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► To cite this version:

Yoann Copard, Frederique Eyrolle, Cécile Grosbois, Hugo Lepage, Loic Ducros, et al.. The unravelling of radiocarbon composition of organic carbon in river sediments to document past anthropogenic impacts on river systems. *Science of the Total Environment*, 2022, 806, pp.150890. 10.1016/j.scitotenv.2021.150890 . hal-04181561

HAL Id: hal-04181561

<https://hal.science/hal-04181561v1>

Submitted on 22 Jul 2024

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2 **The unravelling of radiocarbon composition of organic**
3 **carbon in river sediments to document past anthropogenic**
4 **impacts on river systems**

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15

16 **Highlights**

17 - Aquatic POC in sedimentary archives does not record ¹⁴C from the nuclear industry

18 - $\Delta^{14}\text{C}$ values of terrestrial POC as a tool to determine the transit time of sediments
19 within the watershed

20 - $\Delta^{14}\text{C}$ as a useful tool to document the trajectories of social and economic
21 milestones.

22

23 **Acknowledgement**

24 This work was supported by the Institute for Radioprotection and Nuclear Safety (France)
25 and the NEEDS-Environment funding (PALYNO project, 2017-2018). POC and Palynofacies
26 were performed at the Institut des Sciences de la Terre d'Orléans laboratory. Warm thanks

27 are addressed to Marielle Hatton, Jean-Paul Bakyono, Franck Giner, David Mourier and
28 Cédric Le Corre for their help during field operation, sample preparation and sample
29 analyses.

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The unravelling of radiocarbon composition of organic carbon in river sediments to document past anthropogenic impacts on river systems

ABSTRACT

As carriers of dissolved and particulate loads that connect continental surfaces to oceans, river systems play a major role in the global carbon cycle. Indeed, riverine particulate organic carbon (POC) is a melange of various origins characterized by their own ^{14}C labeling. In addition, civil nuclear activities have brought new ^{14}C source that remains poorly documented. We propose to unravel the $\Delta^{14}\text{C}$ value of POC stored in a sedimentary archive collected downstream the most nuclearized European rivers (the Loire River). We postulate that riverine POC is a mixture of aquatic POC (which could be impacted by the liquid discharge from nuclear industry), terrestrial and petrogenic POC. With a combination of radiocarbon measurements, POC analyses and the palynofacies method, we assessed the respective $\Delta^{14}\text{C}$ value of the POC origins. The gaps between the $\Delta^{14}\text{C}$ values of the sedimentary POC and those of the atmosphere were the result of the dilution from dead-C, the freshwater reservoir effect imprinting the $\Delta^{14}\text{C}$ of aquatic POC and the age and transit time of terrestrial POC within the catchment. Importantly, we consider that the unraveling of radiocarbon composition of riverine POC could be useful to determine either the transit time of material from source to sink, some past industrial or natural events, the resilience of the river system and milestones of the social and economic trajectory of a catchment. For the last three decades, riverine sediments could also act as a source of radiocarbon for the atmosphere.

27 1. INTRODUCTION

28 Since the peak atmospheric ^{14}C level, which occurred in 1963 after nuclear tests, the
29 atmospheric ^{14}C content has been gradually decreasing mainly due to carbon redistribution
30 within the atmosphere and through oceanic and continental pumping (Levin and Hesshaimer,
31 2000; Naegler and Levin, 2009). This decrease was disrupted by fossil fuel emissions
32 containing depleted ^{14}C , which exacerbated the decrease in the $^{14}\text{C}/^{12}\text{C}$ ratio in the
33 atmosphere (Sundquist, 1993; Soon et al., 1999; Levin and Hesshaimer, 2000). As an
34 example, Levin and Hesshaimer (2000) reported that fossil gas emissions contributed to 50%
35 of the reduction in global atmospheric $^{14}\text{CO}_2$ levels in the 2000s. These studies also indicate
36 that since the early 1990s, the biospheric organic compartment that was previously subjected
37 to atmospheric global fallout from nuclear tests has redistributed the ^{14}C that was initially
38 trapped due to mineralization processes, constituting a delayed ^{14}C source for the
39 atmosphere.

40 In rivers, organic carbon (OC), is either associated to suspended particulate matter (i.e.
41 POC, expressed in wt. %) or to sedimentary particles (i.e. sedimentary OC, in wt. %), and
42 comes from various origins, including aquatic organic matter from primary production and
43 terrestrial organic matter from the catchment (soils, superficial formations, sedimentary
44 rocks) delivered to hydrosystems by surface processes (e.g. Marwick et al., 2015). During
45 the photosynthesis pathway that occurs in rivers, the former generally reflects ^{14}C levels
46 ranging from those characterizing the atmosphere to those of the Freshwater Reservoir
47 Effect (FRE) depending on water mass circulation and mixing at the watershed scale (e.g.
48 MacDonald et al., 1987). As a consequence, the radiocarbon concentration in aquatic POC
49 may not be in equilibrium with that of the atmosphere, depending on the levels of dissolved
50 carbonate in water - originating from the weathering of carbonate rocks outcropping in the
51 catchment and / or from groundwater - and may produce CO_2 depleted in ^{14}C (e.g.
52 MacDonald et al., 1987, Ascough et al. 2010); this process is called the hard water-effect
53 (Zhou et al., 2015). In addition, downstream nuclear power plants (NPPs), this aquatic

54 autochthonous POC, is expected to integrate part of the ^{14}C released by NPPs during
55 photosynthesis and may be theoretically enriched in ^{14}C when compared to atmospheric
56 levels. Terrestrial POC can be divided into three main sources: (i) soil POC (POCs) with
57 various ^{14}C concentrations, (ii) POC from various origins held in superficial formations (more
58 and less aged riverine deposits or colluvium) with various and even depleted ^{14}C
59 concentrations (reworked POC, POC_{rw}), and (iii) petrogenic POC (POC_p) devoid of ^{14}C ,
60 which originates from sedimentary rocks or sediments older than 60 ky (dead carbon, e.g.
61 Leithold et al., 2006, Copard et al., 2018). The ^{14}C content in both POCs and POC_{rw} vary
62 with the age of the compounds (e.g., Schmidt et al., 2011) and reflect the ^{14}C content of the
63 atmosphere during their photosynthesis. Prior to its deposition, terrestrial POC, including the
64 petrogenic part, is subjected to different processes occurring in the catchment. These are
65 related to the connectivity of the landscape compartments, as portrayed by the sediment
66 cascade concept (e.g. Fryirs et al., 2007) and to an unknown number of deposition and
67 resuspension cycles during the lateral transport in rivers, as depicted by the POC spiraling
68 concept (Newbold et al. 1982). All these more and less time-consuming processes promote
69 the mineralization of the riverine POC and increase its mean aging. In summary, the higher
70 the residence time of terrestrial POC in the watershed, the greater the probability of
71 enhancing its mineralization and subsequent CO_2 evasion (Raymond et al., 2013).

72 Generally, river sediments essentially consist of detrital material of various grain sizes,
73 driven by the local hydrodynamic conditions, and include evidence of past aquatic primary
74 production. However, sedimentary OC stored in rivers mainly originates from the catchment
75 since (i) the chemical and physical properties of the terrestrial POC make it more recalcitrant
76 than POC_a to further degradation both in the water column and during early diagenesis (e.g.,
77 Hedges et al., 1997) and (ii) the suspended load rapidly limits aquatic primary production in
78 river systems (e.g., Meybeck, 2006). As a consequence, sedimentary archives can be
79 viewed as a mixture of all these POC sources, with rather a terrestrial (e.g. from the
80 catchment) imprint. The ^{14}C measurements performed on bulk sedimentary OC reflect such
81 mixing and each source is characterized by its own ^{14}C labelling.

82 As a consequence, the question of the nature of organic material isolated for further isotopic
83 measurements of ^{14}C levels is crucial and clearly depends on the scientific question and
84 field. As an example, for paleo-environmental issues, ^{14}C activity is mostly measured from
85 organic particles consisting of wood debris, seeds or charcoals extracted from marine,
86 lacustrine or riverine sediments to date materials (e.g., Dezileau et al., 2014). However in
87 sediment archives, some of these organic particles, such as wood debris, may not be
88 suitable for building an age model and assessing sedimentation rates due to the so-called
89 “old wood effect” which lead to aging processes (cf. inbuilt age, Gavin, 2001). Such a bias
90 can also arise when focusing on refractory organic material, such as charcoals, because of
91 their long residence times in the catchment either in river-banks, soils or sediments located
92 upstream of the studied sedimentary deposit (Oswald et al., 2005). However, for these
93 refractory materials, ^{14}C ages are suitable for deciphering the biomass fire or fossil fuel
94 origins of black carbon (Wang et al., 2016). For understanding soil dating and soil C
95 dynamics, ^{14}C analyses are performed on either bulk soil OM (Rumpel et al, 2002) or their
96 related specific organic compounds, or fractions, which were previously isolated after
97 substantial and time-consuming physical, chemical and/or biological extraction (e.g.
98 Trumbore, 2009). Lastly in coastal and marine environments, ^{14}C analyses coupled with
99 analyses other isotopes ($\delta^{13}\text{C}$) can constitute a powerful method of defining the age and the
100 origin of organic particles in water column or in in sediments (i.e. aquatic, terrestrial, fossil,
101 black carbon, e.g. Bauer et al., 2001). For specific marine OM, such ostracods, and with
102 respect to the age reservoir of marine water, ^{14}C measurements can be viewed as a powerful
103 method of dating marine sedimentary deposits (e.g. Berndt et al., 2019).

104 In this study, without any pretreatments to isolate specific organic compounds, we aim to
105 unravel the ^{14}C contents in sedimentary OC held by decadal fluvial deposits located
106 downstream of the nuclearized Loire River (France), by considering the relative amounts of
107 various POC components. Such refinement is expected to highlight the ^{14}C origins and the

108 relative contributions of the various ^{14}C sources, including potential imprints from the nuclear
109 industry.

110

111 **2. BACKGROUND, METHODS AND ANALYSES**

112 **2.1. Coring site characteristics, sampling and age model**

113 The studied sediment core was previously described in Eyrolle et al. (2019). Briefly, several
114 1 m long sedimentary archives were collected near Montjean-sur-Loire (France) on
115 September 2016 (0 ° 51'23.1 "W - 47 ° 23'34.0" N; Altitude: 13 m) on the right bank of an
116 island in the Loire River, just upstream of the estuary influence and downstream of the last
117 NPP (Figure 1). Several 1m long-cores were sampled within a 1m² area using a percussion
118 corer (Cobra TT - SDEC) with tubes 45 mm in diameter in order to collect enough material.
119 Once collected, the cores were transported to the laboratory where 5 cm slices were
120 sampled along the core and immediately stored at -25°C. Then, the samples were separately
121 freeze-dried under dehydrated nitrogen flux to avoid any atmospheric exchange before
122 analyses. This coring site is one of the most well known in the Loire basin as it is based on a
123 previous and very fine study of a sediment core collected in the same site in 2009 where
124 grain size, mineralogical and geochemical proxies have been detailed (Grosbois et al, 2012).
125 In this previous work, the anthropogenic history of the Loire sediments during the XXth
126 century has been evidenced. The main findings showed a wide range of contamination
127 patterns resulting from the urban development, ore-processing and related activities but also
128 from an important river eutrophication leading to endogenic calcite precipitation and a dilution
129 effect of contaminant contents. For this study, sedimentological parameters (fig. 2A and B)
130 show clayey-silty sediments all along the 1-m core with an uniform particle size distribution
131 (80% of silts) and a D₅₀ ranging from 15 to 28 µm. Only sediment layers at the bottom show
132 some variations with a finer layer at 110 cm deep cm deep and a coarser one below 120 cm
133 cm. All these variations make each core comparable to the one studied previously (Grosbois
134 et al, 2012) and, as any significant geomorphological and sedimentological changes

135 occurred for the past 10 years, this present work benefits of information from the 2 previous
136 studies (Grosbois et al, 2012; Eyrolle et al, 2019).

