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Abstract: Excessive inputs of sediment and acceleration of primary production have been observed worldwide in a large number of water bodies. Human-environment interactions were recognized as one of the main drivers of this evolution during the 20th century with the occurrence of major landscape changes and a greater use of agricultural inputs. In this study, we used paleo-production proxies such as chlorophyll-a, organic matter properties (TOC and TN concentrations, $\delta^{13}C$ and $\delta^{15}N$) measured in sediment cores dated with fallout $^{210}Pb_{ex}$ and ^{137}Cs activities for reconstructing changes in accumulation rates and sources of organic matter during the recent period of agricultural intensification (1920-2020). In order to record these changes at the regional scale, sediment cores were collected at the outlet of several headwater catchments (n=9), covering a wide range of land covers / land uses across the Loire River basin (117,000 km²), France. The rates of sedimentary organic matter deposition in the studied water bodies accelerated from 1950 onwards (+48%). Between 1950 and 1970, the signature of sedimentary organic matter indicates a dominant contribution of soil-derived inputs. This period corresponds to major landscape modifications across the basin (land consolidation, stream re-design, implementation of tile drains) driving a general acceleration of erosion rates. Then, from 1960 onwards, chlorophyll-a and C/N proxies indicate an increase in primary production coupled with a decrease of terrigenous supply in agricultural catchments. These proxies were strongly correlated to the agricultural inputs during the 1955-1990 period (e.g., $r=0.9$ between chlorophyll-a content and N inputs), suggesting a progressive eutrophication of these reservoirs driven by increasing fertilizer use. During these 35 years, sedimentary organic matter deposition rates

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37 Abstract

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41 one of the main drivers of this evolution during the 20th century with the occurrence of major
42 landscape changes and a greater use of agricultural inputs.

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50 The rates of sedimentary organic matter deposition in the studied water bodies accelerated
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56 agricultural catchments. These proxies were strongly correlated to the agricultural inputs during the
57 1955-1990 period (e.g., $r=0.9$ between chlorophyll-a content and N inputs), suggesting a progressive
58 eutrophication of these reservoirs driven by increasing fertilizer use. During these 35 years,
59 sedimentary organic matter deposition rates increased on average by 30%. During the 1990s, despite a
60 slight reduction in fertilizer use (-13%), the paleo-production proxies (e.g. chlorophyll-a) still indicate
61 a positive trend suggesting the contribution of another driving factor such as climate warming or again
62 nutrient release from soils and sediments.

63 In the absence of long term geochemical and sediment input monitoring, paleolimnological
64 reconstructions provide a powerful tool to reconstruct past agricultural pressures in rural

65 environments. This study illustrates the impact of intensive farming on water body siltation driven by
66 varying sources of organic material during the 20th century. In addition, these results suggest that
67 eutrophication processes of these reservoirs with contrasting land uses started during the 1960-1970
68 period and are still ongoing nowadays.

69

70 Keywords: terrigenous organic matter, autochthonous production, agricultural inputs, agriculture
71 intensification, soil erosion, Anthropocene

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89 I. Introduction

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91 Intensification of agricultural practices during the second half of the 20th century induced a
92 significant acceleration of soil erosion and sediment delivery worldwide (Dearing & Jones, 2003;
93 Cerdan *et al.*, 2010). This period of agriculture development is associated with a greater use of
94 agricultural inputs including industrially produced fertilizers. Global fertilizer consumption was
95 multiplied by 4.5 between 1960 and 2015 according to the FAO database (Tilman *et al.*, 2001; Lu &
96 Tian, 2017). Both landscape management and the increase use of agricultural inputs have generated
97 deleterious effects on terrestrial and aquatic ecosystems (e.g. Boardman & Poesen, 2006; Rowan *et al.*,
98 2012). Among these adverse consequences, the excessive delivery of sediment may contribute to the
99 siltation of water bodies including dams, ponds, lakes or river systems (e.g. Gellis *et al.*, 2006;
100 Downing *et al.*, 2008; Ahn *et al.*, 2010). Two main sources of material can contribute to this sediment
101 accumulation: sediment originating from terrestrial sources (e.g. soil erosion, river channel erosion)
102 including mineral and organic material supply or organic matter associated with excessive primary
103 production induced by eutrophication (e.g. Meyers & Ishiwatari, 1995; Gudas *et al.*, 2017). Causes
104 and mechanisms of water body eutrophication were widely studied during the 1970s and 1980s in a
105 large number of lakes in the Northern hemisphere (Glooschenko *et al.*, 1974; Hecky & Kilham, 1988).
106 This research clearly outlined the key role of phosphorous (P) and nitrogen (N) point-based and
107 diffuse source pollution in the bloom of algae in aquatic environments (Bennett *et al.*, 2001). Despite
108 substantial management efforts implemented since this period to regulate agricultural N and P inputs
109 and to improve wastewater treatment (EEC, 1991a; b), the excess of phytoplanktonic production
110 remains problematic in coastal and continental waters (Le Moal *et al.*, 2019). According to the 2018
111 European water assessment status, at least 38% of the continental water bodies have agricultural
112 diffuse source pollution in Europe (EEA, 2018).

