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## 4 Depositional environments and historical contamination as a 5 framework to reconstruct fluvial sedimentary evolution

6

7 Sophia Vauclin<sup>1\*</sup>, Brice Mourier<sup>1</sup>, André-Marie Dendievel<sup>1</sup>, Nicolas Noclin<sup>1</sup>, Hervé Piégay<sup>2</sup>, Philippe  
8 Marchand<sup>3</sup>, Anaïs Vénisseau<sup>3</sup>, Anne de Vismes<sup>4</sup>, Irène Lefèvre<sup>5</sup>, & Thierry Winiarski<sup>1</sup>

9

10 <sup>1</sup>Univ Lyon, Université Claude Bernard Lyon 1, CNRS, ENTPE, UMR 5023, LEHNA, F-69518, Vaulx-en-Velin,  
11 France

12 <sup>2</sup>Univ Lyon, Université Jean Moulin Lyon 3, CNRS, ENS Lyon, UMR 5600 EVS, F-69342, Lyon Cedex 7, France

13 <sup>3</sup>ONIRIS, INRAE, LABERCA Route de Gachet-Site de la Chantrerie-CS 50707, Nantes, F-44307, France

14 <sup>4</sup>LMRE, IRSN, PRP-ENV/STEME/LMRE, F-91400, Orsay, France

15 <sup>5</sup>LSCE, CEA, UMR8212, F-91198, Gif-sur-Yvette, France

16

17 \*Corresponding author: [sophia.vauclin@developpement-durable.gouv.fr](mailto:sophia.vauclin@developpement-durable.gouv.fr)

### 18 Abstract:

19 In this study, we explore the variability of sedimentation conditions (grain-size, accumulation rate,  
20 contamination, etc.) according to fluvial depositional environments. Indeed, sediment cores are  
21 commonly used as archives of natural and anthropogenic activities in hydrosystems, but their  
22 interpretation is often complex, especially in a fluvial context where many factors may affect the  
23 quality, continuity, and resolution of the record. It is therefore critical to thoroughly understand the  
24 nature and dynamics of an environment in which a sediment core is sampled to be able to interpret it.  
25 To that end, four depositional environments from a bypassed reach of the Rhône River were  
26 comparatively investigated through geophysical methods in order to assess the range of sedimentation

27 conditions: a floodplain, a semi-active secondary channel, an active secondary channel, and a dam  
28 reservoir. Sediment cores were retrieved from each environment and thoroughly characterized (grain-  
29 size, Total Organic Carbon, organic contaminants, etc.). Robust age-depth models were elaborated for  
30 each of them based on  $^{137}\text{Cs}$ ,  $^{210}\text{Pb}$ , and Persistent Organic Pollutants (POPs) trends. The results show  
31 that each depositional environment recorded a different time-period, and therefore different  
32 contamination levels and trends. In particular, a shift from polychlorinated biphenyls (PCBs) to  
33 polybrominated diphenyl ethers (PBDEs) as the predominant POP in the sediments can be observed,  
34 the tipping point being set in the 1970s. Two types of infrastructure-induced legacy sediments related  
35 to two periods of river engineering in the reach were also identified using grain-size analysis. The  
36 combination of geophysical methods and sediment cores is therefore confirmed as a relevant  
37 methodology that should be promoted in fluvial contexts in order to reconstruct the sedimentary  
38 evolution of fluvial corridors. The study also highlights the challenges of dating recent fluvial sediments  
39 and proposes a multi-proxy dating methodology using POPs contamination trends.

40 **Keywords:** Age-depth model; depositional environment; natural archive; polybrominated  
41 diphenyl ethers; polychlorinated biphenyls; sediment core.

## 42 1. Introduction

43 Monitoring the signals -anthropogenic or natural – that sediments record may provide invaluable  
44 information to understand the long and short-term evolution of a hydrosystem. Indeed, numerous  
45 studies have shown that sediments may be used as archives of meteorological events (e.g. storms:  
46 Vliet-Lanoe et al., 2014; Pouzet and Maanan, 2020, floods: Jones et al., 2012; Toonen et al., 2015),  
47 historical contamination (e.g. Bigus et al., 2014, Dendievel et al., 2020) or other anthropogenic impacts  
48 such as pre-historic gullyng (Dotterweich, 2005; Vanwalleghem et al., 2006), agriculture intensification  
49 (Brooks and Brierley, 1997; Fuller et al., 2015) or river manipulations (Surian and Rinaldi, 2003; Vauclin  
50 et al., 2019). The diversity of anthropogenic disturbances potentially recorded in sediments even led

51 to the establishment of the term “legacy sediments” to refer to sediments resulting from past or  
52 contemporary human activities such as land-use, contamination (Walter and Merritts, 2008; James,  
53 2013; Wohl, 2015) or river engineering (Vauclin et al., 2020). However, in order to use wisely the  
54 information contained in sediment deposits, it is critical to understand their recording ability  
55 depending on their geographical and geomorphological context. Factors such as connectivity of the  
56 environment and sediment accumulation rate, for example, will affect the continuity and precision of  
57 the record, while sediment characteristics such as grain-size or organic carbon content will specifically  
58 affect contamination chronicles (Karickhoff et al., 1979). Some environments such as lakes allow  
59 continuous and regular sedimentation. Sediments deposited in such contexts are therefore reliable  
60 and lasting archives that tend to be preferentially used to study local or atmospheric contamination  
61 trends, anthropogenic activities in the upstream catchment (Guzzella et al., 2008; Guédron et al., 2016)  
62 or long-term climatic or environmental changes (Simonneau et al., 2013; Zolitschka et al., 2013). In  
63 contrast, there is usually a large diversity of depositional environments in fluvial contexts (e.g.  
64 floodplains, oxbow lakes, secondary channels, paleochannels, weir ponds, groyne fields, locks, etc.).  
65 Those environments may have various levels of connectivity and therefore various recording capacities  
66 (Dhivert et al., 2015) and temporalities, making the interpretation of sedimentary archives challenging.  
67 When implemented, river engineering may also profoundly reshape the natural depositional  
68 environments and alter their sedimentation processes. Additionally, sediment accumulation rates in a  
69 fluvial context are often dependent on hydrological events: sustained low-water conditions might  
70 result in a decreased or non-existent sedimentation, while a flood might bring a dozen centimetres of  
71 sediments in the span of a few hours or remobilize previously settled sediments, generating  
72 discontinuous archives. The dating of fluvial sediments is therefore key for a relevant interpretation of  
73 a sedimentary sequence, but might also prove challenging, as classic methods such as  $^{210}\text{Pb}$  or  $^{137}\text{Cs}$   
74 might not give satisfying results due to the continuity issues previously mentioned.

75 In that light, investigating the various fluvial compartments appears as essential for any retrospective  
76 approach implemented from fluvial sediments. It is the only way to potentially reconstruct the entire

77 timeline of a river, where individual sediment sequences are inherently incomplete due to the  
78 changing nature of fluvial systems. It might also allow to establish links between sediments and their  
79 deposition processes, and identify sediment sequences resulting from changes in fluvial systems. It is  
80 especially important as rivers are characterized by complex sedimentary dynamics that are often  
81 further segmented by anthropogenic modification of their morphology and functioning. However,  
82 when suitably interpreted, sedimentary sequences may allow to reconstruct the evolution of a river  
83 corridor in correlation with the anthropogenic pressures it was subject to throughout time. Such an  
84 understanding is essential to implement relevant restoration measures and mitigate further  
85 anthropogenic impact on the river system (Wohl et al., 2005; Arnaud et al., 2015; Slowik, 2015;  
86 Eschbach et al., 2018).

87 The aims of this study are:

- 88 - to highlight the temporal and qualitative variability of fluvial sediment records –especially  
89 regarding contaminant chronicles– depending on their depositional environment,
- 90 - to use these findings to reconstruct the sedimentary evolution of a large river,
- 91 - overall, to propose a robust methodology for the use of fluvial sediment deposits as records  
92 of natural or anthropogenic disturbances in an actively changing fluvial system.

93 To this end, four distinct depositional environments from a 12km-long heavily anthropized reach of  
94 the Rhône River (France) are investigated in a comparative approach. Their dynamics and the  
95 chronology of their records are assessed and compared through a combination of geophysical surveys

96 and dated sediment cores in order to describe the diversity of sedimentary records and reconstitute  
97 the trajectory of this actively adjusting fluvial area.

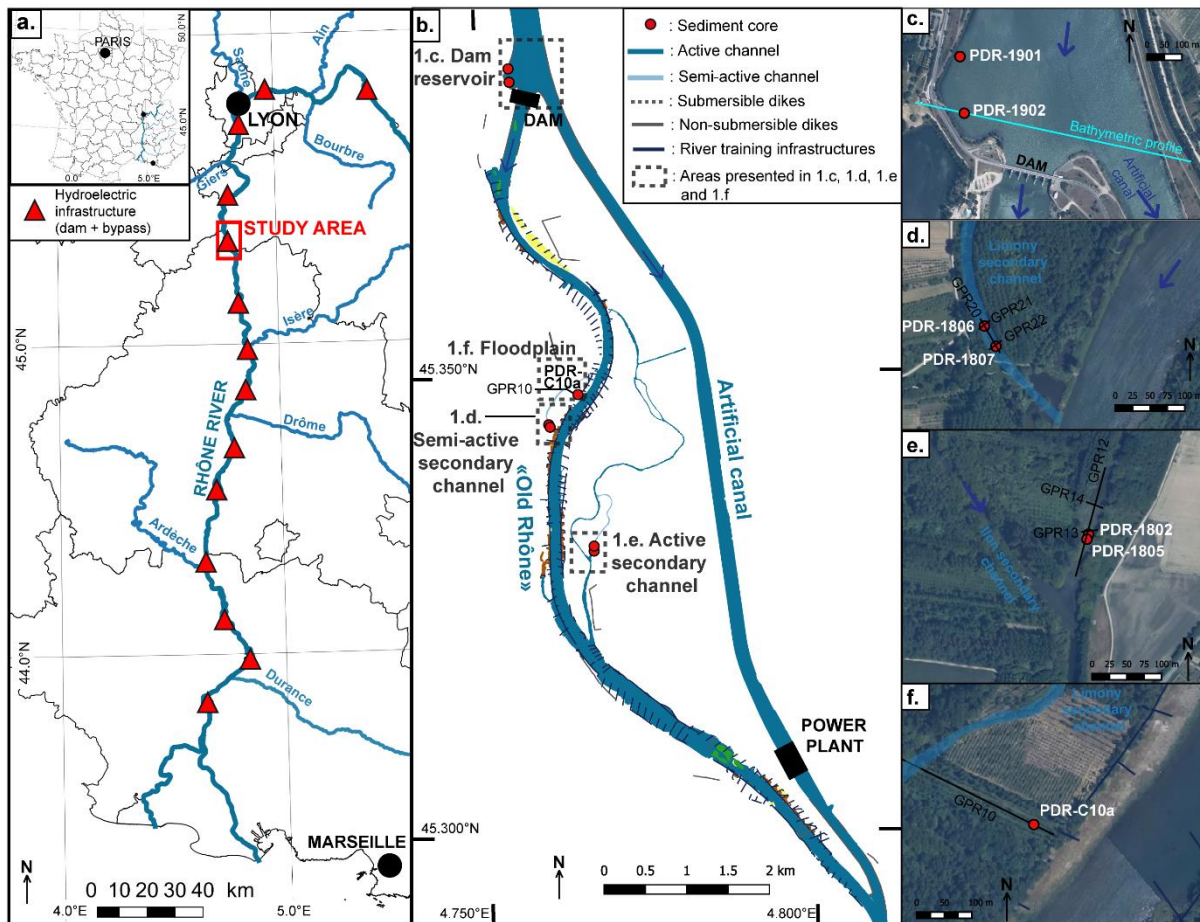
## 98 2. Material and methods

### 99 2.1. Study area

100 The Rhône River is an 812 km-long river that flows from the Furka Glacier in Switzerland to the  
101 Mediterranean Sea in France (Fig. 1.a.). Its catchment covers 98 500 km<sup>2</sup> -of which 90 500 km<sup>2</sup> are in  
102 France- and its average inter-annual discharge is approximately 1 700 m<sup>3</sup>s<sup>-1</sup> at the estuary. It has a long  
103 history of human influence and the earliest traces of agriculture along the river date back to around 7  
104 500 years ago (Berger et al. 2019). As most major rivers in Europe, the Rhône is highly engineered, with  
105 numerous dikes, groynes and 19 dams along its French course. Those infrastructures have impacted  
106 the river in many ways: increased transport capacity, reduced sediment load, channel armouring,  
107 narrowing and incision, increased slope, increased trapping efficiency, etc. (Petit et al., 1996; Bravard  
108 et al., 1999; Arnaud-Fassetta, 2003; Parrot, 2015; Tena et al., 2020).