137 The age model was based on radiocesium and $^{238}\text{Pu}/^{239+240}\text{Pu}$ ratio (Fig. 2C). Radiocesium
138 concentrations with depth show two major peaks at 92.5 cm and 37.5 cm. These peaks are
139 associated to peaking radioactive emissions from atmospheric nuclear tests in 1963 and to
140 fallout from the Chernobyl accident in 1986, respectively, and can be used as chronological
141 signals (fig. 2). The radiocesium peak in 1986 is preceded from 67.5 cm of depth to 37.5 cm
142 by relatively high radiocesium concentrations due to the liquid discharges by the Loire river
143 NPPs gradually implanted since 1957 (UNGG then PWR of Chinon NPP's) then from the
144 beginning of 80's for the WPR. Furthermore, $^{238}\text{Pu}/^{239+240}\text{Pu}$ activity ratios (fig. 2) also show
145 two maximums, one at 77.5 cm depth (0.06 ± 0.01), the other at 52.5 cm (0.08 ± 0.01).
146 Plutonium isotopes initially results from the atmospheric fallout due to nuclear tests
147 performed over 1945-1980 mainly in the northern Hemisphere and from atmospheric
148 deposition related to the explosion of the Transit 5 BN-3 satellite over the Indian Ocean in
149 1964. In the particular case of the Loire River, plutonium activity ratios significantly higher
150 than those characterizing the atmospheric reference values are due to the accidental liquid
151 plutonium discharges in 1969 and 1980 at the Saint-Laurent-Les-eaux NPP's. Thus,
152 $^{238}\text{Pu}/^{239+240}\text{Pu}$ activity ratio peaks constitute, in addition to the radiocesium peaks, additional
153 chronological markers for dating the archive. From these chronological landmarks (1963,
154 1969, 1980, 1986 and 2016), a simplified age model was applied connecting years and
155 depths. All these results for the age model using ^{137}Cs and plutonium isotopes were
156 previously published (Eyrolle et al., 2019) and demonstrated ~~show~~ that the 1 m long archive
157 covers almost the last seven decades (1950–2016) with constant sedimentation rates over
158 two main periods, the 1963–1986 period with 2.4 ± 0.2 cm/y and the 1986–2016 one with 1.3
159 ± 0.2 cm/y as expected from Grosbois et al. (2012).

160

161

162 **2.2. Origins of Particulate Organic Carbon in rivers**

163 As shown in Figure 3, sedimentary OC can be viewed as a mixture of POCs from soil,
164 reworked POC_{rw} from superficial deposits, POC_p from sedimentary rock and POC_a from
165 aquatic primary productivity. The three former components originate from the catchment and
166 are considered as detrital. In particular, POC_{rw} is part of the sedimentary cascade within the
167 catchment (e.g. Fryirs et al., 2007) and is stored in superficial formations such as colluvium,
168 terraces, sediments deposited upstream of the studied station and/or even ancient soils. As a
169 consequence, POC_{rw} consists of a mixture of various types of aged POC. POC_a, POC_s and
170 POC_{rw} correspond to biospheric POC (POC_b) since the age of POC_{rw} can be younger than
171 60 ky (Brugeron et al., 2018). These two latter components are considered a part of
172 terrestrial POC (POC_t). However, an unknown part of POC_{rw} could be devoid of ¹⁴C and
173 could also be assimilated into POC_p. Among all these components, POC_a is the one that is
174 able to reflect liquid radiocarbon releases from the nuclear industry. All these sources exhibit
175 a large range of $\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$ values, with a possible overlap when these two isotopes are
176 combined to decipher the origins of the POC pool and their cycling (fig. 4, modified from
177 Marvick et al., 2015 see also section 2.4). A convenient simplification considers that POC,
178 either in sediments or in suspended particulate matter (POC), can be seen as a binary
179 mixture of dead-C POC (i.e. POC_p and some POC_{rw}) and POC from the biosphere showing
180 a mean ¹⁴C activity (Leithold et al., 2006). This binary mixture concept is frequently used to
181 extract POC_p from POC or sedimentary OC but suggests that the two end-members fulfil
182 three requirements: the mean OC content and the ¹⁴C concentration in POC_b must remain
183 globally constant as the POC_p content (Galy et al., 2008). At first glance, and as seen in the
184 figure 4, the large range of $\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$ values for the different sources able to supply
185 riverine POC implies that the use of this binary mixture remains unreliable. However, if we
186 consider that (i) the intensity of erosion processes in the catchment is variable with space
187 and (ii) the catchment is at the equilibrium state in terms of the climatic, anthropogenic and
188 geological variables ruling erosion, then only some specific sedimentary sources
189 preferentially supply the river (Walling, 1983). Hence, the use of this binary mixture remains
190 a reasonable compromise.

191

192 **2.3. Bulk organic geochemistry and optical analyses of Particulate Organic Matter**

193 The main geochemical characteristics of particulate organic matter were defined with a Rock-
194 Eval 6 pyrolyser (Vinci Technologies), which overcomes OM isolation by digestive processes
195 ((HCl/HF attacks) of the mineral fractions. Basically, this thermal degradation method
196 consists of a pyrolysis and followed by oxidation of the samples (details in Lafargue et al.,
197 1998). The OC content (in wt. %, uncertainty 3 to 5%) is given by the sum of the POC
198 calculated during the pyrolysis (Pyrolysed Carbon, PC) and oxidation (Residual Carbon, RC)
199 steps.

200 Particulate Organic Matter (POM) was also characterized regarding its optical assemblage
201 with quantitative palynofacies signatures (Graz et al., 2010) where each particle class is
202 expressed in mg g^{-1} . After the isolation of the POM from the sample (2-5 g dried at 30°C) by
203 acid digestion (HCl/HF) of the mineral fraction, the POM was then observed under light
204 microscopy (Leica DMR XP) in transmitted light (objective x50) using a grid (10 μm mesh). In
205 this analysis, 200 elementary surfaces were considered in order to obtain an error
206 percentage lower than 5%. The classes of POM are defined on the basis of morphological,
207 textural and colorimetric criteria (Tyson, 1995). The full optical quantitative results were
208 published in Eyrolle et al. (2019). For this study, we focus either on greyish and gelified
209 amorphous organic matter and gelified particles, characterizing the aquatic OM particles
210 (POMa), and the opaque pyrogenic particles defined by some devolatilization vacuoles and
211 their contribution to the total particulate assemblage (POMa and pyro ratio, table 1). In order
212 to convert and keep the weighted contribution of POMa (α term in the equation 5) to the total
213 optical assemblage of POM into the POCa contribution to the POC (section 3.5), and at first
214 approximation, we hypothesize that the weighted contribution of C in OM remains broadly
215 constant whatever the origins of POM.

216

217 **2.4. Carbon 14 analyses**

218 The radiocarbon contents were analyzed using an accelerator mass spectrometer. Sample
219 preparation consisted of washing the sample (0.5 M HCl, 0.1 M NaOH) and then drying it
220 under vacuum to eliminate carbonates. Samples were decarbonated to focus only on the
221 organic counterpart. Decarbonated samples were then sealed in quartz tubes under vacuum
222 with an excess of CuO and silver wire. Tubes were introduced into a furnace at 835°C for 5hr
223 to convert the organic carbon into CO₂. The quartz tubes were then broken under vacuum to
224 release, dry, measure, and collect the CO₂. The graphite target was obtained with a direct
225 catalytic reduction of CO₂ using iron powder as a catalyst (Merck® for analysis reduced, 10
226 µm particles). The reduction reaction occurred at 600°C with excess H₂ (H₂/CO₂ = 2.5) and
227 was complete after 4-5 hr. The iron-carbon powder was pressed into a flat pellet and stored
228 under pure argon in a sealed tube. To reduce contamination from modern carbon or memory
229 effects, all quartz and glass dishes were burned at 450°C for at least 5 hr. A turbo-molecular
230 pump reaching 10⁻⁶ mbar was used to evacuate the vacuum lines. Measurements were
231 performed using the Artemis facility (Moreau et al. 2013) with a 3 MV NEC Pelletron
232 Accelerator coupled with a spectrometer dedicated to radiocarbon dating, measuring ¹²C, ¹³C
233 and ¹⁴C contents and counting the ¹⁴C ions by isobaric discriminations. The analyses
234 required 1 to 100 mg of dry sample (to obtain 1 mg of carbon). Riverine ¹⁴C is commonly
235 expressed as an enrichment factor (Δ¹⁴C in ‰, eq. 1). Δ¹⁴C integrates (i) the year of
236 measurement (y), (ii) the average life of ¹⁴C atom within a sample until it decays (t_{mean} =
237 8267 y), (iii) the year for which the standard (i.e. oxalic acid I; OX-I) is decay-corrected
238 (1950), (iv) the ¹⁴C/¹²C isotopic ratio of the sample (¹⁴R) and (v) the ¹⁴C/¹²C isotopic ratio of
239 the OX-I (¹⁴Rs), readers can refer to Mook and van der Plicht, 1999 and Trumbore et al.,
240 2016 for further details).

241
$$\Delta^{14}\text{C} (\text{‰}) = \left[\frac{{}^{14}\text{R}}{(0.95 \cdot {}^{14}\text{Rs} \cdot \exp^{-(y-1950)/8267})} - 1 \right] \cdot 1000 \text{ (Eq. 1)}$$

242 The average standard deviation of Δ¹⁴C is ± 3‰, and a positive value indicates a ¹⁴C-
243 enrichment from ¹⁴C bomb and/or NPPs releases, a negative value indicates a ¹⁴C dilution
244 from various aged POC while ¹⁴C-avoid compounds age over 60 ky reach -1000‰.

245 (Trumbore et al., 2016). $\delta^{13}\text{C}$ expresses the enrichment or depletion of the ^{13}C stable isotope
246 during physico-chemical or metabolic processes (e.g. photosynthesis) and is calculated as
247 follow (eq. 2):

$$248 \quad \delta^{13}\text{C} (\text{‰}) = [({}^{13}\text{R} / {}^{13}\text{R}_{\text{PDB}}) - 1] \cdot 1000 \text{ (Eq. 2)}$$

249 In this equation 2, ${}^{13}\text{R}$ is the isotopic ratio $^{13}\text{C}/^{12}\text{C}$ of the sample and ${}^{13}\text{R}_{\text{PDB}}$ is that of the
250 international standard (Pee Dee Belemnite, PDB, Smith and Epstein, 1971).

251 Practically, $\delta^{13}\text{C}$ signatures can be used to assess the contributions of various carbon
252 sources transferred by rivers (Aucour et al., 1999). However, $\delta^{13}\text{C}$ signatures of aquatic
253 plants can overlap with those of terrestrial materials (Raymond and Bauer, 2001).
254 Accordingly, the combination of $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ values used in this work (fig. 3 and 5) can
255 help to better determine the C sources from recently fixed C to aged or petrogenic organic C
256 stored in deep soil horizons and sedimentary rocks (Raymond et Bauer, 2001).