113 In the absence of long term geochemical and algae production monitoring in river systems
114 (Minaudo *et al.*, 2015; Chen *et al.*, 2016), trajectories of eutrophication and paleoproduction are less

115 documented over the last century. Paleolimnological techniques based on the collection of sediment
116 cores can provide a useful tool to reconstruct primary productivity over the last century (Brenner *et al.*,
117 1999; Anderson *et al.*, 2014). To this end, a large number of methods was developed in order to
118 reconstruct changes in paleo-productivity in sediment cores (e.g. Meyers, 1994; Lu *et al.*, 2010),
119 including the analysis of pigment contents in lake sediments (Waters *et al.*, 2005; Florian *et al.*, 2015).
120 Chlorophyll-a and its degradation products are the most abundant pigments in lake sediments (Leavitt
121 & Hodgson, 2001). They were widely analyzed for reconstructing histories of eutrophication, and
122 assessing the respective impacts of human activities and climate change (Korosi & Smol, 2012;
123 Stewart *et al.*, 2015). Chlorophyll-a concentration can be quantified through a wide range of chromatic
124 techniques (Millie *et al.*, 1993; Hodgson *et al.*, 1997). In recent years, the potential of the rapid and
125 non-destructive Visual Reflectance Spectroscopy has been demonstrated to detect chlorophyll-a and
126 the associated pheopigments in sedimentary sequences (Wolfe *et al.*, 2006). In this way, a variety of
127 indices were used to infer chlorophyll-a and its degradation products abundances including its relative
128 absorbance depth at 660/670 nm (Rein & Sirocko, 2002), sediment reflectance near 675 nm (Wolfe *et*
129 *al.*, 2006), the absorbance peak between 650 and 700 nm (Michelutti *et al.*, 2010) or again the Q7/4
130 ratio developed by Debret *et al.* (2011).

131 Stable C and N isotope signatures of sediment organic matter ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) provide one of the most
132 widely used methods to identify changes in sources of organic material. This methodology was
133 applied to palaeoenvironmental reconstructions based on the analysis of lake sediments (e.g. Meyers,
134 1994; Huon *et al.*, 2018). The carbon isotopic composition of plants is variable and depends on the
135 source of carbon assimilated. Terrestrial plant record $\delta^{13}\text{C}$ values range from -13 to -30‰ (C3 plants
136 are associated with values of -23 to -30‰ whereas C4 plants correspond to values between -9 and -
137 17‰ with an average value of -13‰) - (Meyers, 1994; Bowsher *et al.*, 2008). Freshwater aquatic
138 plants and plankton have $\delta^{13}\text{C}$ values ranging between -25 and -30‰. Both terrestrial and aquatic
139 organic matter may be distinguished through the comparison of their C/N ratio. Macrophyte residues
140 have C/N ratios >20 but mineralization and aggregation in soils reduces C/N to *ca.* 10 (Meyers, 1994).
141 In addition, $\delta^{15}\text{N}$ can provide information on biogeochemical processes including N consumption by

142 organisms, varying sources of N, denitrification – nitrification and diagenetic processes (e.g. Jinglu *et*
143 *al.*, 2007; Gälman *et al.*, 2009).