109 The study area (known as PDR for « Péage-de-Roussillon ») is located 50 km downstream from the  
110 metropolitan city of Lyon (Fig. 1.a). It consists of a 12 km-long reach with a bypass configuration typical  
111 of the Rhône River: the historical main channel is closed by an upstream run-of-river dam and bypassed  
112 by an artificial canal (Fig. 1.b). The area has undergone two phases of engineering in the last 200 years.  
113 River training and flood protection infrastructures were implemented from the mid-19th century to  
114 the early 20th century: non-submersible dikes were built on the floodplain, and submersible dikes and  
115 groynes were implemented in the river in order to cut secondary arms and straighten the main  
116 channel. From 1974 to 1978, the area was heavily modified to produce hydroelectricity: an 11 km-long  
117 canal was dug out in parallel to the existing one and equipped with a lock and a power plant, and the  
118 historical channel was closed with a run-of-river dam to divert most of the discharge in the canal. As a  
119 result, only a minimum instream flow of 50 to 125 m<sup>3</sup>/s is maintained nowadays in the bypassed  
120 channel (GRAIE, 2016), while the inter-annual discharge upstream of the area is 1080 m<sup>3</sup> s<sup>-1</sup>. The

121 historical channel is still used to accommodate flood flows that exceed the maximum discharge  
 122 processed by the hydropower plant ( $1600 \text{ m}^3 \text{ s}^{-1}$ ).



123  
 124 *Figure 1: a. Location of the study area along the Rhône River; b. Overview of the study area and its river infrastructures (NB:*  
 125 *core PDR-C10a has been previously published in Vauclin et al., 2019); c. Study site: Saint-Pierre-de-Boeuf dam reservoir; d.*  
 126 *Study site: Limony secondary channel; e. Study site: Ilon secondary channel. [IN COLOR]*

127 Inside this area, four fluvial compartments were chosen in an attempt to characterize and compare  
 128 four distinct types of depositional environments. The selected study sites exhibit different hydro-  
 129 sedimentary dynamics which have been created or impacted by various periods and types of river  
 130 engineering, as shown in a planimetric analysis of the area published in Tena et al. (2020).

131 - The Saint-Pierre-de-Boeuf dam reservoir is a recent compartment as it was built in 1978  
 132 upstream of the bypassed section (Fig. 1.b). It is a run-of-river installation, therefore most

133 sediments flow through the dam, especially during flooding. However, a recess zone along the  
134 right bank allows sediment accumulation (Fig. 1.c).

135 - The Ilon secondary channel (Fig. 1.e) was part of a major arm of the Rhône River until the  
136 implementation of river training structures in the 1880s, when it became less connected as  
137 both its inflow and outflow were barred by submersible dikes and groynes (Vauclin et al., 2019;  
138 Tena et al., 2020). In the 1950s, the Ilon secondary channel had lost in width but both its ends  
139 were still linked to a secondary arm of the main stream. Following the bypass implementation  
140 in 1978, the upstream entrance got disconnected and the channel is now supplied by its  
141 downstream end only but still underwater all year-long. It is further referred to as an active  
142 secondary channel.

143 - The Limony secondary channel (Fig. 1.d) used to be an active arm of the river prior to the first  
144 engineering phase. Both its entrance and outflow were then closed by submersible dykes in  
145 the 1880s and it quickly became disconnected from the main channel (Vauclin et al., 2019;  
146 Tena et al., 2020). Nowadays, it is mostly supplied by groundwater and flooded by the river  
147 several times a year. It is further referred to as a semi-active secondary channel.

148 - An overbank floodplain location that used to be flooded regularly prior to the bypass  
149 implementation but is entirely disconnected from the river since the 1980s (Vauclin et al.,  
150 2019). Note that part of the results from this site have already been published (Vauclin et al.,  
151 2019; Vauclin et al., 2020) but are used in this study for a broader comparison of different  
152 depositional environments.

## 153 2.2. Bathymetric and geophysical analysis

154 Bathymetric surveys measured along a transect from the Saint-Pierre-de-Boeuf dam reservoir on which  
155 a sediment core is located (Fig. 1.c) were provided by the CNR (Compagnie Nationale du Rhône;  
156 company in charge of the river management). They were available for six dates: 1980, 1999, 2004,  
157 2008, 2012 and 2018, and allowed to reconstruct the progressive filling of the reservoir with  
158 sediments.

159 Ground Penetrating Radar (GPR) is a non-invasive geophysical method used to obtain a high-resolution  
160 image of shallow subsurface structures (Davis and Annan, 1989). It is based on the generation and  
161 propagation of discrete pulses of high frequency (MHz) electromagnetic energy through the  
162 sediments. It allows the assessment of subsurface spatial structures up to a depth of 10–20 m over  
163 extended areas (Huggenberger et al., 1994; Beres et al., 1999) with a resolution ranging between 10  
164 cm and 1 m (Jol, 1995). It can be used on the ground (GPR) or on shallow water bodies (1 to 2 meters  
165 of water depth) where it is then referred to as Water Ground Penetrating Radar (WGPR: Annan et  
166 Davis, 1977).

167 A 500 m-long GPR survey (GPR10) was acquired on the floodplain site in 2016. Three GPR profiles were  
168 acquired on the active secondary channel (GPR12, GPR13, and GPR14; 160 m, 22 m, and 25 m  
169 respectively; only) in October 2018 (Fig. 1.e). The GPR was used in WGPR configuration from a small  
170 boat as there was a water depth of 60 to 70 cm in the channel at the time. Three GPR profiles were  
171 acquired on the semi-active secondary channel (GPR20, GPR21, and GPR22; 55 m, 18 m, and 20 m  
172 respectively) in October 2018 as well (Fig. 1.d). The measurements were made directly on the ground  
173 as there was no water in the channel at the time. On the floodplain, one 500 m-long survey was  
174 acquired in 2016 (GPR10). All GPR surveys were carried out with the GSSI SIR 3000 system (Geophysical  
175 Survey System Inc., Salem, USA), operated with a shielded antenna at a central frequency of 400 MHz,  
176 running in monostatic mode. In addition, a topographic survey using a GPS Trimble station was  
177 performed to localize the surveys. Velocities of  $0.11 \text{ m ns}^{-1}$  (measured by a common mid-point  
178 procedure) and  $0.076 \text{ m ns}^{-1}$  (from Lin et al., 2009) were used for the terrestrial and aquatic surveys  
179 respectively, in order to calibrate the profile depths. The data processing was performed using the GSSI  
180 Radan 7 software.

## 181 2.3. Sediment characteristics

### 182 2.3.1. Core sampling

183 The core sampling locations were chosen after a quick on-site analysis of the geophysical surveys so  
184 that the cores would cross interesting sedimentary structures. Two underwater cores (PDR-1901 and  
185 PDR-1902) of 72 and 104 cm respectively were retrieved from the dam reservoir in July 2019 with an  
186 UWITEC® corer (Uwitec, Mondsee, Austria) fitted with a 2 m-long and 90 mm-diameter plastic liner.  
187 Two underwater cores (PDR-1802 and PDR-1805) of 94 and 57 cm respectively were sampled from a  
188 small boat in the active secondary channel (Fig. 1.e) in October 2018 with the same corer. Three  
189 terrestrial cores (PDR-1806a, PDR-1806b and PDR-1807) of 82, 74.5 and 78.6 cm respectively were  
190 retrieved from the Limony secondary channel in dewatered state with a Cobra TT percussion driller in  
191 October 2018. Cores PDR-1806b was drilled 1 m apart from PDR-1806a in a 60 cm-deep pit, in order  
192 to combine the two cores; the resulting mastercore (PDR-1806) is 108 cm-long. A 111 cm-long core  
193 (PDR-C10a) was sampled from the floodplain site in 2016 with the same Cobra TT percussion driller.  
194 The detailed characteristics of the studied sediment cores are summarized in Table S1.

### 195 2.3.2. Grain-size characteristics

196 Grain-size was measured on all cores every 0.5 to 1 cm with a Mastersizer 2000© (Malvern Panalytical)  
197 particle size analyzer mounted with a hydro SM small volume dispersion unit. Descriptive grain-size  
198 statistics (D50, mode, D10, D90, skewness, etc.) were computed with the Gradistat software (Blott and  
199 Pye, 2001). To represent the grain-size distributions (GSD) as extensively as possible, heatmaps of each  
200 core were plotted; for each measured sample, the percentage of each grain-size class is represented  
201 by a colour scale. Unmixing of the sets of grain-size distributions was carried out on all sediment cores  
202 simultaneously through end-member modelling analysis (EMMA) with the R package EMMAgeo  
203 (Dietze et al., 2012). EMMA is a compositional method based on the assumption that any GSD is a  
204 mixture of sediment populations corresponding to different mechanisms of production and/or  
205 transport (Weltje and Prins, 2003; Toonen et al., 2015; Dietze et al., 2016). A linear mixing model is  
206 therefore applied to an array of GSDs and allows to identify meaningful end-members that correspond

207 to the most representative populations of the GSDs (Weltje and Prins, 2007). The end-member scores  
208 (percentage of representation of the different end-members) were plotted as a function of the depth  
209 for each core.

### 210 *2.3.3. Geochemical and contaminants analyses*

211 Rock-aval pyrolysis was used to analyze several organic carbon-associated parameters of the  
212 sediments, such as the total organic carbon (TOC). The analyses were carried out at the ISTO laboratory  
213 (Orléans, France) with a 4 cm step on cores PDR-C10a, PDR-1802, PDR-1806, and PDR-1902.

214 Polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) were analyzed. PCBs  
215 were chosen as they are well-studied POPs whose temporal trends are reasonably well-known in the  
216 Rhône River (e.g. Desmet et al., 2012, Mourier et al., 2014, Dendievel et al., 2020). Meanwhile, PBDEs  
217 are emerging POPs that have been little studied in the Rhône, which brings a novelty factor. Besides,  
218 works from other hydrosystems (Nylund et al., 1992, Zegers et al., 2003, Muir and Rose, 2007) suggest  
219 that they have consistent and more contemporary temporal trends, meaning that they could  
220 potentially be used as a time-marker for recent sediment records.

