and sorting processes, grain-size distributions
237 (GSDs) from the West Bengal Sundarbans were analysed following Flood et al. (2016, 2015,
238 2018) using a Malvern Mastersizer 2000™ instrument. Zeros were replaced with a small
239 constant (0.001), with a sensitivity analysis carried out against other small constants (i.e., 0.01
240 and 0.1) showing no discernible variation in median grain-size (i.e., D_{50} μm). GRADISTAT
241 software was used for the analysis of grain-size data in order to calculate grain size statistical
242 parameters, with median grain-size in microns (μm), and silts and clays (i.e., $< 63 \mu\text{m}$)
243 presented from all sites examined (Blott and Pye, 2001).

244 *2.7. Radiocarbon dating and age-depth modelling*

245 Accelerator mass spectrometry (AMS) radiocarbon (^{14}C) dating was carried out in the
246 ^{14}C CHRONO Centre (Queen's University Belfast) to establish the chronostratigraphy of the
247 cores from the Lothian, Gplot, Dhanchi, Dhanchi-2, and Sanekhali (see Table 1).
248 Approximately 3 g of sediment was extracted for each of the samples for ^{14}C AMS radiocarbon
249 dating. Each sample underwent pre-treatment involving an acid wash (HCl) to eliminate
250 carbonates. Both bulk sediment and humic acid extraction was carried out for dating, with
251 humic acid extraction methodology based on that of Lowe et al. (2004). All dates were
252 calibrated against the IntCal13 curve (Reimer et al., 2013) with calibration carried out using
253 the 'Clam' package by Blaauw (2020, 2010) in R Version 4.1.2 (Core Team, 2021). In order to
254 derive rates of sedimentation for these sites, age-depth modelling through the 'Bacon'
255 package in R was used to produce Bayesian age-depth models (Blaauw and Christen, 2011).
256 Bacon calibrates ^{14}C dates against a specified ^{14}C calibration curve, which in this study was
257 IntCal13. Bayesian age-depth modelling was utilised using default settings in Bacon, and
258 these are summarised in Supplementary Figures B8-B11. Accumulation rates (mm yr^{-1}) are
259 presented from the Lothian and Sajnekhali Island cores (see Fig. 7); three age-depth and
260 accumulation models were produced for Sajnekhali due to the presence of age -reversals in
261 the core. Age-depth modelling was not carried out on Dhanchi, Dhanchi-2, and Gplot due to
262 numerous age-reversals and lack of dates. All Bayesian modelled dates are presented in
263 Supplementary Tables C2-C5.

264 3. Results

265 3.1. Binomial logistic and beta regression GLM of TOC

266 The binomial logistic and beta regression models were applied to the TOC (prop.) and Al/Si
267 data from Galy et al (2007) (see Supplementary Table 1). These input data consisted of
268 surface sediments taken from Harding Bridge (n = 28), Sirajganj (n= 32), Mawa (n = 6), and
269 Bhola (n = 13). The results from the binomial logistic regression GLM applied to the Galy et
270 al. (2007) data are shown in Fig. 2, with a pseudo R² value of 0.76. Exploratory modeling
271 revealed under-dispersion in the data (i.e. less variance in the outcome than expected). Here,
272 the dispersion parameter was found to be 0.00019 indicating a high level of under-dispersion;
273 a dispersion parameter of 1 suggests no under or over dispersion in the data. The model was
274 therefore refitted using quasi-binomial distribution, which has no effect on the coefficient
275 estimate, but in this case, reduced the standard errors (Zuur, 2009). We hypothesize that the
276 primary cause of under-dispersion may be attributed to small sample values (Lord and
277 Guikema, 2012; Sellers and Morris, 2017). The Beta regression is also presented (Fig. 3), with
278 a pseudo R² of 0.82.

279

280 3.2. Predicted TOC from the Sundarbans

281 Predicted TOC from the Sundarbans plotted against depth are shown in Fig. 4. Median TOC
282 in the sites ranged between 0.322% (IQR 0.269 – 0.445%) in Lothian; 0.320% (IQR 0.176 –
283 0.422%) in Gplot; 0.307% (IQR 0.256 – 0.354%) in Dhanchi; 0.275% (IQR 0.254 – 0.295%)
284 in Dhanchi-2; 0.288% (IQR 0.267 – 0.363%) in Bonnie Camp; and 0.275% (IQR 0.244 –
285 0.319%) in Sajnekhali. The data from the Indian Sundarbans fall within the range of TOC
286 values found in the Galy et al. (2007) data (i.e., median TOC at 0.46%, IQR: 0.275 – 0.575%,
287 with a minimum and maximum range of 0.03% and 0.82%, respectively).

288

289 3.3. Grain-size analysis results from the Sundarbans

290 Median (D₅₀) grain-size from the Sundarbans are shown in Fig. 5 with grain size variability
291 being composed predominately of the silt-sized fraction (4 – 63 µm), except for the Gplot core
292 which has fine to medium sand. Grain-size data from the Sundarbans reveal an overwhelming

293 majority of the sediment size fractions fall within the silt-size fraction (i.e., 4 – 63 μm) (Fig. 5).
294 The limited grain size variability found in the Sundarbans is reflective of the homogeneous
295 silts and clayey silts of most cores. The mud fraction (i.e., silt and clay, $<63 \mu\text{m}$) is plotted
296 against predicted TOC in the Sundarbans (Fig. 6) where a relatively strong relationship is
297 found between mud and TOC in Lothian (Coefficient (Coef.) = -46.307 ; Standard Error (Std.
298 E) = 4.793 ; $p = <0.001$; model $R^2 = 0.56$) and Dhanchi (Coef. = -60.265 ; Std. E = 6.502 ; $p =$
299 <0.001 ; model $R^2 = 0.55$). A weaker relationship between mud and TOC is found in Dhanchi-
300 2 (Coef. = -58.327 ; Std. E = 8.031 ; $p = <0.001$; model $R^2 = 0.25$) and Sajnekhali (Coef. = $-$
301 35.613 ; Std. E = 4.559 ; $p = <0.001$; model $R^2 = 0.26$); with no discernible relationship found in
302 Gplot (Coef. = -10.336 ; Std. E = 21.409 ; $p = 0.636$; model $R^2 = 0.01$) and Bonnie Camp (Coef.
303 = -6.488 ; Std. E = 2.789 ; $p = <0.05$; model $R^2 = 0.02$). Linear regression of predicted TOC
304 against depth for all sites (Fig. 7) showed there to be relatively strong correlations in Lothian
305 (Coef. = -1460.09 ; Std. E = 96.00 ; $p = <0.001$; model $R^2 = 0.74$), Gplot (Coef. = -591.64 ; Std.
306 E = 83.54 ; $p = <0.001$; model $R^2 = 0.53$) and Dhanchi (Coef. = -1776.96 ; Std. E = 239.68 ; $p =$
307 <0.001 ; model $R^2 = 0.44$); with weaker correlations in Dhanchi-2 (Coef. = -1915.70 ; Std. E
308 = 507.6 ; $p = <0.001$; model $R^2 = 0.081$), Bonnie Camp (Coef. = 1108.01 ; Std. E = 204.24 ; $p =$
309 <0.001 ; model $R^2 = 0.114$), and Sajnekhali (Coef. = 655.41 ; Std. E = 238.96 ; $p = <0.05$; model
310 $R^2 = 0.041$).

311 3.4. Radiocarbon (^{14}C) chronology

312 Table 1 shows the ^{14}C derived chronology from the Sundarbans; due to the nature of age-
313 reversals in the Dhanchi and Dhanchi-2 core data, and the limited number of samples from
314 Gplot ($n = 2$), further age-depth modelling was not carried out on these samples. LOESS-
315 smoothed spline interpolation of the sedimentation rates (mm yr^{-1}) was plotted against depth
316 for Lothian and Sajnekhali cores based on the Bayesian age-depth models (Supplementary
317 Figures B9-B12) and these are presented in Fig. 8. The median sedimentation rate in Lothian
318 was 1.67 mm yr^{-1} (IQR $1.25 - 2.00 \text{ mm yr}^{-1}$), with median sedimentation rates in Sajnekhali
319 ranging from $1.25 - 2.00 \text{ mm yr}^{-1}$.