257

258

259 **2.5. Petrogenic POC content assessment**

260 Mean POCp values (wt. %) in sedimentary archives can be determined by plotting the
261 sedimentary OC contents and the product of these values with the corresponding modern
262 fraction (Fm) of the sample (Galy et al., 2008). Linear modelling provides the POCp content
263 when the line defined by equation 3 crosses the x-axis. However, three criteria must be met
264 prior to the use of this method (Galy et al., 2008): (i) the sedimentary OC in the samples
265 should be seen as a binary mixture of petrogenic and biospheric POC (POCp, POCb), (ii) the
266 POCp content should be almost constant within the sedimentary archive, and (iii) the mean
267 radiocarbon content of POCb (i.e. Fmb) should also be relatively constant within the
268 sedimentary archive. When these criteria are met, the data plotted in the diagram produce a
269 linear trend.

$$270 \quad \text{OC} \cdot \text{Fm} = \text{OC} \cdot \text{Fmb} - \text{POCp} \cdot \text{Fmb} \text{ (Eq 3)}$$

$$271 \quad \text{when } \text{OC} \cdot \text{Fm} = 0, \text{ then } \text{OC} = \text{POCp}$$

272 where OC (sedimentary OC) and POCp are expressed in wt. %, Fm is the measured
273 radiocarbon composition of the sample (fraction of modern C) and Fmb is the radiocarbon
274 composition of the POCb in the sample.

275

276 **2.6. POCa content assessment and Freshwater Reservoir Effect (FRE)**

277 The POCa content was assessed from the bulk organic geochemistry analyses coupled with
278 the optical study of the quantitative palynofacies, as previously described in Eyrolle et al.
279 (2019). During its growth, aquatic OM incorporates carbon from the dissolved inorganic load
280 in rivers and thus, its ^{14}C content most generally reflects part of the dissolved inorganic ^{14}C
281 contents (DI^{14}C) (Aucour et al., 1999). In turn, the DI^{14}C contents mostly depends on the
282 importance of weathering processes of sedimentary rocks (carbonate and, to a lesser extent,
283 siliceous) in the catchment occurring on the surface and in groundwaters, delivering the
284 inorganic dead-carbon dissolved load of rivers (Deevey et al., 1954). Despite exchanges of
285 gaseous inorganic carbon that tend toward equilibrium between atmospheric and river
286 systems, fossil DIC ages the river DIC and consequently the POCa. Hence, the FRE
287 corresponds to the difference between the age of the freshwater carbon reservoirs and the
288 age of the atmospheric carbon reservoirs (Ascough et al. 2010).

289

290 **3. RESULTS AND DISCUSSION**

291 **3.1 $\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$ signals of the riverine POC**

292 At the studied station, the $\Delta^{14}\text{C}$ values in the sediment core ($\Delta^{14}\text{C}\text{-OC}$) exhibit a general
293 increasing trend from the bottom to the surface and can be divided into 3 main periods (fig. 5,
294 tab. 1). The 1923 to 1959 period is characterized by $\Delta^{14}\text{C}$ values ranging from -629 to -463
295 ‰, revealing a POC source that was either severely processed (pedogenesis, sediment
296 cascade and/or POC spiraling) and/or either diluted by a POCp contribution. Although it may
297 be an imprint of aerial aquatic plants, the $\delta^{13}\text{C}$ signature remains mostly inherited from C3
298 plants and/or type III kerogen since aquatic plants held labile C preferentially mineralized (fig.
299 6). The second period, from 1961 to 1986 exhibits two peaks in 1971 (-2 ‰) and in 1986

300 (236 ‰) with depleted $\Delta^{14}\text{C}$ value in 1978 (-105 ‰). The last period, from 1990 (176‰) to
301 2016 (222 ‰), shows weak $\Delta^{14}\text{C}$ variation. The POC sources for these two latter periods
302 were less subjected to these aging processes (particularly from 1961 to 1980) and broadly
303 exhibit a $\delta^{13}\text{C}$ signature of terrestrial C3 plants with a possible imprint of aerial aquatic plants
304 in samples showing positive $\Delta^{14}\text{C}$ values (fig. 6, tab. 1).

305 On the whole, and with the exception of the last period, and the peak in 1986, these $\Delta^{14}\text{C}$
306 values are systematically lower than those registered in the atmosphere over the same
307 periods of time. There are several explanations for the disequilibrium between the $\Delta^{14}\text{C}$
308 calculated from the OC of the riverine sediments and that from the atmosphere. Two of them
309 relate to the POC originating from the catchment: (i) a dilution effect by POCp and the part of
310 POCrw older than 60 ky, leading to reduced ^{14}C contents in sediments and decreased $\Delta^{14}\text{C}$
311 values and (ii) a gap between the ^{14}C activity in POCs and POCrw and that in the
312 atmosphere.

313 For the latter point and, as seen in the figure 5, these gaps can be positive or negative
314 depending on the age of OM stored in the soil profiles of the catchment or in the various
315 superficial deposits and the occurrence of erosion processes over time. Finally, another
316 explanation addresses the $\Delta^{14}\text{C}$ values specific to POCa stored in the sediments. Indeed, the
317 ^{14}C contents of this organic fraction can be expected to reflect several components
318 combining the ^{14}C released by nuclear industry, the local FRE and the equilibrium with the
319 atmosphere.

320 According to the $\delta^{13}\text{C}$ values, most of the organic matter stored in the sediments originates
321 from the C3 terrestrial plants and/or aerial aquatic plants (fig. 6). The shift of +2 ‰ of the $\delta^{13}\text{C}$
322 values with depth (fig. 6, tab. 1) may be attributed to a wide range of factors. Among them, a
323 possible minor changes in the source of the C3-plants, a change in the ratio POCs/POCa in
324 the sedimentary OC, and some microbial processes as seen for the lacustrine sediments
325 (Brenner et al., 1999) or for soil profiles (Torn et al., 2002). For the last factor, this

326 enrichment in ^{13}C may be related to the enhanced metabolism of the lighter isotope, which
327 may be promoted by microbes (e.g. Torn et al., 2002).

328

329

330 **3.2. POCp content in the sedimentary archive**

331 In figure 7 expressing the linear modelling, the results show two linear trends characterized
332 by a correlation coefficient of over 0.95. The trend referring to the period from 1923 to 1959
333 indicates very low POCp content (0.01 wt. %) and the second referring to the period from
334 1961 to 2016, reveals a POCp amount of 0.59 wt%. Accordingly, the two groups of points
335 evidenced in the previous section (1961-1986 and 1990-2016 periods) cannot be clearly
336 identified and would have shown a very bad linear relation in the figure 7. This difference in
337 POCp content over these two periods of time suggests a major change in sediment sources.
338 One of the serious candidates for explaining this change in the beginning of the 1960s could
339 originate from the land use management trends and the conversion of grassland into
340 agricultural area. Indeed re-parcelling, which was marked by an increase in agricultural field
341 size, provoked the near-eradication of hedges coupled with the digging of ditches. These
342 surface processes, which are rooted in the global increase in demand for food production
343 and yield, enhance drainage, reroute a part of the sediments in various reservoirs, and
344 promote erosion and the transfer of new sedimentary sources within the catchment (Foucher
345 et al, 2014). These changes are also the origin of the eutrophication of the Loire River, as
346 evidenced by the increase in authigenic calcite and phosphorous concentrations in a
347 previously studied twin core (Grosbois et al., 2012). Hence, it is reasonable to hypothesize
348 that the POCs, POCrw and POCp could have had a different origin starting in 1961, but no
349 argument can be further advanced at this point to precisely determine the source of this
350 petrogenic counterpart. At first glance, and based on the POCp content close to 0.60 wt.%,
351 the source could be some OC/clay-rich sedimentary rocks, such as marls or schists, with a
352 higher OC content than the other main sedimentary rocks (Ronov and Yaroshevski, 1976,

353 Copard et al., 2007). Such sedimentary rocks from the Mesozoic Age (Lias) outcrop along
354 the Middle Loire River and are a part of Paris Basin.

355

356 **3.3. $\Delta^{14}\text{C}$ signal of POCb over the last decades**

357 By using the POCp/OC ratio (tab. 1), the $\Delta^{14}\text{C}$ values of POCb, including that of both aquatic
358 (POCa) and terrestrial organic carbon (POCs+POCrw), can be calculated (fig. 5) by using
359 equation (4):

$$360 \quad \Delta^{14}\text{C-POCb} = \Delta^{14}\text{C-OC} / (1 - \text{POCp/OC}) \text{ (Eq. 4)}$$

361 with OC and POCp expressed in wt. %, $\Delta^{14}\text{C-POC}$ as the calculated values of the sediment
362 samples, and $\Delta^{14}\text{C-POCb}$ as the theoretical $\Delta^{14}\text{C}$ values of biospheric POC.

363

364 While the first previously identified period (1923 to 1959) exhibits no change, as expected
365 ($\Delta^{14}\text{C-POCb} = \Delta^{14}\text{C-OC}$), due to the low amounts of POCp, the second period (1961 to 2016)
366 shows theoretical $\Delta^{14}\text{C-POCb}$ values (from 12 to 368 ‰) higher than the $\Delta^{14}\text{C-OC}$ ones
367 (from -193 to 222 ‰). These results overall highlight that POCp may significantly dilute the
368 ^{14}C content measured in riverine sediments and in sedimentary archives.

369

370 **3.4. $\Delta^{14}\text{C-POCa}$ over the last decades**

371 Among all the OC components that compose riverine sedimentary archives, POCa is the
372 single component that can be expected to provide evidence for ^{14}C liquid discharge from the
373 nuclear industry. Over the period 1961-2010, the POCa content, assimilated to the OMa
374 ratio, within the studied sedimentary archive showed a low contribution to the total optical
375 assemblage of POM (8% as a mean value, tab. 1). Low amounts of POCa are generally
376 expected in most riverine sedimentary archives since these compounds, which are enriched
377 in lipids, are preferentially degraded during early processes (Burdige, 2007). These results
378 suggest that riverine sedimentary archives cannot be expected to store or reveal the ^{14}C
379 releases from the nuclear industry performed over years. Nevertheless, to subtract this POC

380 component from the POCb signal, we estimated the $\Delta^{14}\text{C-POCa}$ by using additional data
381 sets acquired from radiological monitoring and survey (SYRACUSE IRSN database).
382 Before the beginning of nuclear energy production by NPPs (PWR) in 1980 (fig. 1, Dampierre
383 NPPs) and assuming the absence of ^{14}C nuclear releases before this period of time, the
384 $\Delta^{14}\text{C-POCa}$ values would express a part of the FRE. Thus, the age given by FRE was
385 required in order to estimate the DI^{14}C content of the Loire River upstream NPPs. Even
386 though FRE depends on hydrological conditions and tributary inputs, involving a large range
387 of FRE values, we used a mean FRE value of 1194 ± 30 ^{14}C yr (Coularis et al., 2016), i.e.
388 approximately -143 ‰. This mean FRE value characterizes the upper part of the Loire basin
389 and overcomes the potential influence of NPPs. To assert whether the nuclear releases were
390 recorded in aquatic plants, we used the radiocarbon data from some aquatic mosses ($n=21$)
391 collected upstream and downstream of the Belleville-sur-Loire NPPs as these mosses do not
392 exhibit aerial surfaces that promote $^{14}\text{C}_{\text{atm}}$ fixation during photosynthesis. The Mean $\Delta^{14}\text{C}$
393 values of aquatic mosses collected in 1999 and 2000 upstream and downstream of the
394 Belleville-sur-Loire NPPs reached -164, and -176 ‰ respectively, while the $\Delta^{14}\text{C}_{\text{atm}}$ ranged
395 from 121 to 115 ‰. With respect to the uncertainties associated with FRE, these data show
396 that aquatic mosses in the Loire River are generally in equilibrium with the FRE. Importantly,
397 these data show that the measured mosses do not record the ^{14}C liquid releases from
398 nuclear activities. However, this statement should be considered with caution due to (i) the
399 deficiency of the representativeness of the samples for radiocarbon measurement purposes
400 and (ii) the ^{14}C plume from the nuclear releases, which depends on the local hydrodynamic
401 conditions, remains unknown. For the latter reason, we can hypothesize that if the sampling
402 of aquatic plants are outside the plume, there is obviously no ^{14}C imprint from the NPPs.
403 Finally, a mean value of -10 ‰ C for $\Delta^{14}\text{C-POCa}$ was extracted from the IRSN radiocarbon
404 data and corresponds to the ^{14}C activity measured for the mosses and also for aerial aquatic
405 plants such as *Carex* ($n=13$) and *Phalaris* ($n=8$) growing along the riverbank.