221 The following organic contaminants were quantified in the LABERCA laboratory (ONIRIS Nantes,  
222 France):

- 223 - The seven indicator PCB congeners: PCB 28, 52, 101, 118, 138, 153, 180, of which the sum is  
224 further referred to as  $\sum 7\text{PCBi}$  on cores PDR-1902, PDR-1802, PDR-1806 and PDR-C10a;
- 225 - Nine PBDE congeners: PBDE 28, 47, 49, 99, 100, 153, 154, 183, and 209, of which the sum is  
226 further referred to as  $\sum 9\text{PBDE}$  on cores PDR-1902, PDR-1802 and PDR-1806;

227 The samples were collected at a step of 4 cm, packaged in amber glass vials and then sent to the  
228 LABERCA laboratory for further analysis. After freeze-drying, extraction and purification procedures,  
229 all persistent organic pollutants (POPs) analyses were simultaneously carried out by gas  
230 chromatography coupled with high-resolution mass spectrometry (GC/HRMS) using a 7890A gas  
231 chromatograph (Agilent) coupled with a JMS 800D double-sector high-resolution mass spectrometer

232 (JEOL, Tokyo, Japan). The quantification was ensured by isotopic dilution using 7 <sup>13</sup>C-labeled internal  
233 PCB standards and 9 <sup>13</sup>C-labeled internal PBDE standards added to the samples before the extraction  
234 step. A thorough description of the analysis protocol can be found in Vénisseau et al. (2015) and Liber  
235 et al. (2019). The limits of detection (LOD) for PCBs range from 0.049 (PCB 118) to 0.109 (PCB 28)  
236  $\mu\text{g}\cdot\text{kg}^{-1}$  DW and for PBDEs from 0.002 (PBDE 28) to 0.05 (PBDE 183)  $\mu\text{g}\cdot\text{kg}^{-1}$  DW. The recoveries range  
237 from 60 to 120%.

#### 238 *2.3.4. Sediment dating*

239 Cesium-137 (<sup>137</sup>Cs) was assessed on cores PDR-1802, PDR-1806, PDR-1902 and PDR-C10a. This  
240 anthropogenic radionuclide is typically used to identify two events that appear as peaks in the  
241 radionuclide activity: nuclear weapon testing in 1962-1963 (Ritchie and McHenry, 1990) and the  
242 Chernobyl accident in 1986. Additionally, excess lead-210 (<sup>210</sup>Pb<sub>xs</sub>) activity was assessed on core PDR-  
243 1902 only. <sup>210</sup>Pb is a natural radionuclide that is part of the uranium-238 radioactive decay series and  
244 is continually supplied to sediments from atmospheric deposition. Excess <sup>210</sup>Pb radioactive decay can  
245 be used to model sediment accumulation rate in a sediment core –and by extension, date it- assuming  
246 that the sediments have not been subjected to large scale disturbances (Baskaran et al., 2014). We  
247 assumed that there was neither mixing nor diffusion of <sup>210</sup>Pb<sub>xs</sub> in the core and therefore used the  
248 Constant Flux Constant Sedimentation model in this study (Krishnaswamy et al., 1971; Bruel and  
249 Sabatier, 2020).

250 Samples were collected at steps of 4 to 8 cm, dried for 48h at 40°C and preserved in polystyrene boxes.  
251 Sub-samples from each core were analyzed by counting for at least 24h, using low-background gamma  
252 spectrometry. Gamma emissions were detected with a germanium detector and used to quantify  
253 specific activities of <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>40</sup>K, <sup>210</sup>Pb, and <sup>137</sup>Cs. Those analyses were carried out in the LMRE  
254 laboratory (Laboratoire de Métrologie de la Radioactivité dans l'Environnement, IRSN, Orsay) for cores  
255 PDR-C10a, PDR-1802 and PDR-1806, and in the LSCE laboratory (Laboratoire des Sciences du Climat et  
256 de l'Environnement, Gif-sur-Yvette, France) for core PDR-1902. The mean detection limit is 0.1 Bq.

257 Age-depth modelling was carried out on cores PDR-1802, PDR-1806, PDR-1902 and PDR-C10a using  
258 the R package “clam” (Classical Age-Depth Modelling of Cores from Deposits) v.2.3.4 (Blaauw, 2010).

259 Two models were developed:

260 - Model M1 uses only the most robust time-markers i.e. the surface (date of sampling), <sup>137</sup>Cs or  
261 <sup>210</sup>Pb trends and abrupt grain-size events associated with dated hydrological or human-driven  
262 events.

263 - Model M2 uses the time-markers from model M1 as well as additional time-markers linked to  
264 PCB and PBDE trends (e.g. date of early use of a contaminant, date of restriction of use, date  
265 of ban, etc.).

266 The point in developing those two models was to assess whether contaminant trends can be used as  
267 reliable time-markers in this context and whether they might allow to increase the dating accuracy.

268 The model type (e.g. linear regression, linear interpolation, etc.) was chosen for each core as to obtain  
269 the best possible goodness of fit. The model type and parameter inputs for each model are available  
270 in Supplementary Information (Table S2).

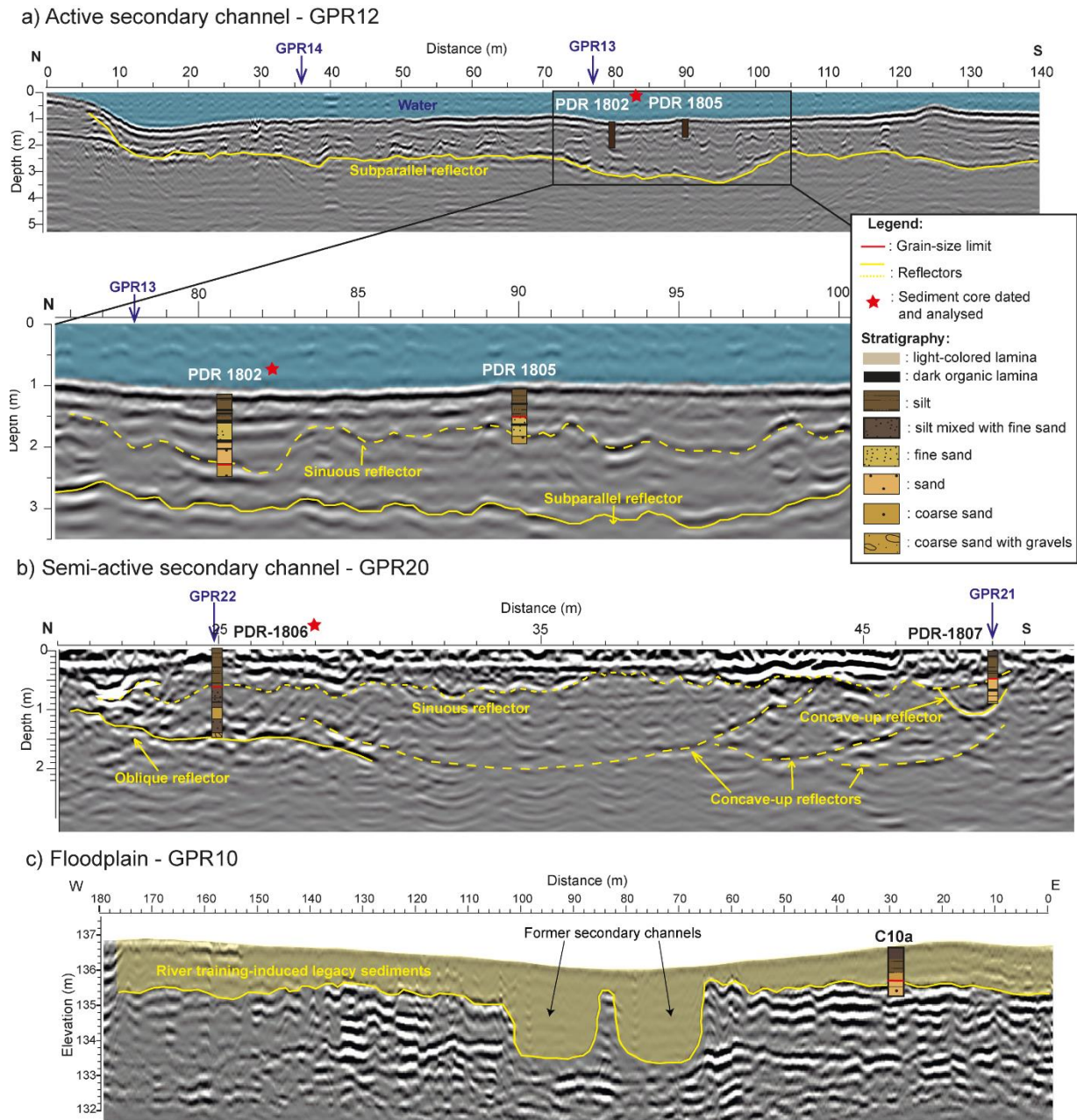
## 271 3. Results

### 272 3.1. Physical characteristics of the depositional environments

273 The bathymetric measurements of a transect of the Saint-Pierre-de-Boeuf dam reservoir over time are  
274 featured in Supplementary Information (Fig. S1). They show the progressive filling of the recess zone  
275 on the right bank side (between roughly 100 and 300 m). In 1980 (two years after the dam  
276 implementation in 1978), the old channel could still be distinguished around 150-200 m. The major  
277 part of the filling (up to 6.5 m of sediments deposited) happened between 1980 and 1999. Since 1999,  
278 up to one additional metre accumulated in the recess zone. Core PDR-1902 was sampled in this zone,  
279 around 150 m on the measured transect. Its compaction rate could not be measured and therefore  
280 had to be assumed; based on previous underwater cores, a 30% rate was used for the representation

281 in Fig. S1. The high uncertainty associated with the compaction rate of core PDR-1902 means that the  
282 bathymetric measurements cannot be used as absolute time-markers. However, they still suggest that  
283 the bottom of the core is older than 1999.

284 The Ilon (active) secondary channel is approximately 20 m large, 300 m long and with a water depth  
285 depending on the season but generally around 1 m. The longitudinal survey GPR12 (Fig. 2.a) shows  
286 that more than 5 m of sediments accumulated in the channel overtime. A continuous subparallel  
287 reflector can be observed along the whole profile around 2-2.5 m. It might correspond to the original  
288 gravel bed of the channel (before 1860, cf. section 2.1). The filling above this reflector is rather  
289 homogeneous with mostly subparallel and sinuous reflectors, indicating a continuous deposition with  
290 little to no reworking. A close-up on the part of the transect where the sediment cores are located  
291 allows to distinguish a continuous sinuous reflector that matches with a grain-size limit observed in  
292 sediment cores PDR-1802 and PDR-1805 (see section 3.2 for more details), proving the consistency  
293 between GPR observations and the results from the core sampling. The survey shows that although  
294 one core is 94 cm-long while the other is 57 cm-long, they deposited roughly in the same time span,  
295 meaning sediment accumulation rates may vary a lot inside this compartment. This variability is  
296 inherent to fluvial sediment deposition and has to be considered for a correct interpretation of  
297 sediment records, which demonstrates the interest of combining sediment cores with geophysical  
298 surveys.