144 Although these proxies were widely used in sediment cores collected in estuarine, coastal
145 areas or even in urban environments to investigate the relationship between nutrient inputs and a
146 change in the water body trophic status (e.g. Voß & Struck, 1997; Rabalais *et al.*, 2007), many fewer
147 studies were conducted in small reservoirs located in continental environments. In addition, to the best
148 of our knowledge, the study of freshwater reservoirs located in contrasted agricultural environments at
149 the large basin scale (<100,000 km²) has never been conducted especially considering the second part
150 of the 20th century.

151 The current research investigates the potential of sedimentary sequences collected at the outlet
152 of contrasted rural catchments (with land cover dominated by forest, grassland or arable land) to
153 reconstruct regional paleoproductivity trends. The objectives of this work are to determine the rates of
154 organic matter deposition in sediments and to identify the sources of organic matter accumulated
155 during the 20th century in these reservoirs, through the discrimination of the terrigenous (soil-derived)
156 *vs.* autochthonous (biological) inputs. Finally, these results will be compared to available agricultural
157 N input data and to current knowledge on soil erosion in these catchments.

158

159

160 II. Site and methods

161

162 1. Study sites

163 The Loire river basin (117,000 km², France) – (Fig. 1) is characterized by contrasted climate
164 (annual rainfall varying between 600 and 1300 mm per year); landform (mean altitude = 300 m, 10%
165 of the catchment >800 m); lithology (sedimentary; metamorphic and magmatic rocks) and land use
166 conditions between its upper and lower parts (Fig.1). Characteristics of the studied sub-catchments
167 were summarized in Table 1.

168 This large basin was impacted by major changes in agricultural practices during the second
169 part of the 20th century. Some areas experienced an intensification of agricultural practices (mainly in

170 the middle and lower catchment sections) - (Foucher *et al.*, 2014) whereas other parts were impacted
171 by land abandonment following rural depopulation (mainly in upper parts of the catchment) -
172 (Foucher *et al.*, 2019b) – Table 2. After 1950, the spreading of “NPK” fertilizers has significantly
173 increased in France in particular in the Loire River basin (Poisvert *et al.*, 2017). In response to these
174 changes, the Loire river basin – as most European basins – has been impacted by eutrophication
175 processes. The first monitoring surveys focusing on primary production have demonstrated the
176 pronounced eutrophic conditions of the Loire river in the early 1980s (Meybeck *et al.*, 1988;
177 Etcheber *et al.*, 2007). A recent study showed that the continuous decline then stabilization of
178 phosphate and nitrate concentrations in the river since 1990 led to the significant reduction of
179 phytoplankton biomass across the whole river system (Minaudo *et al.*, 2015). Nevertheless,
180 eutrophication still represents a crucial environmental problem in this basin nowadays. According to
181 a recent report of the regional water agency, eutrophication remains one of the main causes of
182 degradation of the ecological status of water bodies (lakes, ponds, dams, etc.) with 29% of those in
183 medium ecological status, 18% in low ecological status and even 26% in bad ecological status
184 (AELB, 2013).

185 Nine reservoirs (lakes and ponds, with a surface ranging between 1.2 and 57 ha) located at the
186 outlet of rural catchments (<100km²) were selected in various environments across the Loire river
187 basin to reconstruct trends in rates of sedimentary organic material deposition and to identify the
188 sources of sediment (Fig. 1). These reservoirs were selected among the 14,000 natural and artificial
189 water bodies of the Loire basin because their catchment histories are well known. Furthermore, to the
190 best of our knowledge, these reservoirs were never dredged or drained during prolonged periods.
191 These water bodies are mainly artificial (n=8 artificial and one natural (Tazenat lake)) and they are
192 connected by ephemeral river networks to their drainage area during heavy rainfall. These sites were
193 selected in order to be representative of the diversity of landscape, climate, topography and
194 agricultural practices found across the Loire basin. Although they are unevenly distributed in space
195 across the basin, they cover all the characteristics of the agricultural landscapes found in this region.
196 Among these catchments, four are mainly covered with grassland, three with arable land and two with
197 forests.