406

407 **3.5. Reconstruction of $\Delta^{14}\text{C}$ -POCt over the last decades**

408 By using the POMa contribution (α the contribution to the sedimentary OC, see section 2.3) to the total optical assemblage of POM
409 acquired over the period 1961-2010 with the quantitative palynofacies method (Eyrolle et al.,
410 2019), the $\Delta^{14}\text{C}$ -POCt can be obtained by using the equation (5):

412
$$\Delta^{14}\text{C-POCt} = \Delta^{14}\text{C-POCb} - (\alpha \cdot \Delta^{14}\text{C-POCa}) / (1 - \alpha) \quad (\text{Eq. 5})$$

413 As expected, the $\Delta^{14}\text{C}$ -POCt values are generally lower than that of POCb (fig. 5, tab. 1).
414 These results finally underline that the ^{14}C contents characterizing the terrestrial POC is far
415 from the atmospheric referential value over the whole period covered by the sedimentary
416 archive. As POCt consists of a mixture of POCs and recent POCrw (stored in the catchment
417 as superficial formations), the gaps can be explained since (i) it is well known that POCs
418 shows a large range of ^{14}C contents with depth (e.g. Schmidt et al., 2011, Trumbore, 2009)
419 and (ii) superficial formations, such as old riverine deposits (terraces) or colluviums, can also
420 hold a wide range of OM younger than 60 kyr (e.g. Brown et al., 2009). In other words, the
421 $\Delta^{14}\text{C}$ -POCt values routed by rivers clearly depend on the portion of soil depth and that of the
422 recent sediment, which are both being mobilized by erosion processes. As previously
423 described, the profile can be divided into three main distinct periods:

424 From 1923 to 1959, since there is neither petrogenic (POCp = 0.01 wt.%) nor aquatic
425 contributions to the ^{14}C content of sedimentary OM, the $\Delta^{14}\text{C}$ -POCt signal fit well with that of
426 the $\Delta^{14}\text{C}$ calculated for the sediment. However, the substantial gap between the $\Delta^{14}\text{C}$ -POCt
427 and $\Delta^{14}\text{C}_{\text{atm}}$ values corresponds to a mean age of 6247 ± 847 ^{14}C years for POCt deposited
428 in the sedimentary sink, which is far older than that given for the bulk sediment with the age
429 model (Grosbois et al. 2012; Eyrolle et al., 2019). This involves that erosion processes
430 occurring in the catchment have mobilized essentially highly aged POCt over this period of
431 time. Such POC could be held either by Holocene detrital riverine deposits in the basin,
432 and/or deep soil horizons showing a low turnover of soil organic carbon (Schmidt et al.,
433 2011).

434 From 1959 to 1986 and as seen in the figure 5, at the beginning of the 1960s, a major shift
435 toward higher values occurs for $\Delta^{14}\text{C-POCt}$ (from -463 to -39 ‰). This shift was also
436 enhanced by the occurrence of petrogenic OM (fig. 7, $\text{POCp} = 0.59 \text{ wt.}\%$). In 1969 ($\Delta^{14}\text{C-}$
437 $\text{POCt} = 385 \text{ ‰}$), another shift decreased the $\Delta^{14}\text{C}$ until 1978 ($\Delta^{14}\text{C-POCt} = 136 \text{ ‰}$) and then
438 it increased until 1986 ($\Delta^{14}\text{C-POCt} = 508 \text{ ‰}$). The 1969 shift is due to the beginning of
439 atmospheric fallout from nuclear tests; however a time lag of 5 to 7 years was observed in
440 the sedimentary archive. This time lag is interpreted as the mean transit time from the source
441 of sediments in the catchment to the sedimentary sink in the river system. The subsequent
442 decrease in 1969 is concomitant with the occurrence of carbonized organic particles, likely
443 depleted in ^{14}C content, with vacuole devolatilization as charcoals (fig. 8). This suggests
444 either a significant fire event or an industrial incident along the Loire corridor. These particles
445 supply the sedimentary compartment up to 1975. Afterward, the steady increase in $\Delta^{14}\text{C-}$
446 POCt until 1986 may reflect the residence time of the system, almost 20 years, for such
447 particles.

448 From 1986 to 2010, the $\Delta^{14}\text{C-POCt}$ values steadily decreased from 508 to 139 ‰ while the
449 $\Delta^{14}\text{Catm}$ values in the atmosphere decreased from 197 to 22 ‰ for the same period of time.
450 The mean gap between $\Delta^{14}\text{C-POCt}$ and $\Delta^{14}\text{Catm}$ reaches $212 \pm 65 \text{ ‰}$ over this period of time.
451 This is consistent with the works of Levin and Heishaimer (2000) indicating that the terrestrial
452 biospheric reservoir is a source of ^{14}C for the atmosphere at least since the end of 1980s.

453

454 **3.6. Transit time of POCt from the catchment to the river sediments**

455 Based on our dataset, the transit time of the OM route in the catchment can be assessed by
456 comparing the $\Delta^{14}\text{C}$ peak level in the atmosphere in 1963 with the $\Delta^{14}\text{C-POCt}$ peak in the
457 sedimentary archives. Indeed, the terrestrial OM has previously fixed atmospheric ^{14}C during
458 its growth on the catchment following photosynthesis (e.g. Schurr et al., 2016) prior its route
459 toward the deposition area and its burial in river sediments. As a consequence, this POCt
460 has only kept in memory the atmospheric ^{14}C during its growth. Since there is no additional
461 ^{14}C incorporation in OM after its death, a mean transit time can be estimated by comparing

462 the time lag between the well know $\Delta^{14}\text{C}$ peak level in the atmosphere in 1963 with that
463 preserved within the sedimentary OC (fig. 5). This latter peak may have occurred over the
464 period 1970-1985. The fire event occurring in 1969 prevents any accurate determination of
465 its date. Nonetheless, a minimum and maximum transit time for POCt of 7 and 22 years,
466 referring to the time lags from 1963 to 1970 and 1985, respectively, can be estimated for the
467 Loire basin. Such time lags are in agreement with our previous estimation of 5-7 years made
468 at the beginning 1960s (see above) as those provided by using organically bound tritium
469 (Eyrolle et al., 2019).

470

471 **4. CONCLUSIONS AND PERSPECTIVES**

472 We aim to decipher the $\Delta^{14}\text{C}$ -OC values in sediments from one of the most nuclearized rivers
473 in Europe. For a similar period and as expected, the ^{14}C levels in these materials are far
474 different from those in the atmosphere. There are several reasons for this discrepancy. The
475 first is the dilution of the radiocarbon contents by dead-carbon inherited from the erosion of
476 outcropping sedimentary rocks and aged superficial formations in the catchment. The
477 proportion of the dead-carbon can be easily determined with a linear modelling. Due to the
478 combination of previously published optical measurements, a mean FRE value and a
479 radiocarbon dataset for aquatic organic matter sampled along the river corridor, the
480 contribution of aquatic POC to the $\Delta^{14}\text{C}$ -OC was also calculated. The $\Delta^{14}\text{C}$ -POCa reflects the
481 FRE and not the ^{14}C liquid discharge from the nuclear industry. Hence, both the terrestrial
482 (i.e. soils and younger superficial formations) contribution to the $\Delta^{14}\text{C}$ of the riverine and
483 sedimentary OC and the age of these terrestrial OM could be easily assessed. Despite our
484 inability to split the respective contribution of the soil and the superficial formations to the ^{14}C
485 content of terrestrial organic matter, the $\Delta^{14}\text{C}$ signature of riverine OC could be refined into
486 three parts. In this respect, such a method of $\Delta^{14}\text{C}$ reconstruction would also be a powerful
487 tool for defining the transit time of sediments in a river. For example, in the Loire River, the
488 transit time could be between 7 and 22 years, however future studies are required to validate
489 this tool. This radiocarbon unravelling could also point to the milestones of the social and

490 economic trajectories for a considered catchment and their consequence for sediment and C
491 dynamics in rivers. Here, some major changes in land use, caused by re-parcelling, were
492 identified during the 1960s; these changes, in turn, triggered a reroute of the sediments in
493 the river bringing in some new sedimentary organic matter. The age of terrestrial OM before
494 and after the 1960s clearly indicates the rejuvenation of POCt stored in sediments,
495 suggesting a major change in sedimentary sources in the catchment. In addition, coupled
496 with the palynofacies method of characterizing the particulate organic assemblage, ^{14}C
497 unravelling is suitable for identifying events that occurred in the catchment, such as an
498 industrial accident or a fire event. Here, these events impacted both the radiocarbon
499 concentration of terrestrial POC and the optical assemblage in the river sediments during the
500 end of the 1960s. The incursion of these specific particles depleted in radiocarbon was
501 observed up to 1986, suggesting that the resilience of the riverine system to this event is
502 almost 20 years. Last, the gap between $\Delta^{14}\text{C}_{\text{atm}}$ and that of the terrestrial POC clearly shows
503 that the terrestrial biospheric reservoir is a source term of ^{14}C for the atmosphere since at
504 least 1986.

505

506

507 **Acknowledgments**

508 This work was funded by the Institute for Radioprotection and Nuclear Safety (IRSN-NEEDS,
509 project PALYNO) and a part of the TRAJECTOIRE project from the French National
510 Research Agency (ANR-19-CE3-0009, 2020-2024). Warm thanks are addressed to J.P
511 Bakyono, F. Giner, D. Mourier and C. Le Corre for their precious help during field sampling,
512 sample preparation and sample analyses. The authors also sincerely thank the three
513 anonymous reviewers and the editor for their appreciated help in the improvement the
514 manuscript.