299

300 *Figure 2: Interpreted Ground Penetrating Radar surveys with sediment cores positioned; a) GPR12 from the active secondary*  
 301 *channel; b) GPR20 from the semi-active secondary channel; c) GPR10 from the floodplain. NB: Other transversal GPR surveys*  
 302 *for the active secondary channel (GPR21, GPR22) and the semi-active secondary channel (GPR13, GPR14) are available in*  
 303 *Supplementary Information (Fig. S2). [IN COLOR]*

304 The Limony secondary channel is approximately 20 m-large and 1 km-long, although the geophysical  
 305 surveys and sediment cores are only located in the last 200 m downstream of the channel (Fig. 1.d).  
 306 The longitudinal transect GPR20 (Fig. 2.b) shows multiple oblique and concave-up reflectors between  
 307 depths of 0.5 and 2 m. They may be the result of strong and multiple erosion dynamics, indicating that

308 the channel used to have an important bed mobility. The bottoms of both sediment cores (PDR-1806  
309 and PDR-1807) match with reflectors, which shows again the consistency between the two methods.  
310 Around 50 cm, a rather continuous and sinuous reflector can be identified all along the survey. The  
311 sediment filling above this reflector is more homogeneous, hinting at calmer depositional processes.  
312 GPR12 from the floodplain compartment is roughly perpendicular to the Rhône main channel axis. A  
313 very clear continuous reflector can be observed all along the survey (Fig. 2.c), outlining two former  
314 secondary channels that have been entirely filled in and are not discernible nowadays from the surface.  
315 Other subparallel reflectors are apparent below the continuous reflector while the GPR image is  
316 smooth above: this filling is characteristic of legacy sediments induced by the implementation of river  
317 training infrastructures in the 1880s in the area, as demonstrated in Vauclin et al. (2020). The  
318 continuous reflector also coincides with a grain-size limit observed in the corresponding sediment core  
319 (PDR-C10a).

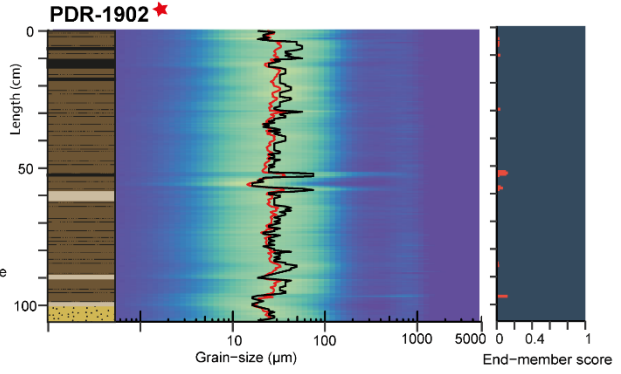
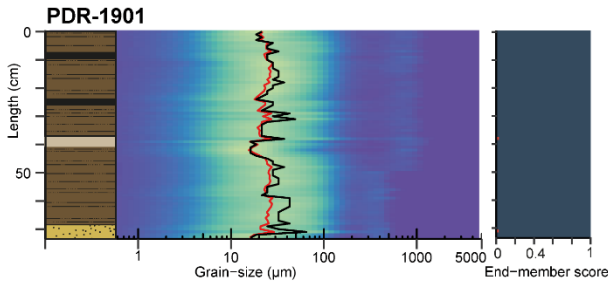
### 320 3.2. Grain-size characteristics of the sediment cores

321 The cores from the dam reservoir (PDR-1901 and PDR-1902) are almost exclusively comprised of silt,  
322 with only a few centimetres of very fine sand at their bottom. At their opening, they exhibited organic  
323 macro-remains and dark organic laminas, as well as a few light-coloured laminas. The cores retrieved  
324 in secondary channels (PDR-1802, PDR-1805, PDR-1806, PDR-1807) have a similar stratigraphic  
325 description: an upward fining pattern of coarse sand, fine sand and silt from the bottom to the top of  
326 the cores (Fig. 3). They also had many noticeable organic macro-remains (leaves, seeds, shells, etc.)  
327 and several darker laminas. The core from the floodplain (PDR-C10a) is characterized by silty deposits  
328 in its top 75 cm and sands at the bottom.

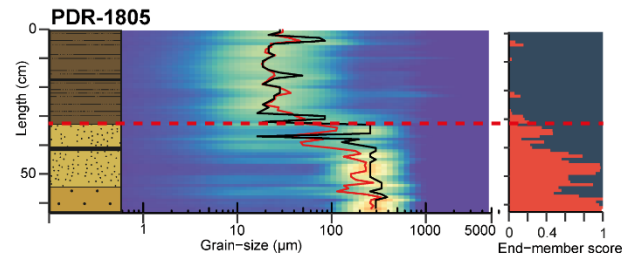
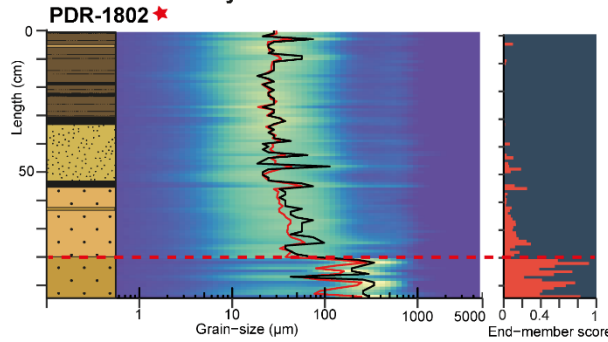
329 The grain-size heatmaps (Fig. 3) from the cores from the dam reservoir highlight a very homogeneous  
330 grain-size distribution, only comprised of fine ( $D_{50} \sim 20 \mu\text{m}$ ) and poorly sorted sediments. In contrast,  
331 two main sections can be distinguished in the cores from the active channel: the top sediments are  
332 fine ( $D_{50} \sim 20\text{-}50 \mu\text{m}$ ) and poorly classified (i.e. the distribution is widely spread around the  $D_{50}$ ), while

333 the sediments at the bottom of the cores are coarser ( $D_{50} \sim 200 \mu\text{m}$ ) and well-sorted (i.e. the  
334 distribution is closely centred around the  $D_{50}$ ). This change in grain-size characteristics can be  
335 observed at 80 cm and 42 cm on PDR-1802 and PDR-1805 respectively (Fig. 3). A similar grain-size limit  
336 can be identified on the heatmaps of the cores from the semi-active secondary channel, at 48 and 40  
337 cm in cores PDR-1806 and PDR-1807 respectively. The transition between the two grain-size patterns  
338 is sharp and is represented as a dotted red line on Fig. 3. A similar distinction between the top (fine,  
339 poorly sorted sediments) and the bottom (coarser, well-sorted sediments) can be observed in core  
340 PDR-C10a from the floodplain environment. The corresponding grain-size limit has already been  
341 analysed in Vauclin et al. (2019) and has been identified as the result of the implementation of river  
342 training infrastructures in the 1880s.

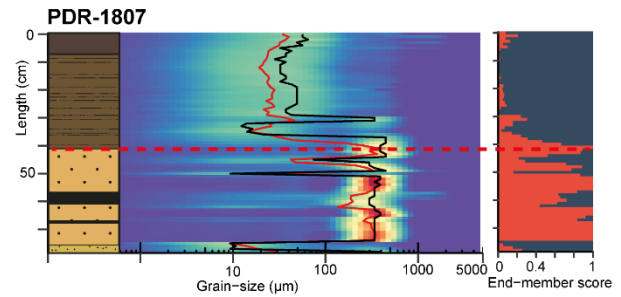
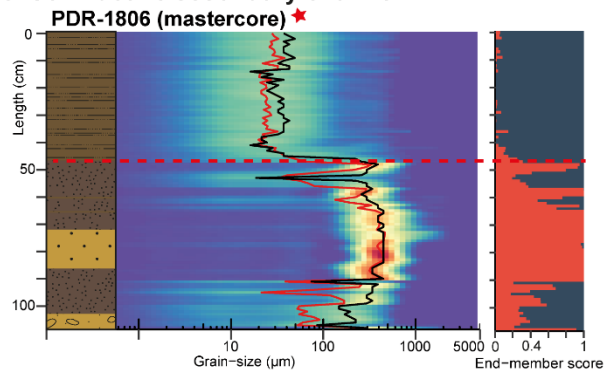
**a. Dam reservoir**



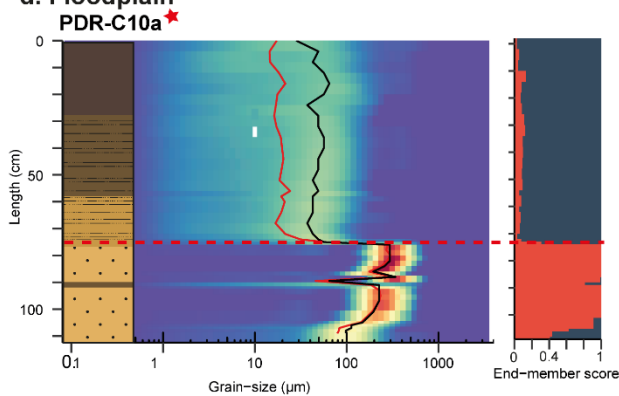
**b. Active secondary channel**



**c. Semi-active secondary channel**



**d. Floodplain**



**Legend:**

Grain-size class frequency (%):



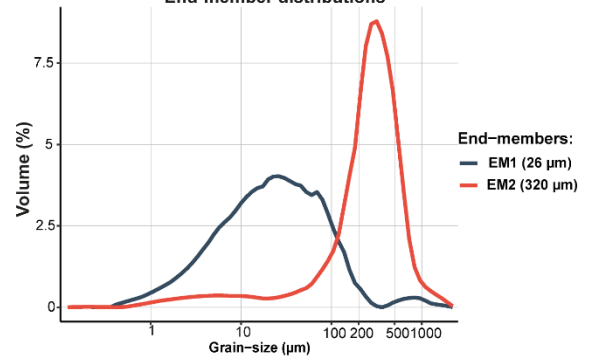
Curves:  
— D50  
— Mode

Stratigraphic legend:

- light-colored lamina
- dark organic lamina
- silt
- silt mixed with fine sand
- fine sand
- sand
- coarse sand
- coarse sand with gravels

- ★ : Sediment core dated and analysed for organic contaminants (cf. figure 4 and 5)
- - - : Grain-size limit

**End-member distributions**



344 *Figure 3: Grain-size characteristics for the seven studied sediment cores in a) the dam reservoir, b) the active secondary*  
345 *channel (Ilon), c) the semi-active secondary channel (Limony), and d) the floodplain. For each core, from left to right:*  
346 *stratigraphic description, heatmap representation of the total grain-size repartition and End-Member scores. [IN COLOR]*

347 The End-Member Modelling Analysis (EMMA) algorithm (Dietze et al., 2012) showed that the corpus  
348 of grain-size distributions (GSDs) was best represented with two end-members (EM). The main modes  
349 are at 26 and 320  $\mu\text{m}$  and represent each around 50% of the grain-size distributions (Fig. 3). This end-  
350 member model has a class-wise explained variance of 52% and a sample-wise explained variance of  
351 93%.

352 The end-member scores –i.e. their percentage of representation of the GSDs- vary depending on the  
353 sediment core and the depth of the sample (Fig. 3). In the reservoir cores (PDR-1901 and PDR-1902)  
354 and in the top part of the 5 other cores, EM1 is largely predominant, which is consistent with the fine  
355 homogeneous sediments observed in the stratigraphic descriptions and heatmaps. Below the grain-  
356 size limit, EM2 is mostly represented which corresponds to the observed sandy sediments. The grain-  
357 size limit identified in the heatmaps can be clearly seen on the end-member score graphs as well. It  
358 appears as an abrupt change in the EMs repartition and supports the observation that there is a sudden  
359 change in the grain-size properties that can be consistently observed all sediment cores except those  
360 from the dam reservoir, and might therefore be linked to river engineering. While this limit was initially  
361 derived from visual observations and grain-size data, the fact that it also appears in the EM score  
362 graphs gives it a statistical weight. Overall, the end-member modelling attests of the existence of two  
363 distinct deposition processes in the studied cores: EM1 is likely representative of a decantation process  
364 in a context of still waters, while EM2 might represent former dynamics of deposition from when the  
365 fluvial corridor was not constrained by the various engineering works.