320

321 4. Discussion

322 4.1. Prediction of TOC from the Indian Sundarbans

323 Using the Al/Si and TOC quantification from Galy et al. (2007) as a 'calibration' dataset, we
324 predicted TOC at six sites in the Sundarbans (this study) which revealed comparable levels of
325 TOC to those reported by Galy et al. (2007). The overall range of predicted TOC would strongly
326 indicate that terrestrial sources of organic matter (OM) are the main source of TOC in the
327 Indian Sundarbans. It has been found that $\delta^{13}\text{C}$ compositions in the Sundarbans are
328 comparable within the global range of mangroves and other coastal ecosystems (Prasad et
329 al., 2017). Sedimentary OM is primarily influenced by terrestrial sources, particularly inputs
330 from C_3 and C_4 metabolic plant pathways for both the Indian and Bangladeshi Sundarbans
331 (Prasad et al., 2017). Plant litter, particulate, and suspended OM are sourced primarily from
332 terrigenous matter, phytoplankton and marine particulates, as the dominant OM end-members
333 in coastal ecosystems (Bala Krishna Prasad and Ramanathan, 2009; Gonnee et al., 2004;
334 Prasad et al., 2017). For the first time (to the authors knowledge), the beta regression
335 approach was used to model geochemical data, thus overcoming the challenges posed by
336 compositional data, particularly the unit sum constraint. The advantages of the beta regression
337 can also be seen in model flexibility in accounting for asymmetries in distributions and

338 proportions; with Gaussian approaches found to be inaccurate in these respects (Cribari-Neto
339 and Zeileis, 2010). Whilst Galy et al. (2007) used a linear regression model to fit the TOC and
340 Al/Si data (Supplementary Figure 1), beta regression (Fig. 3) is the most appropriate approach
341 to model “true” proportional data and its error structure (Cribari-Neto and Zeileis, 2010; Douma
342 and Weedon, 2019).

343

344 *4.2. TOC and grain-size variability in the Indian Sundarbans*

345 There is a well-established relationship between grain size and OM, in that the OM of
346 continental margin sediments increases concomitantly with finer grain size with clays (i.e., <
347 4 μm) found to have twice as much OM as silts (i.e., 4 – 63 μm), and four times as much OM
348 as in fine sands (i.e., 63 – 250 μm), Trask (1939). The proposed mechanism for this
349 relationship is the similarity of settling velocity (i.e., hydrodynamic or 'hydraulic' equivalence)
350 of particulate OM and the fine-grained detrital minerals (Buchanan and Longbottom, 1970;
351 Trask, 1939; Tyson, 1995). Furthermore, OM is also associated with TOC, with OC controlled
352 by sediment properties, and/or impacted by mixing and sorting processes, similar to mixing
353 and sorting of detrital grains (Galy et al., 2015, 2011, 2008, 2007). The results here show that
354 grain-size data from the Sundarbans reveal an overwhelming majority of the sediment size
355 fractions fall within the silt-size fraction (i.e., 4 – 63 μm), reflective of the homogeneous silts
356 and clayey silts characterised for the surface sediments of the lower western tide-dominated
357 delta plain, compared to that found on the eastern fluvial-dominated delta (Allison et al., 2003).
358 Thus, hydrodynamic conditions, and sediment grain size, play a determinant role in the
359 deposition of OM and associated TOC (Tyson, 1995). We observed results in agreement with
360 this interpretation at Lothian and Dhanchi. However, the relationship was less clear at
361 Dhanchi-2 and Sajnekhali, and was not observed at all in the Bonnie Camp and Gplot data.
362 We hypothesise that between-site differences could be driven by various processes of mixing
363 of sand mud fractions under complex hydrodynamic processes within the Indian Sundarbans
364 (*cf.*, Venkatraman et al., 2013). We posit that this complex mixing process may be site
365 specific, given the overall variability of predicted TOC with depth (Fig. 7). Notwithstanding this

366 variation, our predicted TOC results are in relative agreement with previous studies (Allison et
367 al., 2003), with reported ranges from 0.05% to 1.1%.

368 We argue that the variation of D_{50} grain size and TOC in the Indian Sundarbans may
369 reflect a number of non-mutually exclusive processes. Our data primarily support the
370 explanation that the observed between-site differences may reflect the increasingly reworked
371 nature of the sediments (Allison et al., 2003; Flood et al., 2018; Rogers et al., 2013). Sediments
372 in the Sundarbans are considered to be reworked from G-B sourced muds from the delta front
373 (*cf.*, Allison et al., 2003; Rogers et al., 2013), and are understood to be delivered through
374 onshore advection during monsoonal and cyclonic coastal setup events (*cf.*, Allison and
375 Kepple, 2001; Flood et al., 2018). TOC is both inversely correlated with sand content and the
376 sand-mud ratio (*cf.*, Bornhold and Yorath, 1984; Syvitski et al., 1990; Tyson, 1995) and TOC
377 may also be inversely correlated with mean size of the silt fraction (Scheidegger and Krissek,
378 1983; Tyson, 1995). In contemporary sediments the maximum level of TOC is generally better
379 correlated with grain size than in the minimum TOC fraction (Gross, 1967; Romankevich and
380 Shirshov, 2013; Tyson, 1995). The correlation between grain size and TOC may partly reflect
381 the greater surface area of finer particles, in a form of sediment-specific surface area (*cf.*,
382 Tyson, 1995, pp. 87), which is related to the amount of organic carbon that may be absorbed
383 on sediment surfaces (Hedges et al., 1993; Keil and Hedges, 1993). Alternatively, it is known
384 that downcore decreases in TOC content are indicative of rapid sediment respiration rates
385 with burial (Allison et al., 2003). In the active progradational lower delta plain sediments, TOC
386 preservation is low, relative to marsh and mangrove deposits in the more inactive delta
387 complex (Allison et al., 2003).

388 Alternative explanations for temporal variability in TOC throughout the Sundarbans
389 may be related to the influence of seasonal fluctuations between saline flooding and rainwater
390 flushing (Allison et al., 2003). Strong differences associated with poorer drainage and organic
391 preservation are found to coincide with higher OM preservation in parts of the Sundarbans
392 that tend to dry out between annual floods (Kosters, 1989; J. M. Coleman, 1966; Allison et al.,
393 2003). In the Sundarbans sediments OM breakdown is facilitated by the relatively high

394 permeability of the silt-dominated sequence (*cf.*, Allison et al., 2003). Lastly, Tidal
395 morphodynamics play a significant role in the determination of sediment balance within an
396 estuarine system, primarily channel geometry in the form of channel convergence into funnel
397 planimetric shape and sloping characteristics of the bed, and the degree of sinuosity; these
398 morphodynamic factors control tide propagation along the channel (Lanzoni and Seminara,
399 2002). Net sediment transport, in terms of magnitude and direction, over the course of a tidal
400 cycle, affects the tidal asymmetry, leading to unequal duration and/or unequal magnitude of
401 ebb and flood tides (Dronkers, 1986; Lanzoni and Seminara, 2002; Townend and Pethick,
402 2002). Tidal flow asymmetries that are characterized in the form of shorter flood duration and
403 higher flood current maximum (i.e., flood dominance) will lead to landward directed sediment
404 transport; with shorter fall periods and greater ebb current (i.e., ebb dominance) leading to net
405 seaward directed sediment transport (Dronkers, 1986; Lanzoni and Seminara, 2002; Townend
406 and Pethick, 2002). Whilst it is not yet clear, morphodynamic process may be influencing the
407 observations at Bonnie Camp and Sajnekhali.