515

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698

699 **Figures caption**

700 Figure 1: *study site showing the coring site and the spatial distribution of nuclear power*
701 *plants along the Loire River (France), and its main contributors (from Eyrolle et al., 2019).*
702 *Nuclear technologies are indicated as UNGG (natural uranium, graphite and gaz) and PWR*
703 *(Pressurized water reactors) with their respective period of use.*

704
705 Figure 2: (A): *core sedimentary log showing the main lithology, (B): particle size distribution*
706 *and mean diameter D50 (μm), (C): activities of radiocesium (Bq kg^{-1}) and Pu ratio ($\times 1000$)*
707 *signals leading to the age model. The two peaks of ^{137}Cs in 1963 (due to peak emissions*
708 *from nuclear weapons tests in the atmosphere), then in 1986 (due to atmospheric fallout*
709 *from the Chernobyl accident in Europe), and the two peaks of $^{238}\text{Pu}/^{239+240}\text{Pu}$ activity ratio in*
710 *1969 and 1980 (occurring after the Saint-Laurent-des-Eaux nuclear reactor accidents) were*
711 *used as chronological references (from Eyrolle et al., 2019). Uncertainties on years are*
712 *estimated to be ± 2 years. For A, B and C, the bottom core was fixed at 125 cm in order to*
713 *homogenise the beginning of these signals.*

714
715 Figure 3: *conceptual view of various particulate organic carbon (POC) sources in a*
716 *catchment (from the Bleone river system, upstream Digne les Bains, Alpes de Haute*
717 *Provence, France). POCa: aquatic POC as the autochthonous POC for the river system.*
718 *POCp (petrogenic POC), POCs (soil POC), POCrw (reworked POC), both considered as*
719 *allochthonous and respectively originating from: outcropping rocks, soils, superficial*
720 *formations. the two color codes for POCrw correspond to POC older (grey) and younger*
721 *(sepia) than 60 ky BP. Red circle correspond to a theoretical coring site containing*
722 *sedimentary OC. POCa, POCs and POCrw correspond to the biospheric POC (POCb) while*
723 *POCt includes POCs and POCrw. See also section 2.2 for details. (picture from Goolge*
724 *earth).*

725

726 Figure 4: $\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$ signatures of the main sources of riverine POC (adapted from
727 Marwick et al., 2015). Atmospheric $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$ from atmospheric CO_2 (Graven et al.,
728 2017), C3 plants: $\delta^{13}\text{C}$ (Kohn et al., 2010, O'Leary, 1988), – C4 plants $\delta^{13}\text{C}$ (Cerling et al.,
729 1997), $\Delta^{14}\text{C}$ for C3 and C4 plants are in equilibrium with the atmosphere (e.g. Trumbore,
730 2009) - Aquatic plant: $\delta^{13}\text{C}$ for submerged and aerial plants (Chappuis et al., 2017), $\Delta^{14}\text{C}$
731 (SYRACUSE IRSN database) – $\delta^{13}\text{C}$ of kerogen (Tyson, 1995) (fossil OM, Tyson, 1995) is
732 divided into three main type of organic matter: the type III encompasses all the terrestrial
733 higher plants origin while the type I and II combine the lacustrine/bacterial and marine
734 origins. Upward arrow corresponds to a possible contamination in ^{14}C either by ^{14}C gas or
735 liquid releases by nuclear activities. A combination of C3-C4 plants can occurs in soils
736 (mixing), pedogenesis in soils strictly involves POCs, sedimentary cascade in the catchment
737 involves all the terrestrial POC (POCrw POCs) including POCp while POC spiralling in
738 streams afflicts all the POC sources.

739

740 Figure 5: $\Delta^{14}\text{C}$ signals of the atmosphere reservoir and measured $\Delta^{14}\text{C}$ at the most
741 downstream fluvial station of the Loire river basin of sedimentary OC (black circle); green
742 diamond: rebuilt $\Delta^{14}\text{C}$ of biospheric POC (POCb), red diamond: rebuilt $\Delta^{14}\text{C}$ of terrestrial
743 POC (POCt=POCs+POCrw) from the catchment.

744

745 Figure 6: $\Delta^{14}\text{C}$ and $\delta^{13}\text{C}$ signatures of sedimentary OC stored in the Loire River. Samples are
746 ranged in the aged and reworked or C3 / aerial aquatic plants POC signatures. While
747 samples in black circles (belong to period 1923-1959) appear depleted in ^{14}C , these samples
748 do not hold dead-C but rather show an aged POCs or POCrw signature. To the opposite,
749 those in red circles (belong to the period 1961-2016) show a significant POCp contribution
750 despite their high $\Delta^{14}\text{C}$ values, suggesting a significant enrichment in ^{14}C due to the
751 atmospheric source. See also the legend of the figure 4 for further detailed explanations.

752

753 Figure 7: *linear modelling (from Galy et al., 2008) applied to the sediment samples. Red*
754 *diamonds correspond to the period 1961-2016 (POCp = 0.59 wt.%) while the red one to the*
755 *1923-1959 period (POCp = 0.01 wt.%).*

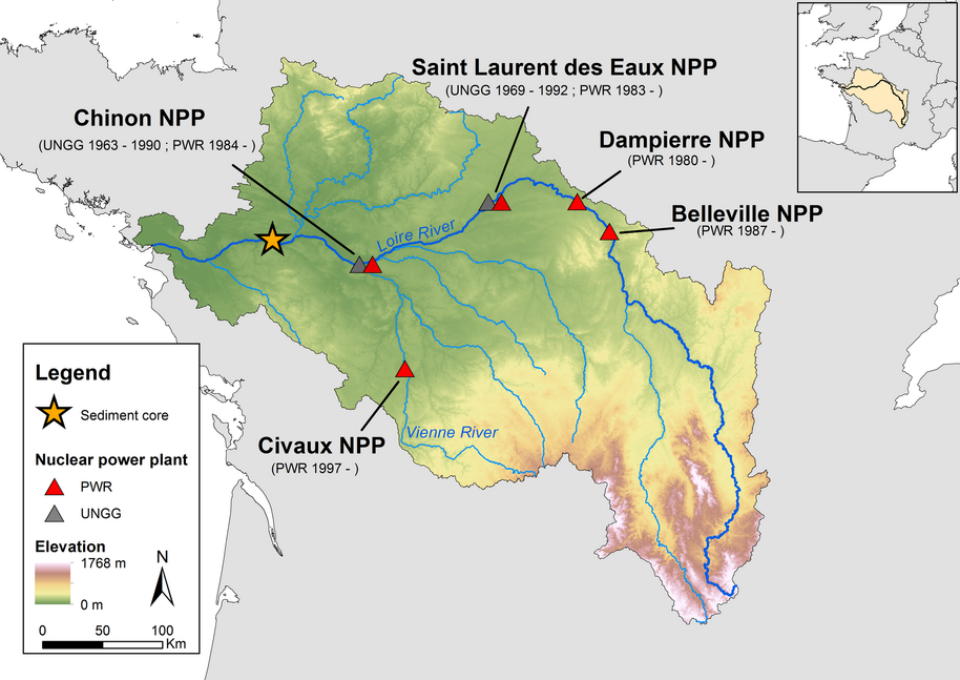
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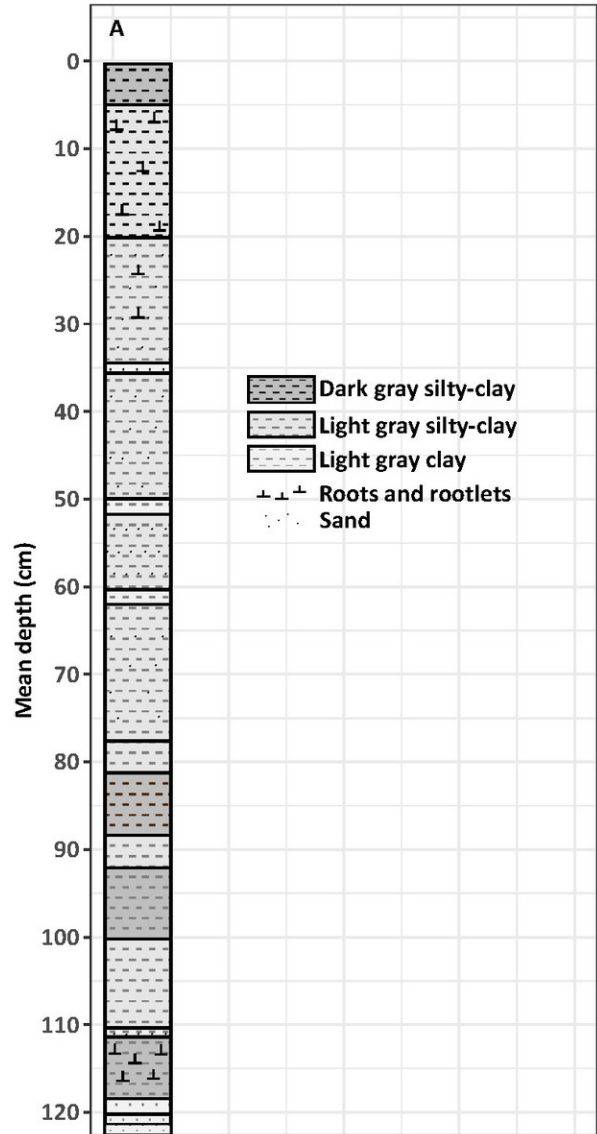
757 Figure 8: *sedimentary pyrogenic ratio signal with time. The pyrogenic particle identified with*
758 *the palynofacies method consists in opaque and corroded particle showing devolatilization*
759 *vacuoles (Graz et al., 2010). If the background level of these particles is less than 1% of the*
760 *total organic particles throughout the sedimentary core, a short period of time (1969-1976) is*
761 *characterised by higher ratio suggesting a fire (natural or industrial) event with a resident*
762 *time of 7 years.*