### 366 3.3. Vertical trends in the geochemical characteristics of the sediments

#### 367 3.3.1. Vertical trends in Total Organic Carbon

368 The Total Organic Carbon (TOC) contents in the cores (Fig. 4) are comprised between 0 and 5%. The  
369 vertical trend in TOC in PDR-1902 (dam reservoir, Fig. 4.a) is monotonous at around 2%. In PDR-1802,  
370 the TOC is low (<1%) before the grain-size rupture (active secondary channel, Fig. 4.b), then increases  
371 up to 3% at 55 cm, before falling back at ~2% until the surface. In PDR-1806 (semi-active channel, Fig.  
372 4.c), the TOC content is null in the bottom of the core until ~50 cm from the top. It then increases up  
373 to 4-5% close to the surface. In PDR-C10a (floodplain, Fig. 4.d), the TOC is almost null below 75 cm,  
374 increases slowly towards the surface, and then more sharply in the last ten centimeters. It is worth  
375 noting that the increase in TOC systematically coincides with the grain-size ruptures identified  
376 previously (red dotted line in Fig. 4) in all cores.

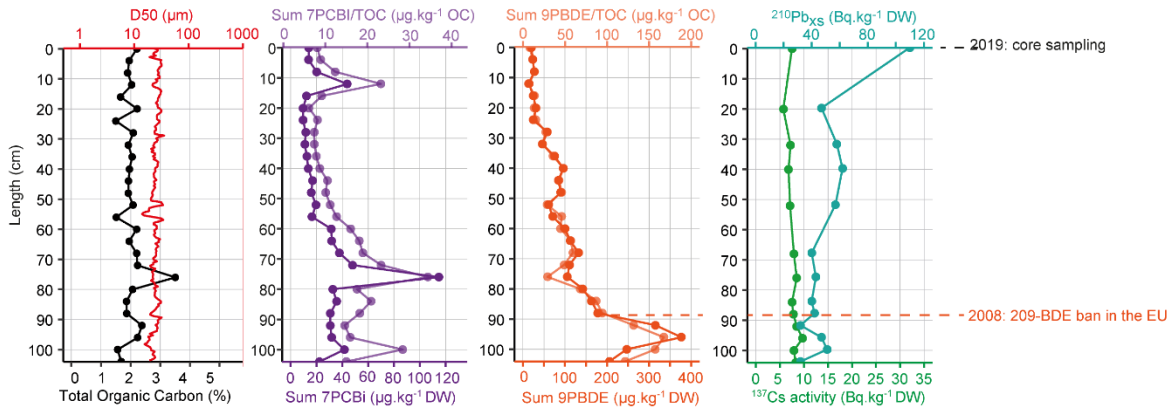
#### 377 3.3.2. Vertical trends in persistent organic pollutants

378 In core PDR-1902 (dam reservoir, Fig. 4.a), the  $\Sigma 7\text{PCBi}$  concentration follows a decreasing trend from  
379 the bottom ( $\sim 40 \mu\text{g kg}^{-1}$ ) to the top ( $\sim 10\text{-}20 \mu\text{g kg}^{-1}$ ). Two sharp peaks can however be observed at 78  
380 cm ( $120 \mu\text{g kg}^{-1}$ ) and 12 cm ( $40 \mu\text{g kg}^{-1}$ ). The  $\Sigma 9\text{PBDE}$  concentration starts around  $200 \mu\text{g kg}^{-1}$  at the very  
381 bottom of the core, peaks at  $\sim 400 \mu\text{g kg}^{-1}$  at 96 cm and then decreases steadily from  $200 \mu\text{g kg}^{-1}$  at 90  
382 cm to  $\sim 20 \mu\text{g kg}^{-1}$  close to the surface. The TOC normalization does not significantly modify the  
383 observed trends in this core.

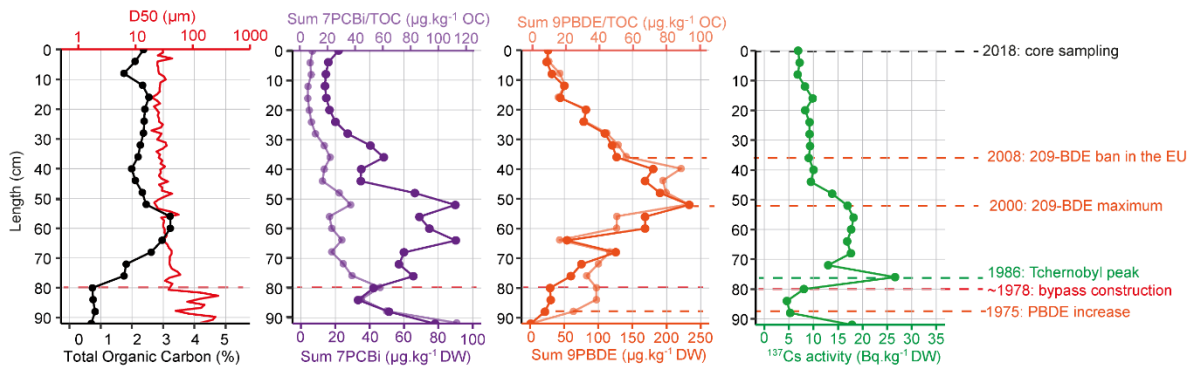
384 In core PDR-1802 (active secondary channel, Fig. 4.b), the  $\Sigma 7\text{PCBi}$  concentration is around  $80 \mu\text{g.kg}^{-1}$  at  
385 the very bottom of the core and fluctuates between  $\sim 40$  and  $90 \mu\text{g kg}^{-1}$  until 50 cm. It then decreases  
386 steadily to reach a stable value of  $20 \mu\text{g kg}^{-1}$  close to the surface. When normalizing the  $\Sigma 7\text{PCBi}$   
387 concentration to the TOC (in order to remove a potential confounding effect due to the affinity of  
388 persistent organic pollutants for organic matter), the trend is significantly modified: we observe an  
389 exponential decrease in  $\Sigma 7\text{PCBi}/\text{TOC}$  from the bottom of the core to the surface. The  $\Sigma 9\text{PBDE}$   
390 concentration is null at the very bottom but increases rapidly and peaks at  $\sim 250 \mu\text{g kg}^{-1}$  around 50 cm.

391 It decreases then steadily to reach  $\sim 25 \mu\text{g kg}^{-1}$  at the surface. The TOC normalization does not change  
392 significantly the overall vertical trend observed for PBDEs.

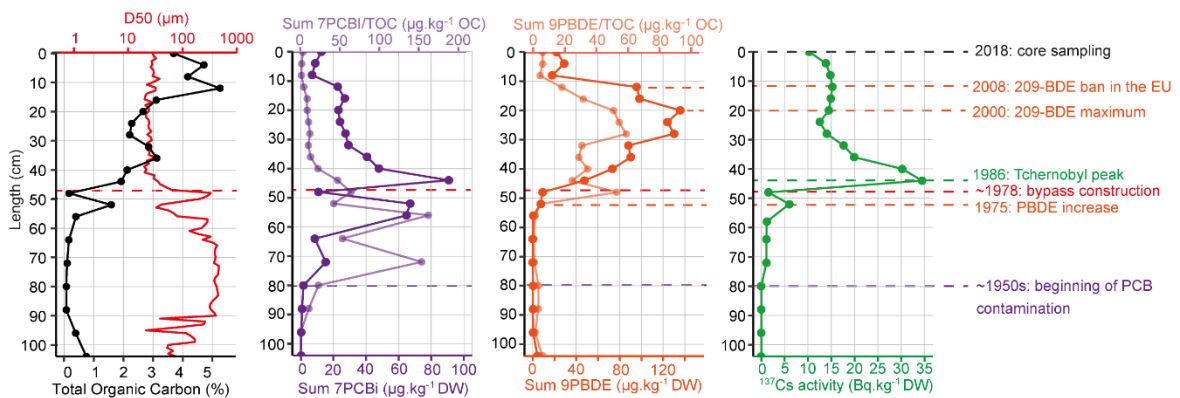
**a) PDR-1902 (dam reservoir)**



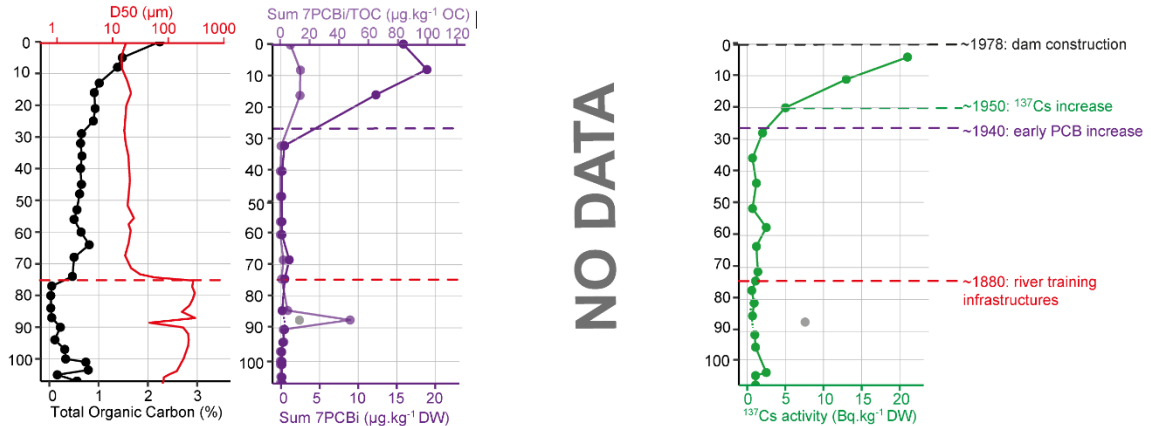
**b) PDR-1802 (active secondary channel)**



**c) PDR-1806 (semi-active secondary channel)**



**d) PDR-C10a (floodplain)**



394 *Figure 4: Vertical trends in geochemical characteristics in cores a) PDR-1902; b) PDR-1806; c) PDR-1802; d) PDR-C10a. For*  
395 *each core, the following characteristics are presented from left to right: Total Organic Carbon (TOC) content, D50, sum of the*  
396 *7 indicator PCB<sub>i</sub>, sum of 9 PBDE, <sup>137</sup>Cs activity. For organic contaminants, the darkest curve represents the raw concentrations*  
397 *while the lightest curve represents the concentrations normalized to the TOC. NB: the sum of 9PBDE was not measured in core*  
398 *PDR-C10a. [IN COLOR]*

399 The  $\Sigma 7\text{PCBi}$  concentration is null in the bottom part of core PDR-1806 (semi-active channel, Fig. 4.c)  
400 but increases quickly starting at 80 cm from the surface. It peaks at a value of  $\sim 100 \mu\text{g kg}^{-1}$  at 44 cm,  
401 then decreases steadily to reach a background value of  $\sim 5\text{-}10 \mu\text{g kg}^{-1}$  close to the surface. When  
402 normalizing the  $\Sigma 7\text{PCBi}$  concentration to the TOC, the vertical trend is significantly modified: the  
403 increase is sharper starting at 80 cm, and the peak happens lower in the core, between roughly 55 and  
404 70 cm. From 55 cm to the surface, an exponential decrease is then observed. In this core, the  $\Sigma 9\text{PBDE}$   
405 concentration is null between the bottom of the core and  $\sim 50$  cm from the surface. We can then  
406 observe a sharp increase in concentration until  $\sim 140 \mu\text{g kg}^{-1}$  around 20-30 cm. Starting at 20 cm, the  
407 concentration decreases rapidly until  $20\text{-}30 \mu\text{g kg}^{-1}$  close to the surface. The TOC normalization does  
408 not significantly impact the PBDEs vertical trend.