408

409 *4.3. Sedimentation rates and burial of TOC*

410 The inconsistency of ¹⁴C ages throughout the sites dated in the Indian Sundarbans (Table 1)
411 likely reveals a complicated sedimentary environment (*cf.*, Nian et al., 2018). The introduction
412 of old C may have taken place with sediment storage and reworking along the dispersal path
413 from the delta plain to delta mouth as a main cause of older dates and age reversals (*cf.*,
414 Stanley and Chen, 2000). Self-cannibalisation of tidally-deposited sediments is understood to
415 be most important in the Sundarbans, particularly in the eastern delta complex with regards
416 reworking of sediments (Allison, 1998b; Woodroffe, 2010). Radiocarbon dates of the Holocene
417 sequence of the Indian Sundarbans need to be complimented with a multi-method
418 geochronological approach, such as optically stimulated luminescence (OSL) dating quartz
419 silt (Chamberlain et al., 2017). Although tidally reworked sediments, may prove too challenging
420 for OSL dating (Chamberlain et al., 2017). The assessment of the uncertainties involved in the
421 reconstruction of the sedimentation rates from the Indian Sundarbans is both dynamic and

422 complex, and highlights the limitations of ^{14}C dating methods in this environment (*cf.*, Nian et
423 al., 2018).

424 The Indian Sundarbans however is considered to be sediment starved, eroding, and
425 cut-off from major distributary sources of sediment (Flood et al., 2018). Estimates of mid-late
426 Holocene sedimentation on the most eastern extent of the tidal delta plain in Bangladesh
427 reveal mean rates of $\sim 1.1 \text{ cm yr}^{-1}$, with accretion attributed to a mix of flood pulse sediment
428 and reworking of older sediments from the shelf or tidal channel beds (Allison and Kepple,
429 2001; Rogers et al., 2013). Accretion rates due to this net sedimentation are approximately
430 equal to the mean regional rate of relative sea-level rise (RSLR) of $\sim 1.0 \text{ cm yr}^{-1}$ (Rogers et
431 al., 2013). Accretion is complicated by varying estimates of subsidence rates (*i.e.*, increasing
432 water level) across the delta plain at *c.* $\sim 3.0 \text{ mm yr}^{-1}$ (Becker et al., 2020, and references
433 therein).

434 The sedimentation rate in the Sundarbans is higher during the wet monsoon season
435 (*i.e.*, June-October), with greater frequency and deeper inundation of island sites carrying
436 suspended sediment loads (Hale et al., 2019). The primary future challenge facing the Indian
437 Sundarbans is the sustainable delivery of sediment and accretion in the tide-dominated
438 mangrove sites (Hale et al., 2019). Reduction in mangrove areas worldwide in recent decades
439 may have consequences for OM exchange at the land-ocean boundary (Jennerjahn and
440 Ittekkot, 2002). In the case of the Sundarbans, possible worst-case scenarios for reduced
441 sediment delivery range between 14–18%, along with an increase in the duration of the dry
442 season (*i.e.*, November-May) by one month or more has the potential to reduce overall
443 sediment deposition by approximately half of what is presently observed (Hale et al., 2019).
444 Such a worst-case scenario results in a sedimentation rate that is below the rate of local sea
445 level rise (*cf.*, Hale et al., 2019), resulting in a threat to the continued viability of the
446 Sundarbans as a sustainable form of C burial. The successful burial of atmospheric C in the
447 Sundarbans is predicated on the successful accretion and sustainability of sediment delivery
448 which at present looks more precarious.

449

450 **5. Conclusions**

451 In this study we explored the variation of TOC in the Indian Sundarbans from six cores
452 collected from five sites. The main study findings are:

453 (1) The data from Galy et al. (2007) in combination with the beta regression modelling
454 approach, has given reliable predictions for TOC for the Indian Sundarbans. TOC has
455 varied across sites, with accumulation rates coincident with accretion across sites.

456
457 (2) Radiocarbon (^{14}C) dating has shown the variability of sedimentation rates over the
458 last 5,000 years; with sedimentation found to be greatest at Sajnekhali relative to
459 Lothian – with uncertainty of ^{14}C dates associated with the reworked nature of delta
460 sediments found in this study.

461
462 (3) Variation in TOC over the last 5,000 years has illustrated the role of sediment
463 depositional processes in controlling TOC burial, with TOC found to accumulate
464 alongside decreasing grain-size. The observed between-site differences reflecting the
465 increasingly reworked nature of the sediments associated with tidal advection as the
466 primary driver of TOC delivery.

467
468 The study has found that TOC burial and sedimentation are intimately related in the Indian
469 Sundarbans, but with TOC burial reliant on the sustainable delivery of detrital sediment to
470 island sites, future rates of TOC will be ultimately predicated on successful retention of tidal
471 sedimentation in the context of flood risk management using embankments to resist further
472 marine incursions on inhabited tidal islands (Adnan et al., 2020).

473

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485

486 **Author contributions**

487 RPF developed the study concept, JDO obtained funding for field work, RPF and JDO carried
488 out field work jointly, and RPF carried out all laboratory analyses as part of PhD study. MGM
489 developed the binomial logistic and beta regression methods for TOC prediction. GTS carried
490 out Bayesian age-depth modelling and manuscript review; IDB edited figures and manuscript
491 text for publication. The first manuscript draft was written by RPF and revised by all co-authors.
492 All authors contributed to the intellectual content expressed herein.

493

494 **Declaration of Competing Interest**

495 The authors declare that there is no conflict of interest.

496

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780

List of figures

Fig. 1. Location of the Ganges-Brahmaputra Delta, with inlay map showing location of Hardinge Bridge (Ganges), Sirajganj (Brahmaputra), Mawa, and Bhola (Lower Meghna) sampled by Galy et al. (2007), with Lothian, Gplot, Dhanchi, Dhanchi-2, Bonnie Camp, and Sajnekhali (this study).

Fig. 2. Binomial logistic regression GLM applied to Galy et al. (2007) TOC (prop.) and $\ln(\text{Al/Si})$ data with 95% CI ribbon. Pseudo R^2 value of the binomial logistic regression GLM = 0.76.

Fig. 3. Beta regression (pseudo R^2 value of beta regression = 0.82) with quantiles (2.25% and 97.5% plotted as a ribbon around model). As a result of the beta distribution, on which the model predictions are made, being skewed (i.e., the data are fitted to the beta distribution to account for the fact that they cannot be >1.0 or <0.0), standard deviation and confidence intervals used in linear models cannot be used here; instead quantiles of the predicted beta distribution are used here to show the predicted range of values.

Fig. 4. Predicted TOC in percentage format (%) from; (a) Lothian, (b) Gplot, (c) Dhanchi, (d) Dhanchi-2, (e) Bonnie Camp, and (f) Sajnekhali (beta regression model coefficients used).

Fig. 5. Median grain-size (D_{50} μm) from: (a) Lothian, (b) Gplot, (c) Dhanchi, (d) Dhanchi-2, (e) Bonnie Camp, and (f) Sajnekhali. The overall median grain size in Lothian was 38.12 μm (IQR 32.69 – 42.05 μm), Gplot at 92.18 μm (IQR 31.44 – 151.49 μm), Dhanchi at 28.19 μm (IQR 25.69 – 31.58 μm), Dhanchi-2 at 31.23 μm (IQR 28.27 – 32.62 μm), Bonnie Camp at 31.60 μm (IQR 30.32 – 32.73 μm), and Sajnekhali at 33.39 μm (IQR 31.26 – 35.86 μm).

Fig. 6. Linear regression of how predicted TOC changes with median size fraction for all sites (blue line), where $D_{50} < 63 \mu\text{m}$; (a) Lothian, (b) Gplot, (c) Dhanchi, (d) Dhanchi-2, (e) Bonnie Camp, (f) Sajnekhali; a trend of increasing TOC with decreasing size is found (a, c, d, f) with no strong relationship found in two sites (b & e).