763

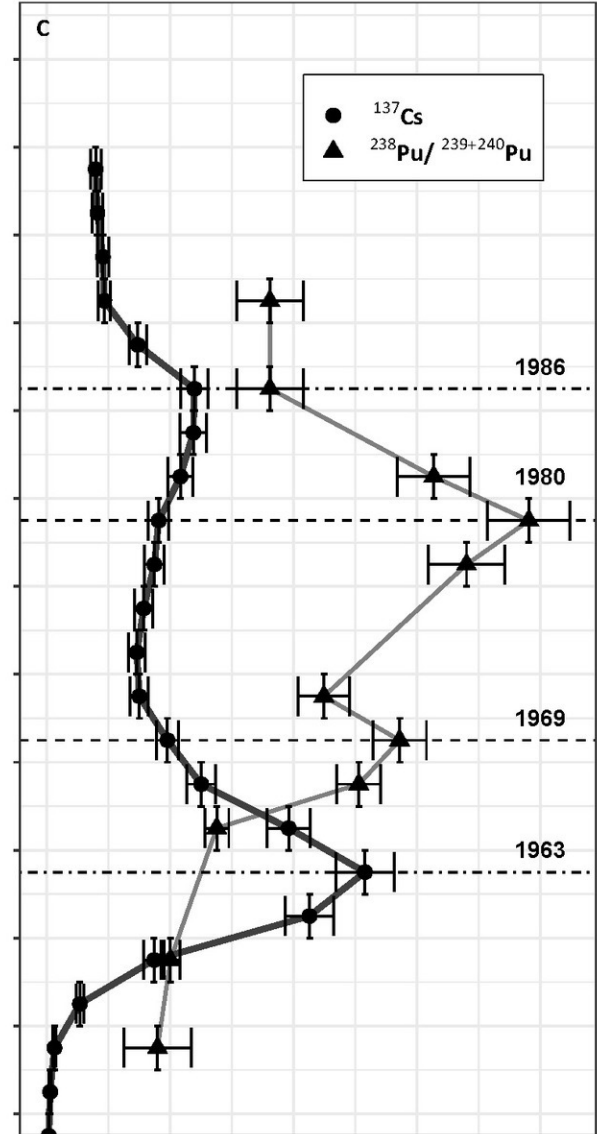
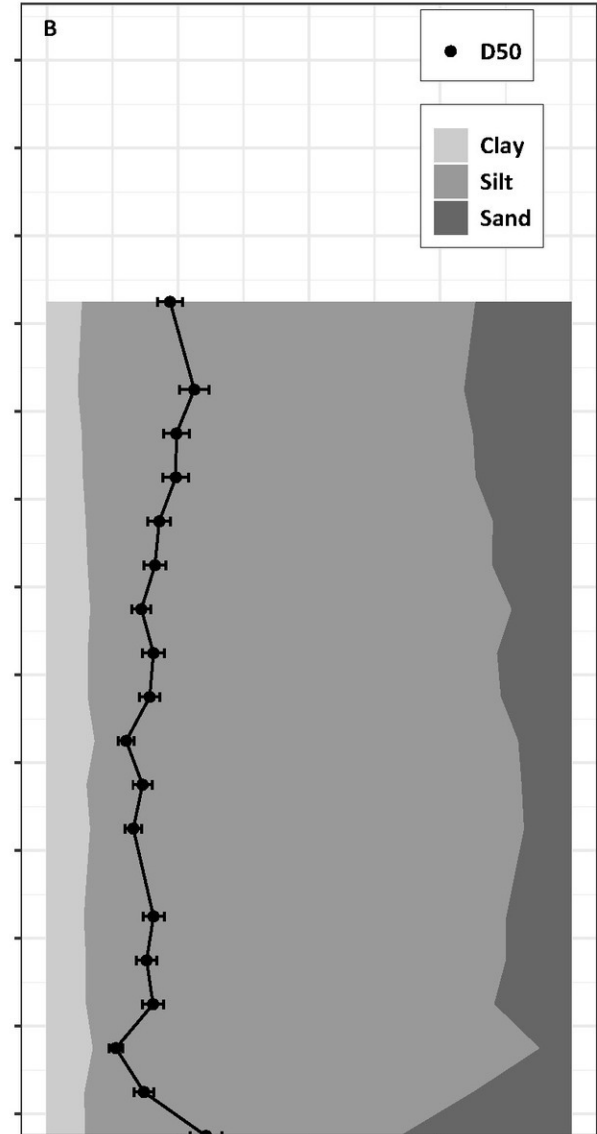
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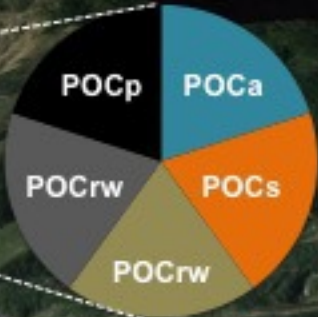
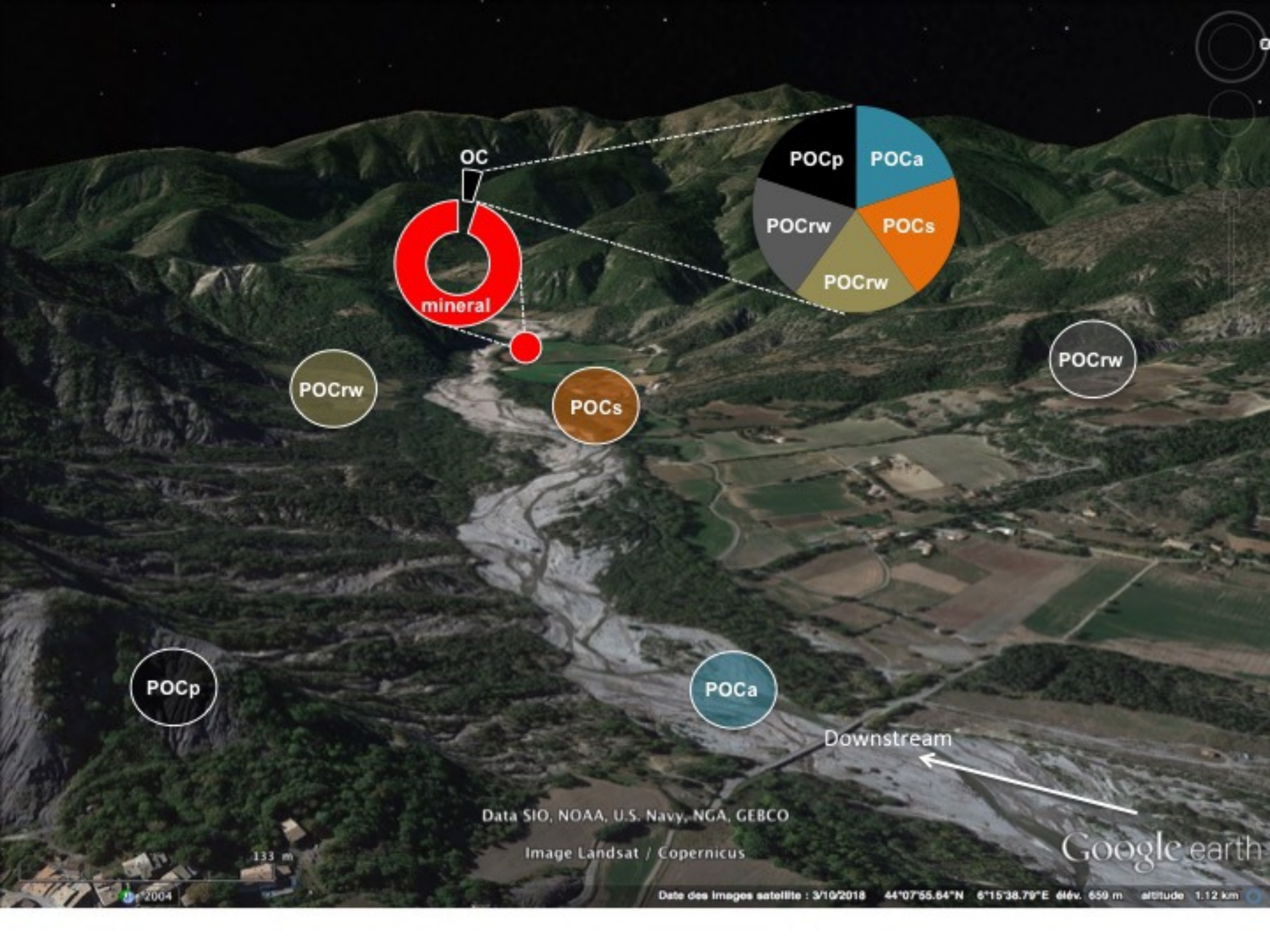




Core sedimentary log



Activity of ^{137}Cs (Bq kg^{-1}) and $^{238}\text{Pu}/^{239+240}\text{Pu}$ activity ratio ($\times 1000$)