409 In core PDR-C10a (floodplain, Fig. 4.d), the  $\Sigma 7\text{PCBi}$  concentration is null in the bottom part of the core.  
410 The first measured concentration is  $\sim 12 \mu\text{g kg}^{-1}$  around 18 cm, and the contamination levels then stay  
411 similar until the surface. The TOC normalization shows that the contamination is minimal compared to  
412 the other cores but does not modify the tendency.

413 It is worth specifying that the congener repartition for PCBs and PBDEs is similar in all three cores and  
414 does not vary significantly with depth. The highly chlorinated PCB congeners (PCB-  
415 138/PCB153/PCB180) are predominant, generally representing between 60 and 80% of the  $\Sigma 7\text{PCBi}$ .  
416 Meanwhile, congener 209 (BDE-209, the main component of the deca-BDE commercial mixture)  
417 represents between 93 and 99% of the  $\Sigma 9\text{PBDE}$  measured in the samples and was therefore mainly  
418 considered in further interpretations.

419 **3.3.3. Radionuclides activity trends**

420 The  $^{137}\text{Cs}$  activity profile for core PDR-1902 (dam reservoir, Fig. 4.a) is monotonous and comprised of  
421 background values between 5 and 10  $\text{Bq kg}^{-1}$ . No activity peak is observed. The  $^{210}\text{Pb}_{\text{xs}}$  activity profile  
422 globally decreases from  $\sim 120 \text{ Bq kg}^{-1}$  at the top to 40-60  $\text{Bq kg}^{-1}$  at the bottom of the core. While a few  
423 measure points slightly deviate from that decreasing trend (e.g. at 20 cm), all were nevertheless taken  
424 into account for the modelling.

425 In core PDR-1802 (active secondary channel, Fig. 4.b), the  $^{137}\text{Cs}$  activity is around 20  $\text{Bq kg}^{-1}$  at the very  
426 bottom of the core and decreases onto the last 8 cm. This declining trend hints at the existence of an  
427 earlier peak that was not recorded in the core. The activity then increases to peak at 25  $\text{Bq kg}^{-1}$  around  
428 76 cm. Afterwards, it decreases quickly between 44 and 78 cm, and more progressively between 44  
429 cm and the surface, where the activity stalls around 5  $\text{Bq kg}^{-1}$ .

430 The  $^{137}\text{Cs}$  activity in core PDR-1806 (semi-active secondary channel, Fig. 4.c) is null from the bottom of  
431 the core to 80 cm from the surface, then very low ( $< 5 \text{ Bq kg}^{-1}$ ) until 58 cm. We can then observe two  
432 activity peaks of different intensities: a 6  $\text{Bq kg}^{-1}$  peak at 52 cm, followed by a temporary decrease and  
433 more straightforward peak of 35  $\text{Bq kg}^{-1}$  at 44 cm. Above this second peak, the  $^{137}\text{Cs}$  activity decreases  
434 overall to reach 10  $\text{Bq.kg}^{-1}$  at the surface, although a slight increasing trend can be observed again  
435 between 12 and 25 cm.

436 In core PDR-C10a (floodplain, Fig. 4.d),  $^{137}\text{Cs}$  activity is null in the bottom of the core and increases from  
437 20 cm onwards. It starts at around 5  $\text{Bq kg}^{-1}$  and reaches 20  $\text{Bq kg}^{-1}$  close to the surface.

### 438 3.4. Age-depth modelling

#### 439 3.4.1. Date markers

440 The grain-size change identified in cores from the secondary channels (PDR-1802 and PDR-1806; cf.  
441 section 3.3) and the floodplain (PDR-C10a) can be used as a date marker. Indeed, a similar change has  
442 already been observed in sediment cores from the Rhône floodplain, in this study area and two other  
443 sites along the river corridor (Vauclin et al., 2019; Vauclin et al., 2020). In those cases, it was proven

444 that the abrupt grain-size modification was due to the first period of river engineering (~1880) that  
 445 created a partial disconnection between the channel and the floodplain, and sediments above the limit  
 446 were therefore defined as infrastructure-induced legacy sediments. This mechanism is illustrated here  
 447 in the floodplain core (PDR-C10a). In the cores from the secondary channels, however, the grain-size  
 448 change is located just below <sup>137</sup>Cs activity peaks and therefore has to be more contemporary than the  
 449 1880s. We therefore concluded that it was caused by the second period of river engineering (bypass  
 450 implementation) in 1978. As a consequence, we considered that the <sup>137</sup>Cs activity peaks in cores PDR-  
 451 1802 and PDR-1806 were the result of the 1986 Tchernobyl accident since both are located above the  
 452 1978 grain-size limit.

453 Tipping points in organic contaminants trends were used as secondary date markers in models M2 (in  
 454 addition to absolute date-markers, cf. section 2.3.4), based on production/consumption temporal data  
 455 or already published dated contamination trends from other sedimentary archives. Standard errors  
 456 were associated with POPs time-markers to take into account the lag time between the legislation, its  
 457 implementation at the national scale and the time needed to record a decrease in the sediment record  
 458 (Table S2). All date markers used in this study and their justification and/or reference are available in  
 459 Table 1.

Date marker	Date	Sediment cores	Reference/explanation
<b>Absolute date markers</b>			
<sup>137</sup> Cs activity peak	1986	PDR-1806, PDR-1802	See section 3.5.1
Grain-size change	1978	PDR-1806, PDR-1802	See section 3.5.1
<b>Secondary date markers</b>			
PCB concentration increase	1950	PDR-1806	Brevik et al., 2002b; Desmet et al., 2012; Lorgeoux et al., 2016; Dendievel et al., 2020

PBDE concentration increase	1975	PDR-1806, PDR-1802	Nylund et al., 1992; Muir and Rose, 2007; Kohler et al., 2008; Lorgeoux et al., 2016; Abbasi et al., 2019
209-BDE maximum	2000	PDR-1806, PDR-1802	Kohler et al., 2008; Abbasi et al., 2019
209-BDE ban in the EU	2008	PDR-1806, PDR-1802, PDR-1902	European Parliament, 2002/95/CE, European Court of Justice ruling, April 1 <sup>st</sup> 2008

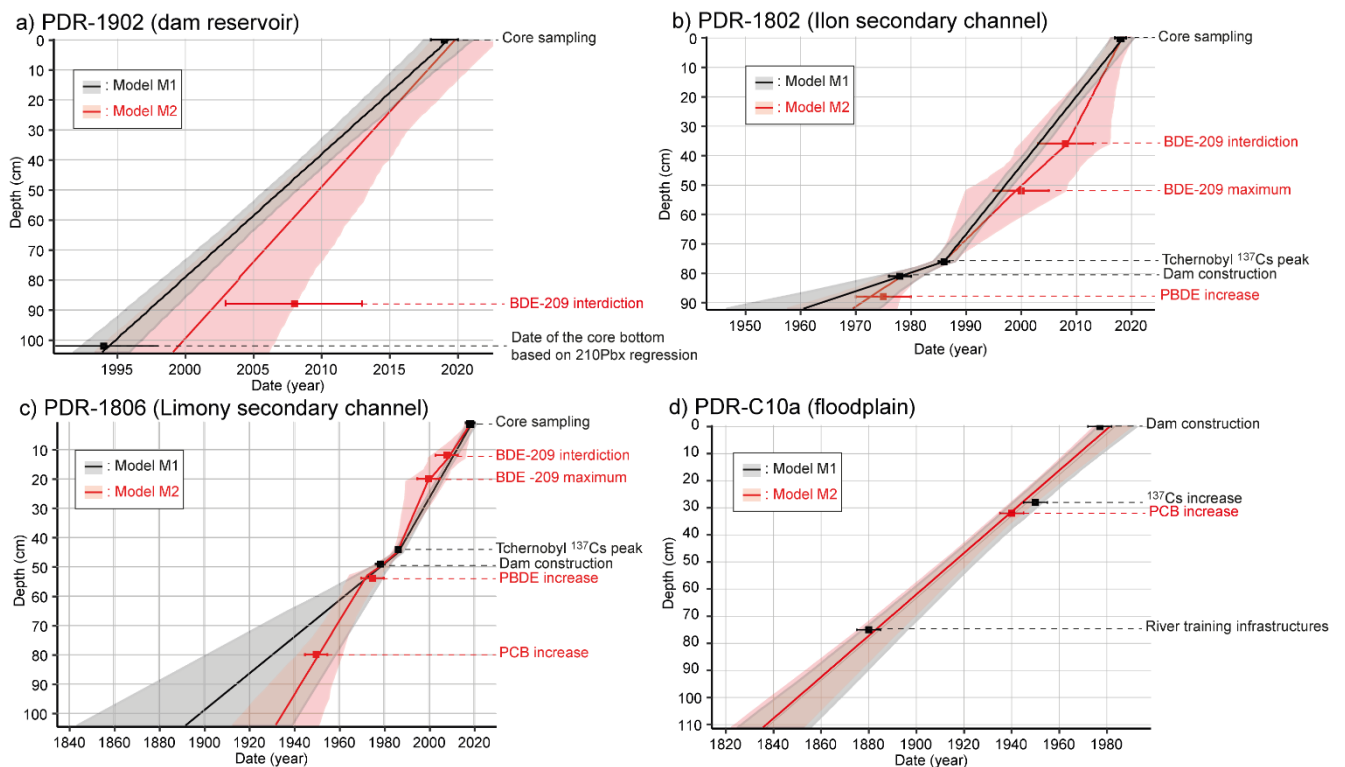
460 *Table 1: Date markers used to build the age-depth models*

### 461 3.4.2. Age models

462 Model M1 for core PDR-1902 was developed from <sup>210</sup>Pbxs modelling exclusively. The coefficient of  
463 determination of the model was  $R^2=0.61$ , and the resulting sedimentation rate in the reservoir  
464 computed from the regression slope is  $4.12 \text{ cm yr}^{-1}$ . According to this model, the bottom of core PDR-  
465 1902 would date back to 1993 (Fig. 5.a). Model M2 additionally considers the PBDE decreasing trend  
466 in the core and results from a linear regression. It estimates a constant sedimentation rate of  $5.0 \text{ cm}$   
467  $\text{yr}^{-1}$  in the reservoir, the bottom of the core therefore dating back to 1999. While both models give  
468 relatively similar results, the bathymetric measurements (Fig. S2) suggest that the bottom of the core  
469 is older than 1999, which renders model M1 more plausible.

470 Models M1 and M2 for core PDR-1802 were obtained with a linear interpolation model (Fig. 5.b).  
471 According to model M1, the bottom of the core dates back from 1960. The resulting sedimentation  
472 rate is around  $2.3 \text{ cm yr}^{-1}$  between 1986 and 2018. Model M2 gives very similar results to model M1:  
473 the date of the bottom of the core is estimated at 1969 and the sedimentation rate is around  $\sim 1.2$   
474  $\text{cm}\cdot\text{year}^{-1}$  until 1986,  $1.7\text{-}1.8 \text{ cm yr}^{-1}$  between 1986 and 2006 and  $3.2 \text{ cm yr}^{-1}$  between 2006 and the  
475 surface. The consistency between the two models suggest that the dating of core PDR-1802 is overall  
476 valid, although the dating of very bottom of the core beyond the older time-marker should be

477 considered with caution as the results had to be extrapolated. This also indicates that the additional  
 478 time-markers linked to the PBDE contamination trend in model M2 are reliable in this case.