Fig. 7. Linear regression of predicted TOC against depth for all sites (blue line); (a) Lothian, (b) Gplot, (c) Dhanchi, (d) Dhanchi-2, (e) Bonnie Camp, (f) Sajnekhali; a trend of increasing TOC is found from the bottom to the top of cores (a-d), and a decreasing TOC trend from the bottom to the top of cores (e & f).

Fig. 8. LOESS-smoothed spline interpolation of sedimentation rates (mm yr^{-1}) for Lothian (a) and Sajnekhali (b-d) based on Bayesian age-depth model. The median sedimentation rate (mm yr^{-1}) in Lothian is 1.67 mm yr^{-1} (IQR $1.25 - 2.00 \text{ mm yr}^{-1}$). Three age-depth models (Supplementary Figures B9, B10, B11) were carried out on Sajnekhali due to the presence of age-reversals; median sedimentation rates range from 2.00 mm yr^{-1} (IQR $1.43 - 2.16 \text{ mm yr}^{-1}$) (b), 2.00 mm yr^{-1} (IQR $1.43 - 2.50 \text{ mm yr}^{-1}$) (c), 1.25 mm yr^{-1} (IQR $1.00 - 2.50 \text{ mm yr}^{-1}$) (d).

List of tables

Table 1: ^{14}C chronology from the Sundarbans.

Appendix A. Supplementary material

Supplementary Material A1. R Markdown for Estuarine-deltaic controls on coastal carbon burial in the western Ganges-Brahmaputra delta over the last 5,000 years (HTML).

Supplementary Material A2. R Markdown for Estuarine-deltaic controls on coastal carbon burial in the western Ganges-Brahmaputra delta over the last 5,000 years (PDF).

Supplementary Material A3. R Markdown Code for Supplementary Material A1, A2 (RMD File).

Supplementary Table A4. Sundarbans Al/Si data file used in Supplementary Material A3 (.csv file).

Supplementary Table A5. TOC data from Galy et al. (2007) used in Supplementary Material A3 (.csv file).

Appendix B. Supplementary figures

Supplementary Figure B1. TOC for sediments of the Ganges (filled blue symbols), the Brahmaputra (filled green symbols), Lower Meghna (filled orange symbols) as a function of Al/Si (Galy et al., 2007). A positive linear relationship ($R^2 = 0.89$, $p \leq 0.001$) with sediments from all three rivers having a similar positive trend between TOC and Al/Si, indicative of similar OC loadings (Galy et al., 2007). In sandy and quartz-enriched bed sediments, TOC is found to be very low (circles); TOC linearly increases with the relative proportion of aluminium-enriched fine-grained minerals found in the suspended sediments (squares) (Galy et al., 2007). Sediments from the three rivers show a comparable positive trend, indicative of the similar OC loading relative to their composition (Galy et al., 2007). Best fit (solid black line) and 95% confidence interval (CI: dashed black lines) are shown in the figure (after Galy et al., 2007).

Supplementary Figure B2. Median grain-size (D_{50} μm) from: (a) Lothian, (b) Gplot, (c) Dhanchi, (d) Dhanchi-2, (e) Bonnie Camp, and (f) Sajnekhali.

Supplementary Figure B3. $< 63 \mu\text{m}$ grain-size fraction from: (a) Lothian, (b) Gplot, (c) Dhanchi, (d) Dhanchi-2, (e) Bonnie Camp, and (f) Sajnekhali.

Supplementary Figure B4. Linear regression of how predicted TOC changes with median size fraction (D_{50}) for all sites (blue line); (a) Lothian ($R^2 = 0.54$), (b) Gplot ($R^2 = 0.52$), (c) Dhanchi ($R^2 = 0.55$), (d) Dhanchi-2 ($R^2 = 0.25$), (e) Bonnie Camp ($R^2 = 0.02$), (f) Sajnekhali ($R^2 = 0.05$).

Supplementary Figure B5. Relationship between the TOC and clay fraction ($< 4 \mu\text{m}$) for all sites; (a) Lothian, (b) Gplot, (c) Dhanchi, (d) Dhanchi-2, (e) Bonnie Camp, (f) Sajnekhali.

Supplementary Figure B6. Relationship between the TOC and silt fraction (4 – 64 μm) for all sites; (a) Lothian, (b) Gplot, (c) Dhanchi, (d) Dhanchi-2, (e) Bonnie Camp, (f) Sajnekhali.

Supplementary Figure B7. Relationship between the TOC and mud (i.e., silt and clay fraction, < 63 μm) for all sites; (a) Lothian, (b) Gplot, (c) Dhanchi, (d) Dhanchi-2, (e) Bonnie Camp, (f) Sajnekhali.

Supplementary Figure B8. Linear regression of predicted TOC against depth for all sites; Lothian ($R^2 = 0.74$), Gplot ($R^2 = 0.53$), Dhanchi ($R^2 = 0.44$), Dhanchi-2 ($R^2 = 0.08$), Bonnie Camp ($R^2 = 0.11$), Sajnekhali ($R^2 = 0.04$); Lothian, Gplot, Dhanchi, and Dhanchi-2 show a trend with rising TOC towards core surfaces, with Bonnie Camp and Sajnekhali showing reverse trends in predicted TOC values.

Supplementary Figure B9. Bayesian age-depth models constructed with the 'Bacon' age-depth modelling package in R for Lothian Island core ^{14}C dates. Bayesian age-depth modelling utilised default settings within 'Bacon' (i.e., a piece-wise linear model with 5 cm sections, a gamma prior for sedimentation rate with a mean of 20 and shape 1.5, a beta prior for memory with mean of 0.7 and strength of 4, and a student-t distribution to deal with outlying dates). All modelled dates are presented in Supplementary Table 2. The grayscale on the model represents the likelihood, where darker gray indicates the greater likelihood that the model ran through that section of the core. The top panel shows three plots for each model: the left panel plot shows the model stability; the middle plot shows the prior (green line) and posterior (gray filled) distributions of accumulation mean; and the panel plot on the right shows the prior (green line) and posterior (gray filled) distributions of memory properties.

Supplementary Figure B10. Bayesian age-depth model constructed with the 'Bacon' age-depth modelling package in R for Sajnekhali Island core. All ^{14}C dates from Sajnekhali (see Table 1) were included in this model run. Mean 95% confidence ranges 380 yr, min. 61 yr at 0 cm, max. 506 yr at 145 cm; 62% of the dates overlap with the age-depth model (95% ranges). All modelled dates are presented in Supplementary Table 3.

Supplementary Figure B11. Bayesian age-depth models constructed with the 'Bacon' age-depth modelling package in R for Sajnekhali Island core. This model run was performed on a subset of ^{14}C dates from Sajnekhali (i.e., UBA-22971, UBA-22968, UBA-22967, UBA-22966, see Table 1). Mean 95% confidence ranges 514 yr, min. 58 yr at 0 cm, max. 701 yr at 275 cm 100% of the dates overlap with the age-depth model (95% ranges). All modelled dates are presented in Supplementary Table 4.

Supplementary Figure B12. Bayesian age-depth models constructed with the 'Bacon' age-depth modelling package in R for Sajnekhali Island core. This model run was performed on a subset of ^{14}C dates from Sajnekhali (i.e., UBA-22972, UBA-22970, UBA-22969, see Table 1). Mean 95% confidence ranges 558 yr, min. 57 yr at 0 cm, max. 724 yr at 180 cm, with 75% of the dates overlap with the age-depth model (95% ranges). All modelled dates are presented in Supplementary Table 5.

Appendix C. Supplementary tables

Supplementary Table C1: Total suspended sediment (TSS), Al/Si, TOC and of river sediments and gravels from Galy et al. (2007); Al/Si and TOC data used in beta regression model.

Supplementary Table C2: Lothian Bacon Bayesian age-depth model interpolated calibrated ages (input data for Supplementary Figure B9).

Supplementary Table C3: Sajnekhali Bacon Bayesian age-depth model interpolated calibrated ages (input data for Supplementary Figure B10).

Supplementary Table C4: Sajnekhali Bacon Bayesian age-depth model interpolated calibrated ages (input data for Supplementary Figure B11).