POC_{rw}

POCs

POC_{rw}

POCp

POCa

Downstream

Data SIO, NOAA, U.S. Navy, NGA, GEBCO

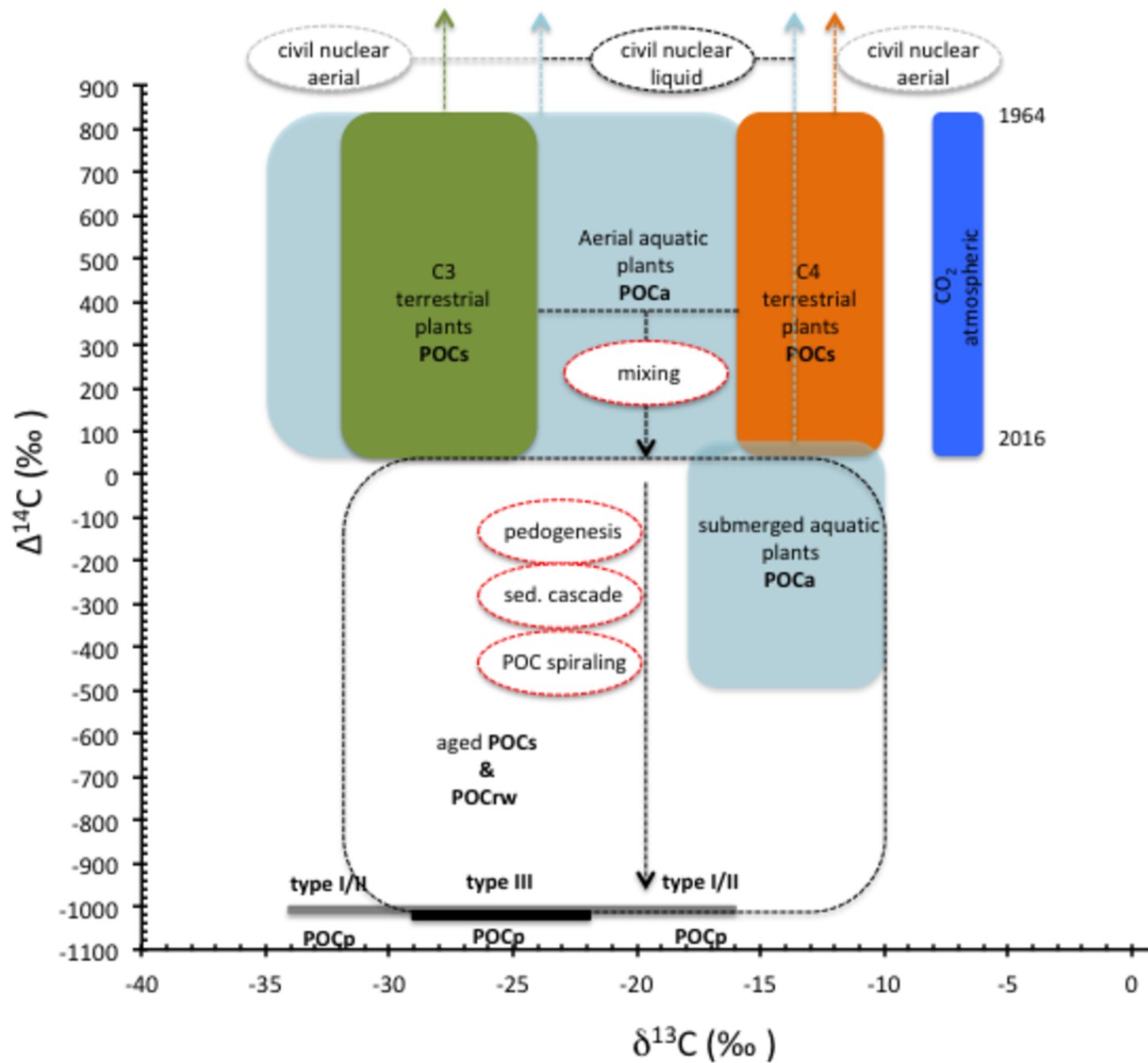
Image Landsat / Copernicus

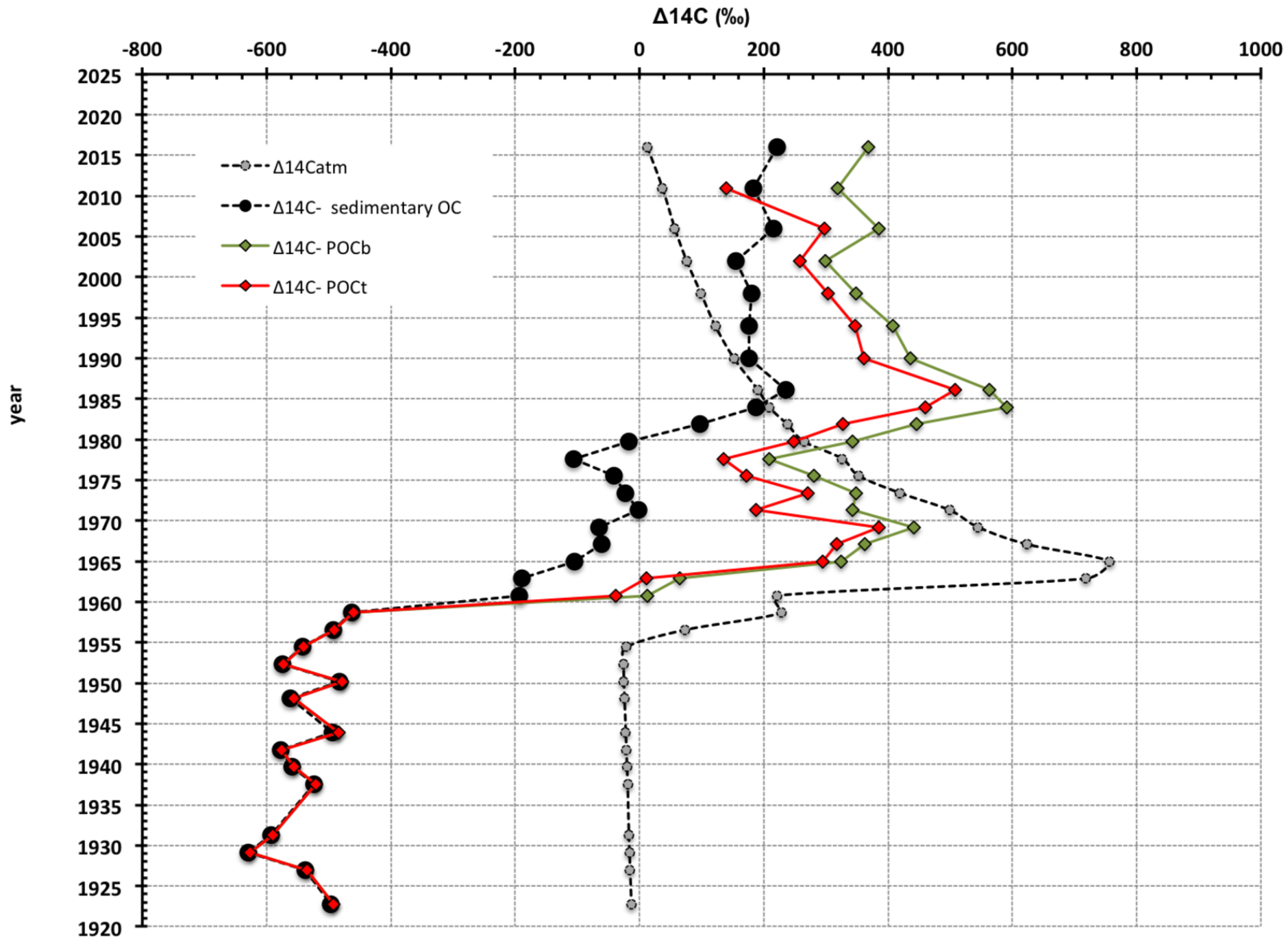
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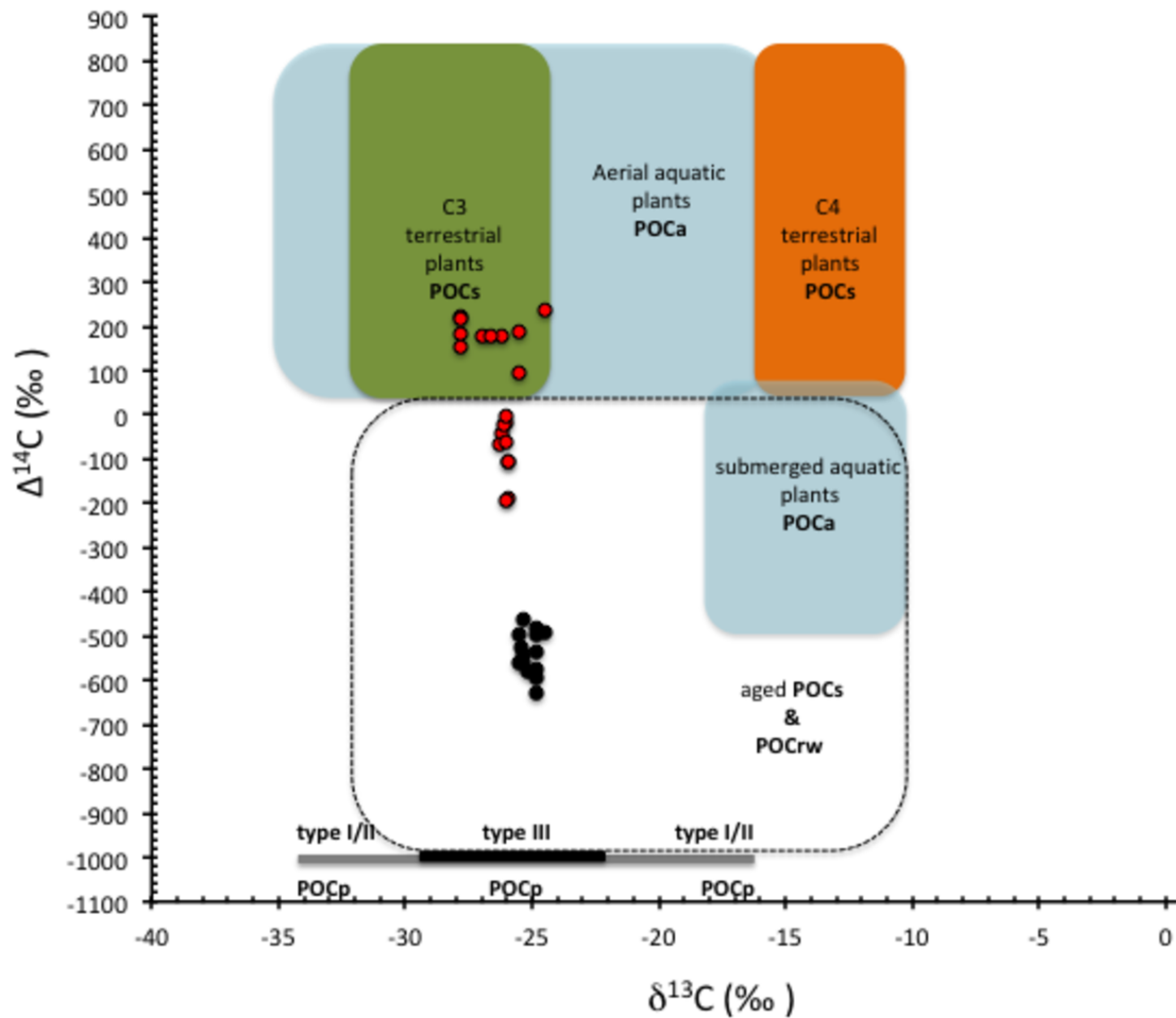
133 m

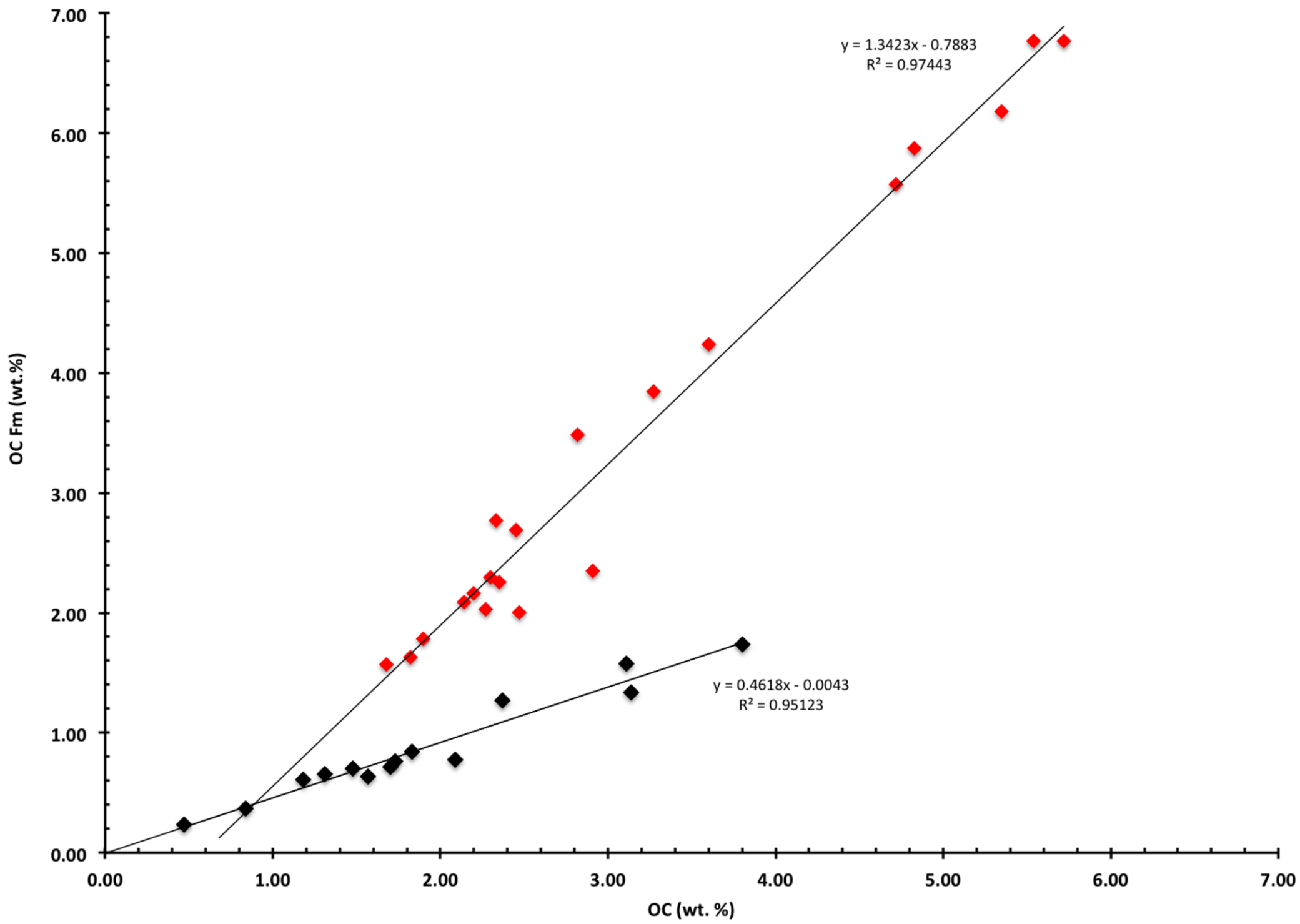
2004

Date des Images satellite : 3/10/2018 44°07'55.64"N 6°15'38.79"E élév. 650 m altitude 1.12 km