479

480 *Figure 5: Age-depth models for cores PDR-1902, PDR-1802, PDR-1806 and PDR-C10a. [IN COLOR]*

481 Models M1 and M2 for core PDR-1806 were obtained with a linear interpolation model (Fig. 5.c).  
 482 According to model M1, the bottom of the core dates back from 1890, however this date results from  
 483 a large extrapolation -the latest time-marker being positioned at 48 cm- and is therefore unreliable.  
 484 The sedimentation rate is estimated at  $0.6 \text{ cm yr}^{-1}$  between the bottom of the core and 48 cm (time-  
 485 marker:  $^{137}\text{Cs}$  peak 1986), then increases at  $1.3 \text{ cm yr}^{-1}$  until the surface. Model M2 gives a very similar  
 486 estimation for the top 48 cm of the core, with a sedimentation rate comprised between 1 and  $2 \text{ cm yr}^{-1}$   
 487 between 1986 and 2018. Between 50 and 104 cm, however, the slope of the modelled line is steeper,  
 488 with an estimated  $1.4 \text{ cm yr}^{-1}$  sedimentation rate. According to this model, the bottom of the core  
 489 dates back from 1933, although it should still be considered with caution as it results from  
 490 extrapolation. As the two models give consistent results for the top 48 cm of the core, we can assume  
 491 that the dating of this section is reliable. The two additional time-markers related to the PBDE

492 contamination trend in model M2 therefore appear as accurate. Regarding the bottom half of the core,  
493 model M2 is likely the most accurate of the two, as the beginning of PCB contamination in sediments  
494 is widely accepted as dating back from the early 1950s (Wania, 1999; Breivik et al., 2002a; Breivik et  
495 al., 2002b) and such a trend has already been observed in sediment core from the Rhône River (Desmet  
496 et al., 2012; Mourier et al., 2014).

497 The best results for the age-depth modelling of core PDR-C10a (Fig. 5.d) were obtained with a linear  
498 regression model. Only one POPs time-marker is additionally used in model M2 compared to M1,  
499 therefore the two models are practically identical. A consistent sedimentation rate of  $0.76 \text{ cm yr}^{-1}$  is  
500 estimated in both cases, with the bottom of the core dating back from 1840. However, the last reliable  
501 time-marker is the grain-size change resulting from the river training infrastructures implementation  
502 in the area around 1880, at 75 cm. Below this point, the model results are highly uncertain as they  
503 stem from extrapolation.

## 504 4. Discussion

### 505 4.1. Multi-proxy dating in a complex fluvial setting

506 Dating often constitutes the basis for the interpretation of other information recorded in a sediment  
507 sequence, such as identifying contamination trends or relating grain-size patterns to hydrological  
508 events. However, achieving a robust dating of sediments from a complex fluvial setting is challenging  
509 and requires extensive interpretation in its own. Two types of dating methods can be distinguished  
510 based on the information they provide (Notebaert et al., 2011): discrete methods such as radiocarbon,  
511 Optically Stimulated Luminescence or  $^{137}\text{Cs}$  will provide an absolute date for a few discrete points in  
512 the core, while continuous methods based on the presence and evolution of tracers -for instance  
513 contaminant trends- will allow to identify time periods based on their vertical repartition in the  
514 deposits. In fluvial contexts, several processes might render these dating methods ambiguous:  
515 sedimentation hiatuses due to incision, erosion, or the intermittent nature of a depositional  
516 environment (e.g. floodplain), fluctuations of sediment accumulation rate, post-depositional processes

517 (e.g. bioturbation, contaminants/radionuclides migration or leeching), etc. (Lewin and Macklin, 2003;  
518 Bábek et al., 2008; Matys Grygar et al., 2016). The different dating methods will be more or less  
519 affected by those processes: the use of  $^{137}\text{Cs}$  activity measurements might prove little informative if a  
520 sedimentation hiatus coincides with an activity peak or if the sediments were deposited after 1986,  
521 while a varying sedimentation rate is especially misleading when trying to infer a time period from a  
522 known contamination trend.

523 A combination of complementary methods is therefore optimal for a robust dating of a sediment  
524 sequence in a complex fluvial system, as illustrated in this study where no less than five types of time-  
525 markers ( $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  activity, contamination trends, grain-size changes, bathymetric  
526 measurements) were necessary to build reliable age-depth models for each of the four depositional  
527 environments. Indeed, few discrete time-markers were available in our samples: the deposits are too  
528 recent for radiocarbon dating to give relevant results, and clear  $^{137}\text{Cs}$  peaks could only be identified in  
529 two out of the four depositional environments. We therefore had to use continuous and less robust  
530 indicators such as contamination trends. PCBs temporal trends in sediments are reasonably well-  
531 known, having been studied in many European Rivers (Bábek et al., 2008; Malina et Mazlova, 2017;  
532 Lorgeoux et al., 2016, Dendievel et al., 2020) including the Rhône River (Desmet et al., 2012, Mourier  
533 et al., 2014). They started being produced in the 1930s but the contamination often appears in  
534 sediments from the 1950s only (Lorgeoux et al., 2016, Dendievel et al., 2020). Their emissions peaked  
535 in 1975 in France (Breivik et al., 2002b), which coincides with the prohibition of their use in open  
536 systems, and then decreased. They were then banned from use and sale in closed system as well in  
537 1987. In our case, unfortunately, only core PDR-1806 recorded the complete PCB temporal trend, and  
538 the large discrepancy between the concentration and the TOC-normalized trends (Fig. 4.b) rendered  
539 unreliable any date indication other than the initial increase in concentrations (~1950s). PBDEs have  
540 been less investigated and there is no consensus yet about their temporal repartition in sediment  
541 deposits. However, their concentrations follow a well-defined pattern that seems consistent in the  
542 cores from the aquatic environments (Fig. 4.a, 4.b and 4.c), meaning they could be used as geochemical

543 tracer. As for PCBs, their use is restricted in time: they started being produced in 1970 (Lauzent, 2017),  
544 and were progressively banned from use and production by the European Union from 2004 to 2008  
545 (European Parliament, 2002/95/CE). Additionally, results from dated sediment cores (Nylund et al.,  
546 1992, Zegers et al., 2003, Muir and Rose, 2007) suggest a contamination peak in the early to late 1990s  
547 depending on the congener, BDE-209 tending to peak in the early 2000s as the use of the deca-mix  
548 was regulated later than the others.

549 Relying on contamination trends -especially ones that were less studied- is risky to date sediment  
550 archives in such a complex setting, therefore our approach was to construct two separate age-depth  
551 models for the aquatic cores: model M1 from the absolute time-markers only ( $^{137}\text{Cs}$ , grain-size changes,  
552 sampling date), inevitably imprecise but reliable; and model M2 using contamination trends as  
553 additional time-markers. Both models ended up being similar in the four cores, which proves the  
554 robustness of our approach. In the case of PDR-1806, the use of PCBs contamination trend even  
555 allowed to significantly improve the age-depth model in the lowest part of the core. This methodology  
556 also confirms the potential for PBDE contamination trends to be increasingly used as time-markers in  
557 sediment deposits, although larger-scale studies would be useful to ascertain it. Finally, the lack of  
558 time-markers in the most recent core (PDR-1902, Fig. 4.a) highlights the interest of investigating  
559 emergent contaminants trends in order to retain the ability to date future sediment cores where  
560 conventional date-markers such as  $^{137}\text{Cs}$  peaks and historical organic contaminants may not be  
561 present.

## 562 4.2. Characterization of the depositional environments

563 One of the aims of this study is to demonstrate that a thorough characterization of the fluvial  
564 depositional environment from which a sediment core will be sampled is essential and time efficient.  
565 While the four depositional environments have been previously described from a geographical and  
566 geophysical point of view, this section intends to formally characterize these environments based on

567 previous geomorphology studies and illustrate incidentally the interest of such characterization for  
568 sediment archive interpretation.

569 The dam reservoir environment in this study cannot readily be compared to most reservoirs in terms  
570 of depositional processes, as the run-of-river dam is punctually erased during floods and few sediments  
571 are therefore deposited in the axis of the dam (Fig. S2). The coring zone where sediments accumulated  
572 corresponds to a recess zone where there is little flow velocity under normal discharge conditions and  
573 is likely closer to a harbour basin in terms of context. The environment was never dredged and there  
574 is no sedimentological nor bathymetric evidence to show that sediment remobilization might have  
575 happened during floods: the core sediments are fine (silt to clay) and homogeneous, and the  
576 bathymetric lines are in chronological order. Few date markers were available in the core sampled in  
577 the reservoir, which renders essential the knowledge that sedimentation is likely continuous and  
578 sustained in time in this environment: it supports the use of  $^{210}\text{Pb}$ s decay and a linear regression age-  
579 depth modelling to establish the chronology of the core.

580 The Ilon (active) secondary channel was affected by the river training phase in the 1880s: the upstream  
581 end of the larger secondary channel to which the Ilon channel is connected to was barred by a dyke  
582 (Vauclin et al., 2019, Tena et al., 2020). Later on, the dam implementation quickly resulted in the  
583 disconnection and siltation of the upstream part of the channel, which is a common consequence of  
584 bypassing in the Rhône River as demonstrated in Dépret et al. (2019). Both phases can be identified  
585 on the GPR profile due to a distinct and continuous subparallel reflector (dotted line on Fig. 2.a) that  
586 matches temporally with the dam-induced legacy sediment limit in the cores (Fig. 3.b). While the grain-  
587 size analysis shows that sediments below the limit are coarser (fine sand) than those above (silt), both  
588 units appear similar on the GPR survey: poorly defined subparallel reflectors that are indicative of a  
589 calm deposition process. A deeper subparallel reflector is speculated to correspond to the pre-  
590 infrastructure gravel-bed (Fig. 2.a). Overall, this depositional environment is therefore characteristic  
591 of a passive channel fill (Aslan, 2013). The river training infrastructure initiated the filling, and the

592 severe flow reduction associated with bypassing emphasized the disconnection, resulting in the  
593 deposition of finer sediment without altering the sedimentary architecture. Understanding the  
594 chronology of the filling process of this environment was invaluable to reinforce the dating of the  
595 corresponding sediment core.

596 The Limony (semi-active) secondary channel was artificially disconnected from the main channel by  
597 the building of dykes at its upstream and downstream ends in the 1880s (Vauclin et al., 2019; Tena et  
598 al., 2020). Both the sediment cores and the GPR results prove that two distinct phases of infilling  
599 followed. The first phase is characterized by numerous oblique and concave reflectors on the GPR  
600 profile (Fig. 2.b) and fine to medium sands in the corresponding section of the sediment cores (Fig.  
601 3.c). Both aspects are indicative of an active fill (Aslan, 2013), with deposition of bedload elements and  
602 possible migration of bar or dune forms recorded as the oblique reflectors. The second phase is  
603 characterized by subparallel/sinuuous reflectors (Fig. 2.b) and silt deposits (Fig. 3.c), indicating a more  
604 passive fill due to severe flow reduction (Aslan, 2013) -which in this case is due to the dam  
605 implementation in 1978. Dépret et al. (2019) also found that water level lowering following bypassing  
606 in the Rhône River resulted in a rapid dewatering process, which is confirmed here by the sharp  
607 increase in organic carbon content in this layer (Fig. 4.c). This depositional environment is therefore a  
608 hybrid of an active and passive channel fill, strongly influenced by river engineering. This interpretation  
609 was essential to identify the grain-size change as a date-marker that contributes to the robust dating  
610 of core PDR-1806. Additionally, it is worth noticing that its intermittent connection to the main channel  
611 might have resulted in a discontinuous sediment record.