Supplementary Table C5: Sajnekhali Bacon Bayesian age-depth model interpolated calibrated ages (input data for Supplementary Figure B12).

Supplementary Table C6: Outputs from binomial logistic regression GLM of Galy et al. (2007) TOC and $\ln(\text{Al/Si})$ data used in Fig. 3 (main text).

Supplementary Table C7: Outputs from beta regression GLM of Galy et al. (2007) TOC and $\ln(\text{Al/Si})$ data used in Fig. 4 (main text).

Supplementary Table C8: Predicted TOC data from the Indian Sundarbans sites (i.e., from Lothian, Gplot, Dhanchi, Dhanchi-2, Bonnie Camp, and Sajnekhali) used in Fig. 4 and Fig. 7 (main text).

Supplementary Table C9: Median grain size (D_{50} μm) and predicted TOC from beta regression from the Indian Sundarbans sites (i.e., from Lothian, Gplot, Dhanchi, Dhanchi-2, Bonnie Camp, and Sajnekhali) used in Fig. 5 and Fig. 6 (main text).

Supplementary Table C10: Grain size data in quarter phi (ϕ) intervals from the Indian Sundarbans sites (i.e., from Lothian, Gplot, Dhanchi, Dhanchi-2, Bonnie Camp, and Sajnekhali).

Supplementary Table C11: X-ray fluorescence (XRF) geochemical composition from the Indian Sundarbans sites (i.e., from Lothian, Gplot, Dhanchi, Dhanchi-2, Bonnie Camp, and Sajnekhali); Al and Si elemental ratios used in this study.