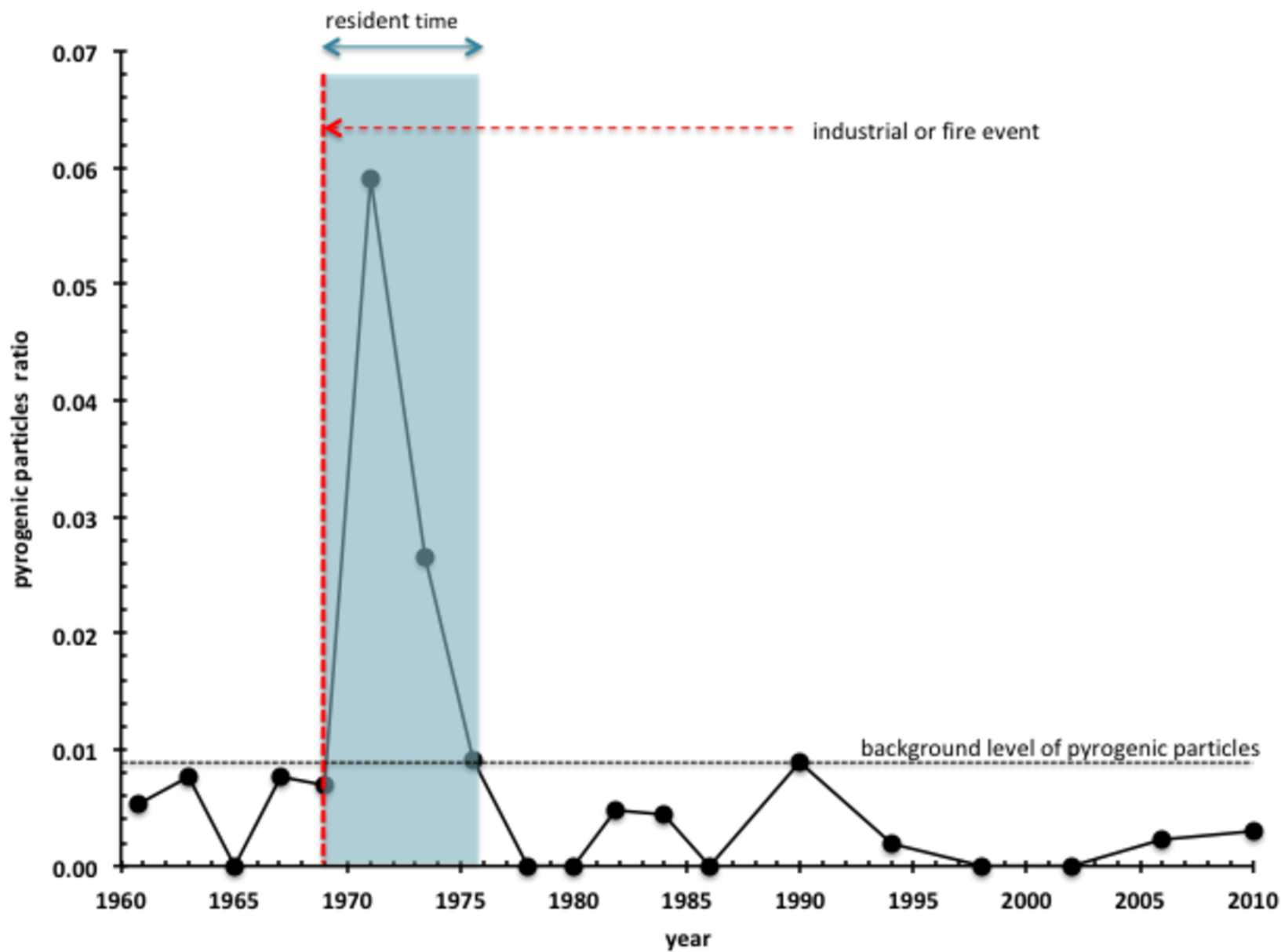
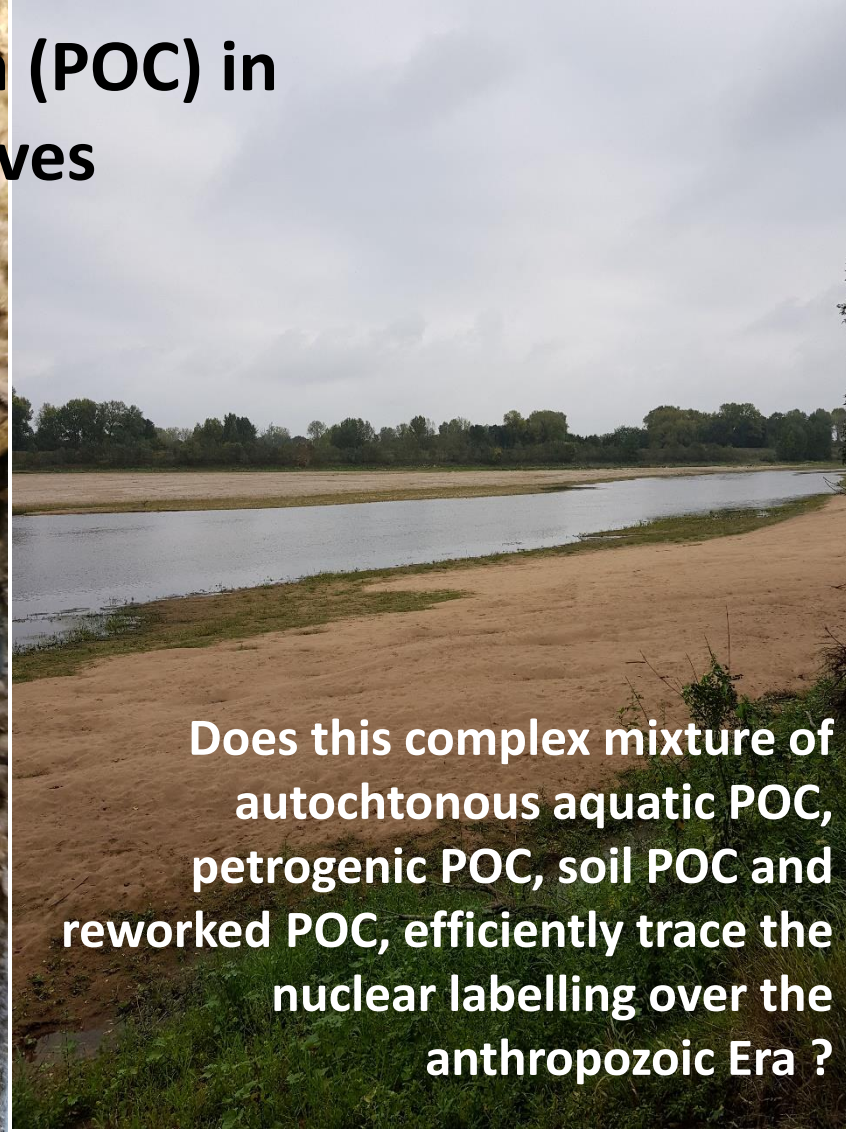


table 1: values of the main isotopic parameters of the riverine samples and that of the atmosphere ($\Delta^{14}\text{C}_{\text{atm}}$, Graven et al. 2017). Ages of the samples are calculated with an uncertainty of +/- 2 years from some isotopic methods ($^{238}\text{Pu}/^{239+240}\text{Pu}$ and ^{137}Cs , see Eyrolle et al., 2019). POMA and Pyro are two ratios corresponding respectively to the aquatic and the carbonized particles contributions to the total optical assemblage of OM given by the palynofacies method (Graz et al., 2010, see Eyrolle et al., 2019 for details). Fm corresponds to the fraction of modern as described in Mook and Van der Plitch (1999), POCp content is defined in the section 3.2 and in the equation 1. Little available sediment for the period 1923-1959 and 2016 explains the absence of optical results.

depth (cm)	age	$\Delta^{14}\text{C}_{\text{atm}}$ (‰)	$\Delta^{14}\text{C}$ -POC (‰)	$\delta^{13}\text{C}$ (‰)	Fm	POC (wt.%)	POCp/POC	$\Delta^{14}\text{C}$ -POCb (‰)	$\Delta^{14}\text{C}$ -POCt (‰)	PO Ma	Pyro
2.5	2016	13	222	-27.8	1.22	5.54	0.106	368	N/A	N/A	N/A
7.5	2011	37	183	-27.8	1.18	5.72	0.103	319	139	0.15	0.003
12.5	2006	57	216	-27.8	1.22	4.83	0.122	385	298	0.08	0.002
17.5	2002	76	156	-27.8	1.16	5.35	0.110	299	258	0.04	0.000
22.5	1998	99	180	-27	1.18	4.72	0.125	349	304	0.04	0.000
27.5	1994	122	177	-26.2	1.18	3.60	0.164	408	347	0.06	0.002
32.5	1990	152	176	-26.6	1.18	3.27	0.180	436	362	0.07	0.009
37.5	1986	191	236	-24.5	1.24	2.82	0.209	563	508	0.05	0.000
42.5	1984	209	188	-25.5	1.19	2.33	0.253	591	460	0.12	0.004
47.5	1982	238	97	-25.5	1.10	2.45	0.241	446	327	0.11	0.005
52.5	1980	265	-17	-26	0.98	2.20	0.268	343	248	0.09	0.000
57.5	1978	326	-105	-25.9	0.89	2.27	0.260	209	136	0.07	0.000
62.5	1976	353	-41	-26.2	0.96	2.35	0.251	281	172	0.10	0.009
67.5	1973	419	-23	-26.1	0.98	2.14	0.276	349	271	0.07	0.026
72.5	1971	499	-2	-26	1.00	2.30	0.257	342	187	0.13	0.059
77.5	1969	545	-64	-26.3	0.94	1.68	0.351	442	385	0.05	0.007
82.5	1967	624	-61	-26	0.94	1.90	0.311	363	318	0.04	0.008
87.5	1965	756	-105	-25.9	0.90	1.82	0.324	325	295	0.03	0.000
92.5	1963	718	-190	-25.9	0.81	2.47	0.239	64	12	0.05	0.008
97.5	1961	221	-193	-26	0.81	2.91	0.203	12	-39	0.05	0.005
102.5	1959	228	-463	-25.3	0.54	2.37	0.004	-461	-461	N/A	N/A
107.5	1957	73	-493	-24.5	0.51	3.11	0.003	-491	-491	N/A	N/A
112.5	1954	-21	-543	-25.3	0.46	3.80	0.003	-541	-541	N/A	N/A
117.5	1952	-25	-575	-24.8	0.43	3.14	0.003	-573	-573	N/A	N/A
122.5	1950	-25	-482	-24.8	0.52	1.18	0.008	-478	-478	N/A	N/A
127.5	1948	-24	-562	-25.3	0.44	0.84	0.012	-556	-556	N/A	N/A
137.5	1944	-22	-495	-25.5	0.50	0.47	0.021	-484	-484	N/A	N/A
142.5	1942	-21	-578	-25.2	0.42	1.70	0.006	-576	-576	N/A	N/A
147.5	1940	-20	-559	-25.5	0.44	1.73	0.006	-556	-556	N/A	N/A
152.5	1938	-19	-523	-25.4	0.48	1.48	0.007	-520	-520	N/A	N/A
167.5	1931	-17	-592	-24.8	0.41	1.57	0.006	-590	-590	N/A	N/A
172.5	1929	-16	-629	-24.8	0.37	2.09	0.005	-627	-627	N/A	N/A

177.5	1927	-15	-537	-24.8	0.46	1.83	0.005	-535	-535	N/A	N/A
187.5	1923	-13	-497	-24.8	0.50	1.31	0.008	-493	-493	N/A	N/A

Particulate Organic Carbon (POC) in riverine sedimentary archives



**Does this complex mixture of
autochthonous aquatic POC,
petrogenic POC, soil POC and
reworked POC, efficiently trace the
nuclear labelling over the
anthropozoic Era ?**