612 Finally, the floodplain environment described in this study could be defined as a floodbasin: the lowest  
613 lying parts of a floodplain that act as a stilling basin in which suspended fines can settle from overbank  
614 flows after the coarser suspended debris have been deposited on levees (Aslan, 2013; Allen, 1965).  
615 Sediments from the floodplain environment in our study area are indeed fine (Fig. 3.d) and the absence  
616 of reflectors in the upper layer on the corresponding GPR profile (Fig. 2.c) is an indication of a

617 decantation process, resulting in a homogeneous sedimentary architecture with no tangible reflector.  
618 In our case, the screening of coarser materials has been emphasized by the implementation of in-  
619 channel infrastructures (Vauclin et al., 2019). Floodbasins often exhibit a network of small channels  
620 (Allen, 1965), which used to be the case in the studied area, as proven by the two paleochannels clearly  
621 identifiable on GPR10 (Fig. 2.c). Those filled channels are not discernible on the field nor recorded in  
622 the historical maps, likely due to their small size: this demonstrates the interest of using GPR or another  
623 geophysical method to understand the sedimentary context of a sediment core.

#### 624 4.3. Influence of depositional environments on sediment archives

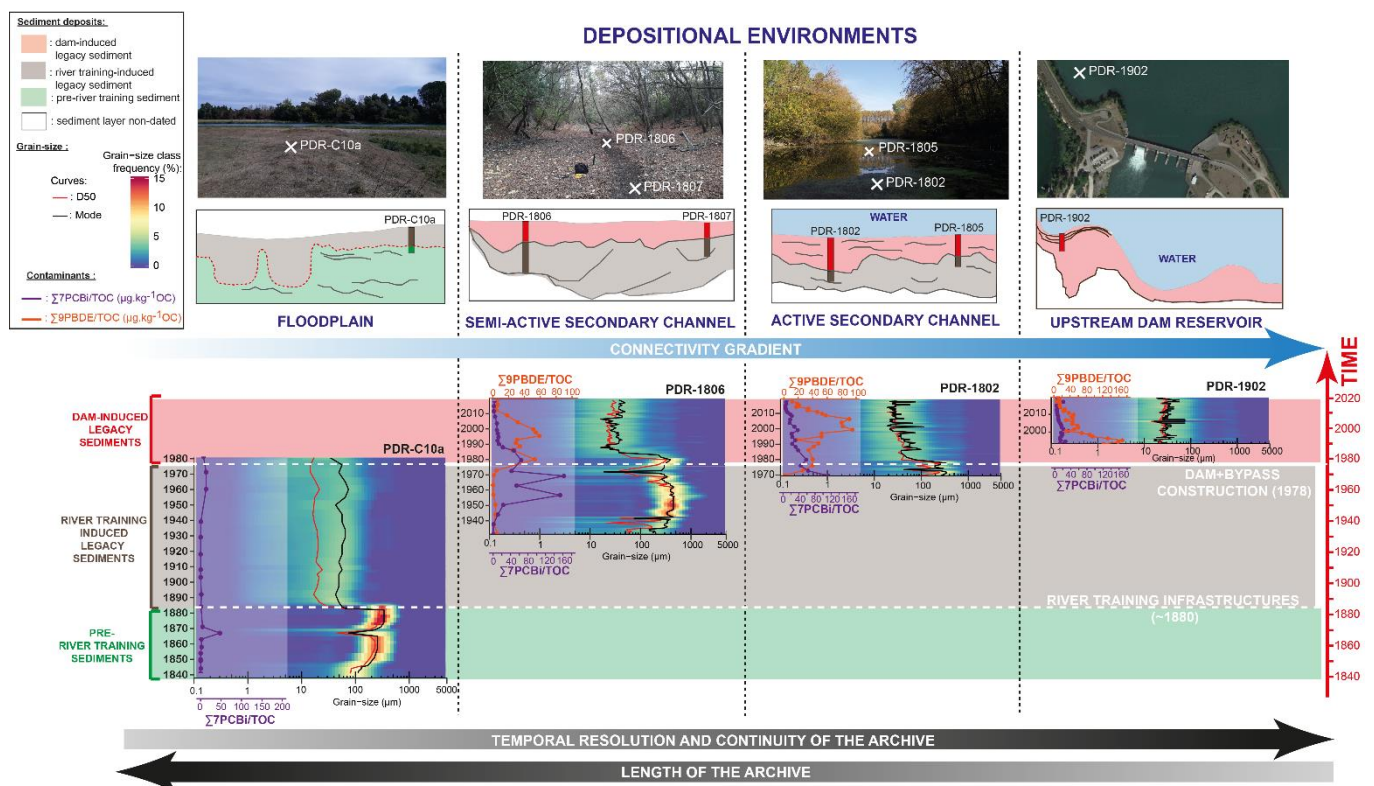
625 When studying a sedimentary archive, one is ideally looking for a compromise between a good  
626 temporal resolution and a long-term record, in an environment as preserved as possible. Those  
627 features depend strongly on the depositional environment and are influenced by natural and  
628 anthropogenic disturbances. As a consequence, finding a good sampling site for a sediment core is  
629 rarely straightforward and requires investigation and/or a good knowledge of the study area.

630 Floodbasins have often been used as archives of historic contamination as they are less prone to  
631 reworking than channel deposits and thus constitute long-term records of fine-grained sediments that  
632 efficiently retain contaminants (Matys Grygar et al., 2016). The counterpart is that the record is most  
633 likely intermittent, sediments being deposited in high-discharge conditions only. An estimation of the  
634 overflow frequency is therefore essential for the interpretation of sedimentary sequences from such  
635 an environment. In aggrading or corseted systems, the floodplain might not be actively flooded  
636 anymore, meaning that even sediments close to the surface could be a few decades old. In our case,  
637 the floodplain has been entirely disconnected from the channel since the late 1970s, and interpreting  
638 the pollution trend from core PDR-C10a (Fig. 4.d) as the whole contamination chronicle would have  
639 led to a wrong estimation of the pollution levels and dynamics in the area. Lewin and Macklin (2003)  
640 noted that channel stability may cause overbank sedimentation to become self-limiting as the  
641 floodplain aggrades and progressively becomes too elevated to be overflowed, and therefore reduces

642 over time the preservation potential of the floodplain environment. This is consistent with our  
643 observations following the river training infrastructures implementation: the decline in floodplain-  
644 channel connectivity was first translated into the deposition of finer-grained legacy sediments (Vauclin  
645 et al., 2019), and was later exacerbated by the severe flow reduction associated with the dam  
646 construction. River engineering therefore caused the loss of a recording environment, but it should  
647 also be noted that the river training infrastructures stopped lateral channels migration, facilitating  
648 sediment deposition without major reworking in the margins for several decades. Despite the recent  
649 hiatus, the floodplain in our study still displays the longest sediment record (~1840s-1970s, cf. Fig. 6),  
650 and therefore remains an essential sedimentary archive when properly interpreted.

651 Abandoned or partly disconnected channels are often considered as a good alternative to floodplains  
652 to study recent river history (Bábek et al., 2008, Van Metre et al., 2008; Van Metre et al., 2015; Mourier  
653 et al., 2014) as they are usually characterized by a reasonably good preservation potential and a better  
654 stratigraphic resolution and continuity than floodplains due to their higher connectivity with the main  
655 channel. The two secondary channels we studied showed similar recording tendencies but were also  
656 influenced by the respective timings and dynamics of their disconnection. The temporal extent of the  
657 archive is related to the date of the disconnection: the secondary channel that was partly disconnected  
658 from the 1880s onward recorded a longer chronicle (1930s-2018) than the other channel (1960s-2018)  
659 whose partial disconnection is mainly related to the bypass implementation in the 1970s. This  
660 illustrates again the influence of human-induced river modification on the recording ability of the  
661 various depositional environments. Additionally, the temporal resolution of the archive is linked to the  
662 connection frequency of the channel environment (Fig. 6): the Ilon channel which is connected most  
663 of the year has an accumulation rate more than twice as high ( $\sim 3.2 \text{ cm}\cdot\text{year}^{-1}$ ) as the Limony channel  
664 ( $\sim 1.3 \text{ cm}\cdot\text{year}^{-1}$ ) that is only connected under high discharge conditions. Finally, the nature of the  
665 channel fill influences the continuity and reliability of the archive: a passive fill such as in top parts of  
666 the secondary channel cores ensures a deposition without reworking and therefore an archive without

667 significant hiatus, while an active channel fill -such as in the bottom half of core PDR-1806- is more  
 668 subject to sediment remobilization and errors in the archive interpretation.



669

670 *Figure 6: A summary of the influence of depositional environments on sedimentary records: contamination trends and grain-*  
 671 *size events recorded depending on the environment (from left to right: floodplain, semi-active secondary channel, active*  
 672 *secondary channel and upstream dam reservoir). [IN COLOR]*

673 A fully anthropogenic depositional environment such as the run-of-river dam reservoir in this study  
 674 presents the highest temporal resolution and continuity, allowing to reliably discern subtle grain-size  
 675 and contamination variations. It is however temporally limited to the date of construction of the  
 676 infrastructure and might pose more practical challenges for the retrieving of sediment cores because  
 677 such environments tend to accumulate large amounts of sediments due to their proximity to the main  
 678 stream. Indeed, we did not manage to secure a core longer than ~1m although we know from the  
 679 bathymetric measurements that more than 5 m of sediments were stored in our sampling location.

680 Overall, each of the four depositional environments investigated in this study present strengths and  
 681 weaknesses as potential sediment archives. It is highly unlikely to come across a depositional

682 environment in a fluvial system that satisfies all the requirements of an ideal sedimentary archive:  
683 long-term record, good temporal resolution and continuity (i.e. absence of hiatuses). However, it is  
684 possible to reconstruct a composite archive by correlating several dated sedimentary sequences  
685 sampled in different deposition environments in a common study area, as illustrated in Fig. 6. While  
686 demanding, this methodology provides a robust archive that accounts for the spatial and temporal  
687 variability of the preservation potential of recent alluvial sediments. This is especially relevant when  
688 trying to understand the evolution of a complex and highly anthropized fluvial system such as the  
689 Rhône River, as river engineering tends to strongly modify the functioning of existing depositional  
690 environments (floodplain, secondary channels) and create new ones (reservoir), making the  
691 interpretation of sediment sequences even more challenging.

## 692 5. Conclusion

693 This work demonstrates the strong influence of the type of depositional environment on the sediment  
694 archive potential in a fluvial context. Indeed, the comparative and multi-compartments approach  
695 implemented in this study proved that the four studied environments recorded four different time-  
696 periods, and therefore different hydromorphological events and contamination trends. The different  
697 archives also show various degrees of continuity and temporal resolution that can only be inferred  
698 from a thorough understanding of the depositional environment and its functioning. For this purpose,  
699 the combination of geophysical surveys with sediment core following a rigorous sampling design is  
700 optimal, and we believe that such combination should be applied more frequently when investigating  
701 sediment deposits in actively shifting or adjusting river corridors. This is especially true in highly  
702 anthropized hydrosystems, as we proved that river engineering modified the recording potential of all  
703 four studied environments. The characteristics of a depositional environment can also provide  
704 invaluable information to elaborate or specify age-depth models of sedimentary archives, a task that  
705 can otherwise prove challenging in a fluvial context, especially with contemporary cores where  
706 conventional date-markers ( $^{137}\text{Cs}$  activity peaks, historical contaminants trends) might not be

707 recorded. In a context of growing environmental concerns regarding hydrosystems, a better  
708 understanding of the impact of depositional environments on the recording potential of fluvial  
709 sediment is essential. It may allow to correctly identify areas with a higher contamination and/or  
710 remobilisation risk, but also to assess filling dynamics and the prospective need for restoration or  
711 maintenance work such as dredging. It is an indispensable tool for current and future monitoring of  
712 complex river systems that are continually evolving in association with anthropogenic activities.

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