Figure 1

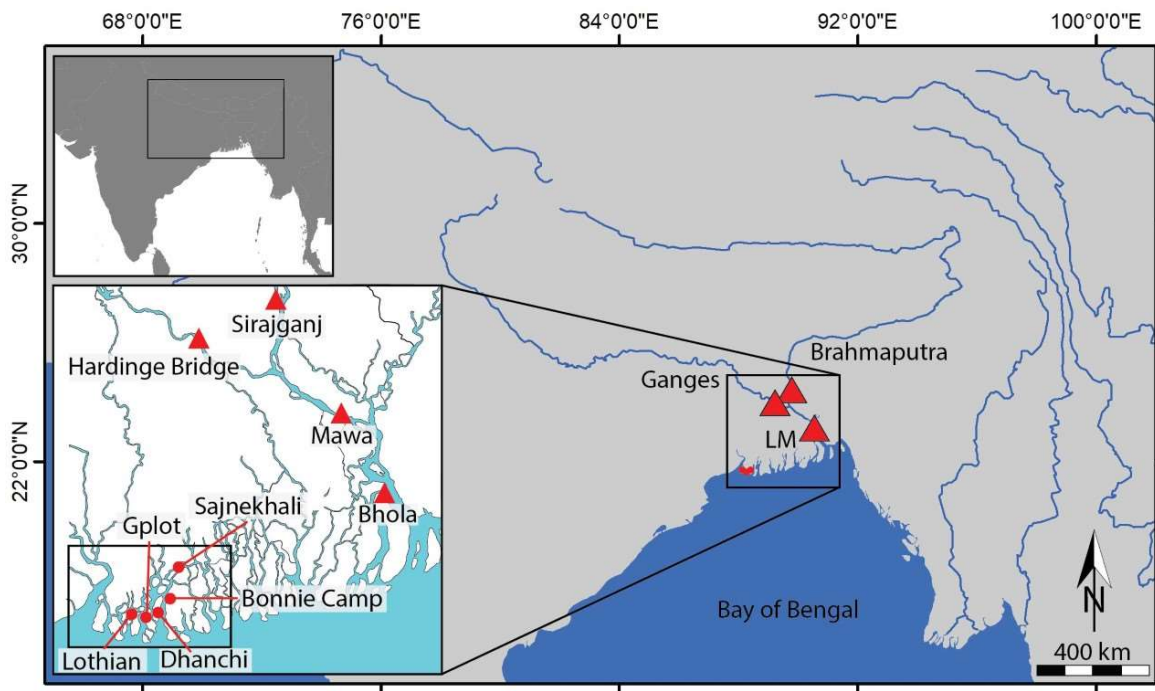


Figure 2

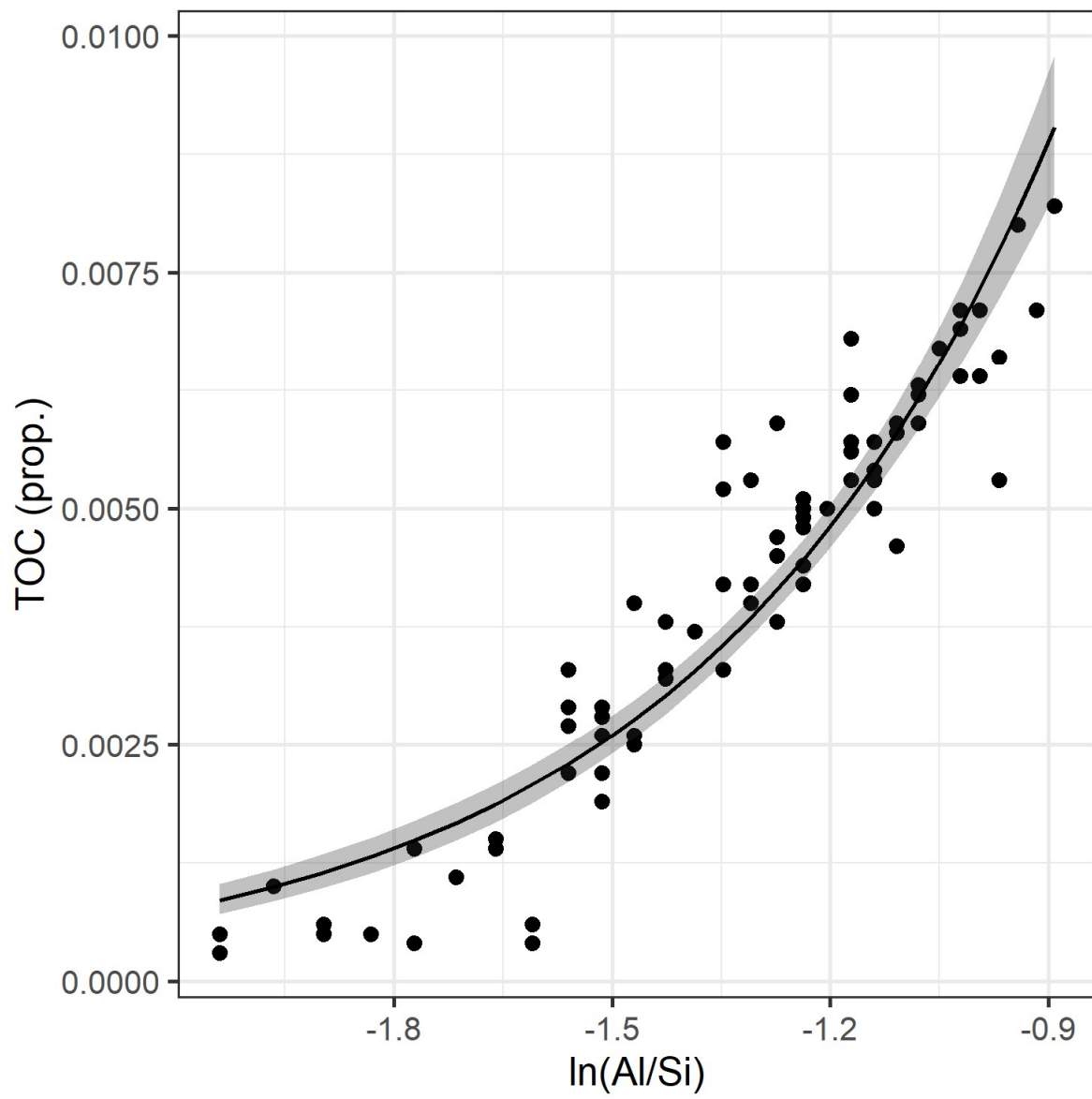


Figure 3

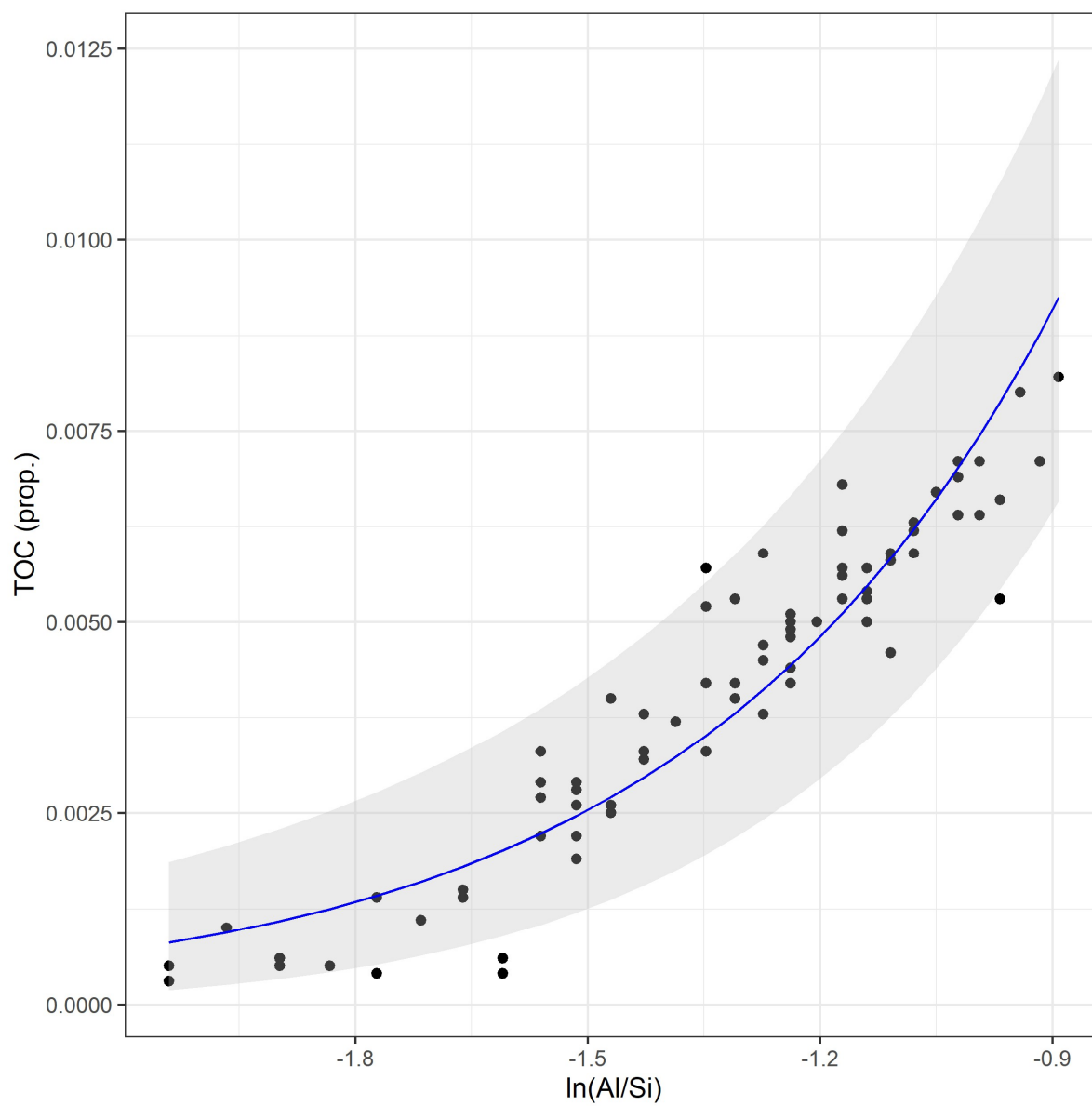


Figure 4

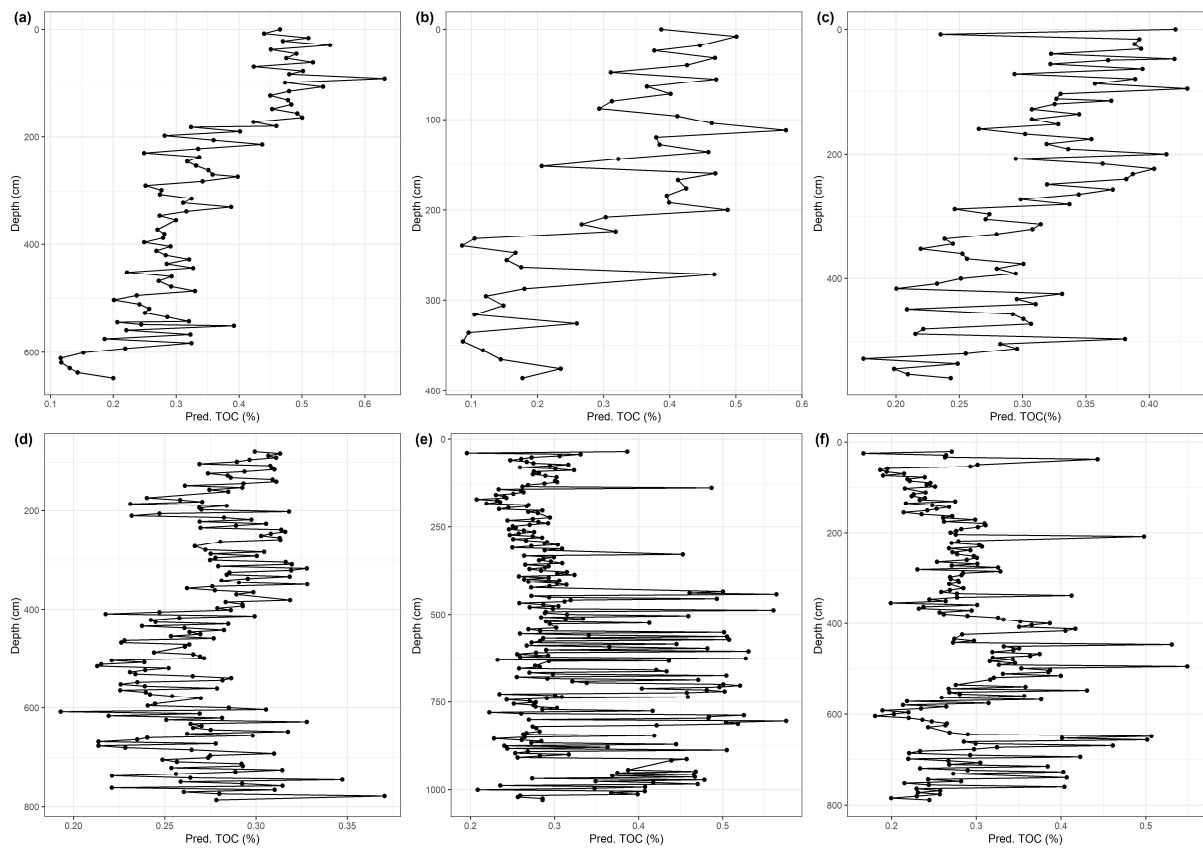


Figure 5

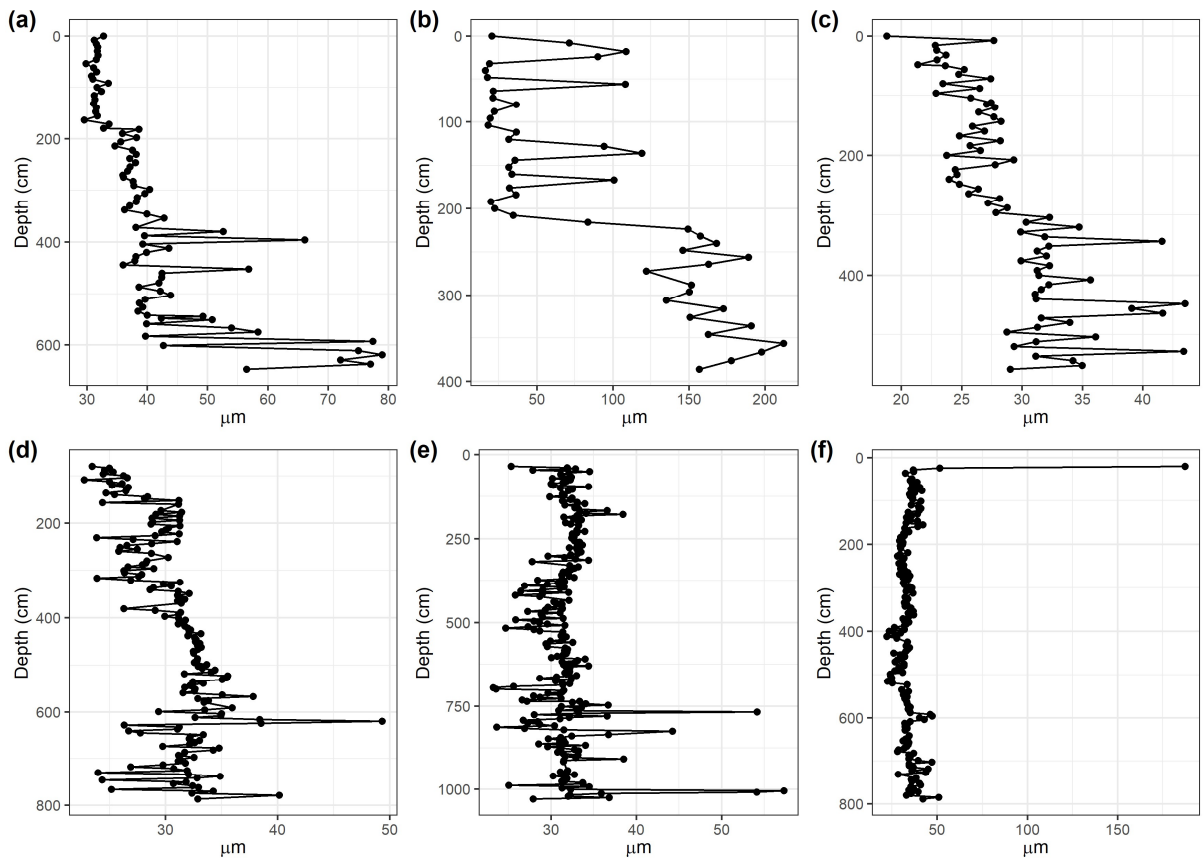


Figure 6

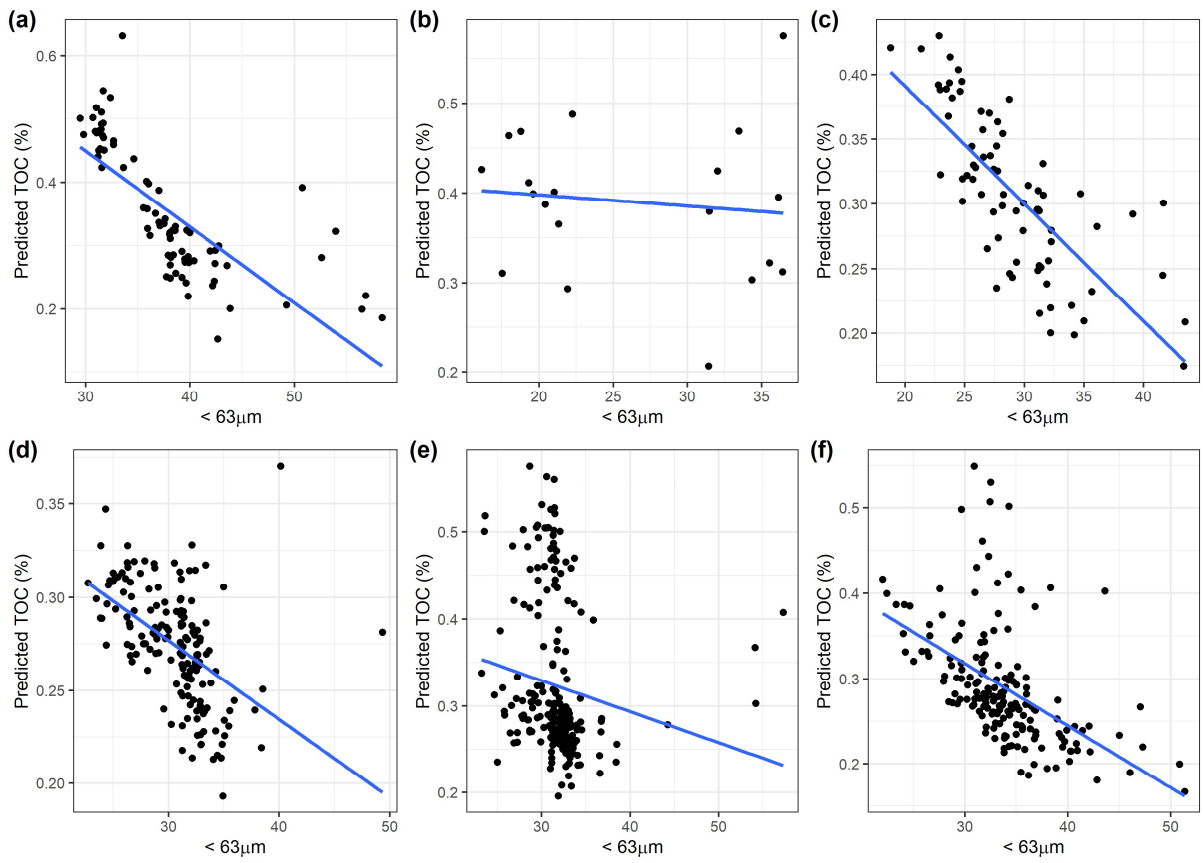


Figure 7

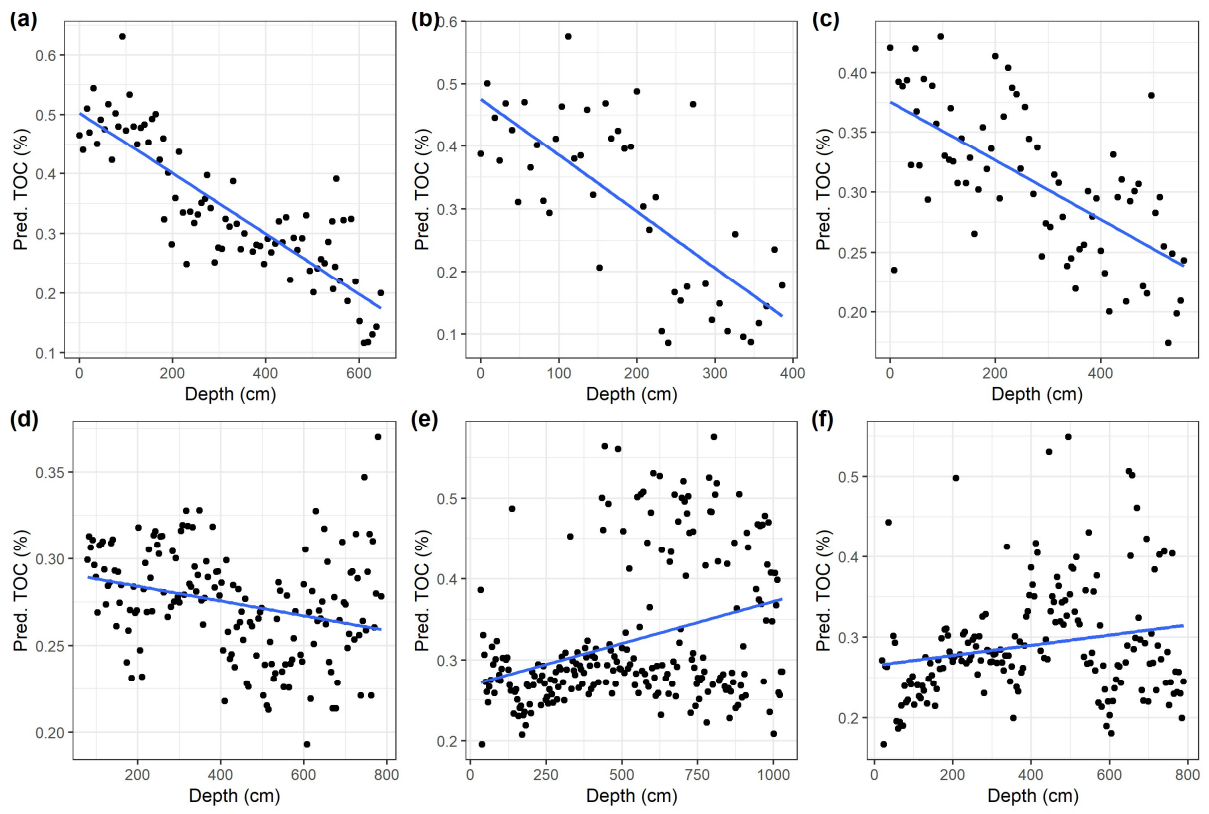


Figure 8

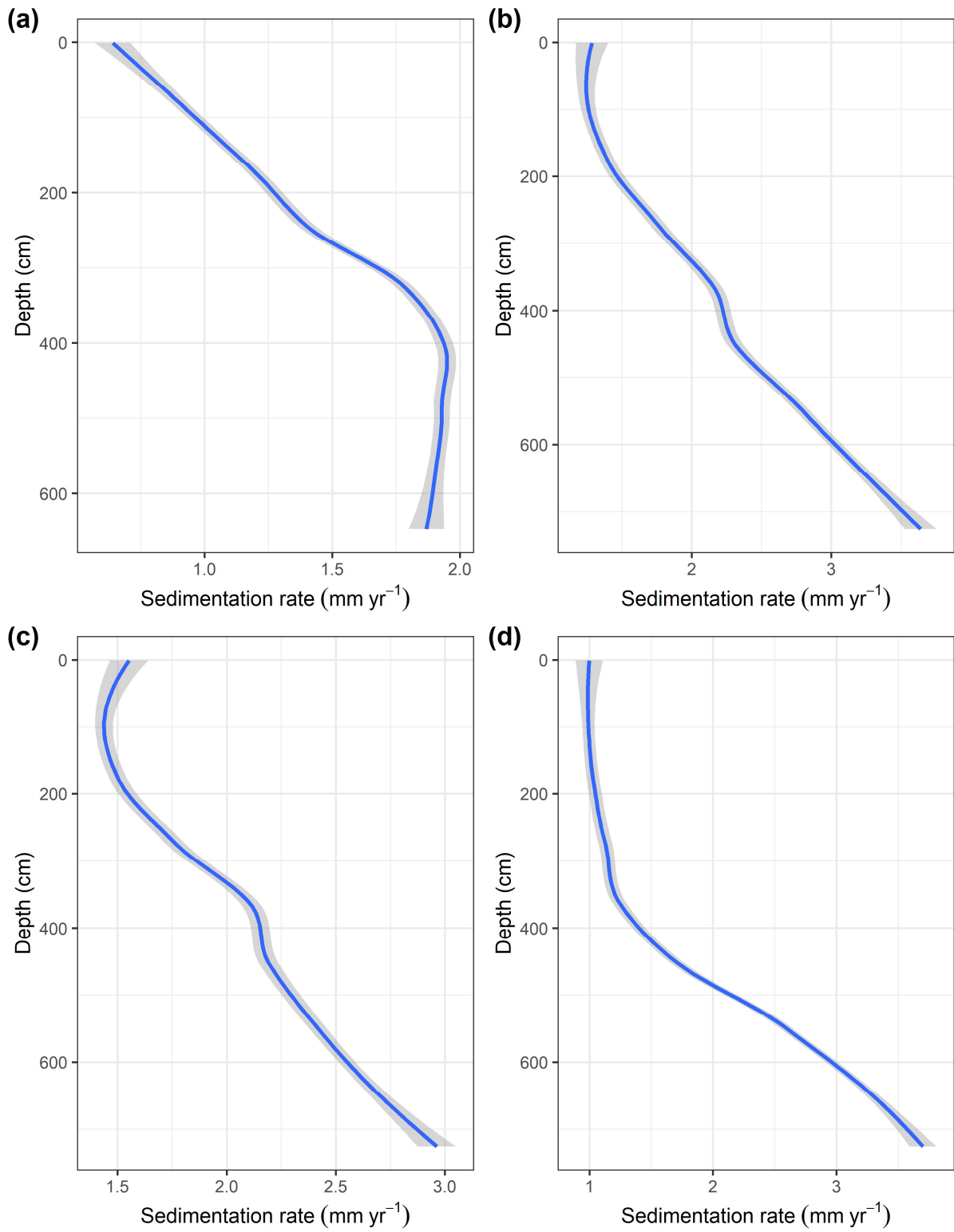


Table 1

Site	ID	Depth (cm)	^{14}C age ($\pm 2\sigma$)	Calendar age with 2σ error (cal yr BP)	Material
Lothian	Surface	0		-60*	NA
	UBA-20910	70	1271 ± 25	1277-1178	Humic acid
	UBA-20571	164	1868 ± 28	1873-1727	Humic acid
	UBA-20569	214	2409 ± 39	2698-2347	Humic acid
	UBA-21170	274	2868 ± 49	3155-2865	Humic acid
	UBA-18154	647	4291 ± 35	4959-4826	Bulk sediment
Gplot	Surface	0		-60*	
	UBA-18992	184	4291 ± 37	4961-4826	Bulk sediment
	UBA-18993	224	3383 ± 37	3719-3510	Bulk sediment
Dhanchi	Surface	0		-60*	
	UBA-21040	78	2635 ± 34	2841-2724	Humic acid
	UBA-21039	158	1620 ± 28	1565-1414	Humic acid
	UBA-20908	160	2777 ± 33	2951-2790	Humic acid
	UBA-21038	214	1437 ± 34	1384-1294	Humic acid
	UBA-18989	280	4245 ± 36	4867-4649	Bulk sediment
	UBA-18991	512	4528 ± 40	5312-5047	Bulk sediment
UBA-18152	558	4081 ± 26	4801-4447	Bulk sediment	
Dhanchi-2	Surface	0		-61**	
	UBA-23517	137.5	3281 ± 35	3586-3409	Humic acid
	UBA-23516	239	2585 ± 34	2769-2519	Humic acid
	UBA-23515	335	2438 ± 31	2700-2357	Humic acid
	UBA-23514	433	3127 ± 37	3444-3240	Humic acid
	UBA-23513	530	3070 ± 48	3383-3161	Humic acid
	UBA-23512	628	2815 ± 40	3055-2798	Humic acid
	UBA-23511	725	2596 ± 38	2782-2518	Humic acid
Sajnekhali	Surface	0		-61**	NA
	UBA-22973	86	ND	ND	Humic acid
	UBA-22972	185	2991 ± 35	3328-3063	Humic acid
	UBA-22971	283	2205 ± 30	2316-2146	Humic acid
	UBA-22970	383	3356 ± 40	3692-3481	Humic acid
	UBA-22969	479	3562 ± 44	3974-3721	Humic acid
	UBA-22968	577	3249 ± 35	3561-3397	Humic acid
	UBA-22967	673.5	3356 ± 37	3690-3482	Humic acid
	UBA-22966	726	3389 ± 33	3707-3563	Humic acid

Date of sample retrieval 2010* and 2011**. ND: No data.