198 The characteristics of these catchments are summarized in Table 2.

199

200 2. Materials and methods

201

202 Fieldwork was conducted between January 2017 and June 2018. Sediment cores (n=9) were
203 collected in the nine water bodies using an Uwitec gravity corer (Uwitec®, Mondsee, Austria)
204 equipped with a 90 mm PVC liner and a floating platform available at the Tours University (GeHCo
205 laboratory, France). In each artificial reservoir, the sediment core was collected in the dam vicinity
206 (\approx 50 to 100m from the dam) in the zone where the sedimentation rates are the highest. In the natural
207 reservoir of Tazenat, the sediment core was retrieved in the center of the water body. Given the
208 relatively small size of these reservoirs (<52 ha), it is assumed that the sediment properties recorded at
209 these sampling sites are representative of those found across the entire water bodies with the exception
210 of variations for the sedimentation rates.

211 Sampling locations and bathymetric data were recorded using a Garmin® Echomaps depth sounder
212 (Garmin, Lenexa, USA).

213

214 2.1. Chronology

215 Sediment dating of the nine cores (selected to be representative of the entire water bodies)
216 were established using Caesium-137 (^{137}Cs) and excess Lead-210 ($^{210}\text{Pb}_{\text{ex}}$) activities measured in 130
217 samples of dried sediment material (*ca* 10g). These samples were regularly collected along the
218 sediment cores with on average one sample taken every 6 cm. Gamma spectrometry measurements
219 were obtained using coaxial N- and P- type HPGe detectors (Canberra/Ortec®) available at the
220 Laboratoire des Sciences du Climat et de l'Environnement (Gif-sur-Yvette, France). The ^{210}Pb
221 activities were determined at 46.5 keV. $^{210}\text{Pb}_{\text{ex}}$ was calculated by subtracting the supported activity
222 from the total ^{210}Pb activity (measured at 46.5 keV) using two Radium-226 daughters, Lead-214
223 (average count at 295.2 and 351.9 keV) and Bismuth-214 (609.3 keV). All measurements were

224 corrected for background level determined every two months as well as for detector and geometry
225 efficiencies. Activities were also decay-corrected to the sampling date (Evrard *et al.*, 2016).

226 Age of the sediment sequence layers was determined using the Constant Rate of Supply model
227 (CRS), (Appleby & Oldfield, 1978). This model assumes a constant rate of $^{210}\text{Pb}_{\text{ex}}$ supply from
228 atmospheric fallout, although sediment accumulation is allowed to vary throughout time. For
229 improving the $^{210}\text{Pb}_{\text{ex}}$ age model of some cores, the corrected CRS model described in Appleby (2001)
230 was used. The $^{210}\text{Pb}_{\text{ex}}$ age model validation was carried out through the identification of ^{137}Cs peaks in
231 the sedimentary sequences. This artificial radionuclide may originate from two sources in Western
232 Europe: the Chernobyl accident (1986) and the thermonuclear weapons testing (maximal emissions in
233 1963). Fukushima fallout was shown to be negligible in France (Evrard *et al.*, 2012). To distinguish
234 between both potential ^{137}Cs sources, Americium-241 (^{241}Am), a daughter product of plutonium-241,
235 was used to identify the ^{137}Cs peak attributed to the maximum nuclear bomb fallout of 1963 (e.g
236 Cambay *et al.*, 1989). Information related to sediment core dating was summarized in Table 3.

237

238 2.2. Laboratory analyses

239 Measurements of Visible Reflectance Spectroscopy (VRS) were obtained using a portable
240 spectrophotometer (Konica® Minolta 700-D) equipped with a 3 mm target radius. Spectral reflectance
241 was measured between 400 and 700 nm, with a 10 nm resolution. Before each set of measurements,
242 the device was calibrated using a white and black calibration box. Measurements were continuously
243 recorded on the fresh sediment surface with a 1-cm resolution, in each core.

244 The absorbance peak between 650 and 700 nm was extracted for a sequence of samples
245 selected in the sedimentary sequences to cover 5-year periods on average as a spectral index to
246 measure the summed concentration of all compounds associated with both primary and degraded
247 chlorophyll-a (chlorophyll-a + all chlorophyll-a isomers + pheophytin-a + pheophorbide-a) -
248 (Michelutti & Smol, 2016). The area under the absorbance peak from 650 to 700 nm was calculated
249 following the procedure described in Das *et al* (2005). These values provide a relative information on
250 chlorophyll-a occurrence index derived from the true area calculation. In order to reduce the noise of

251 the colorimetric time series, a LOESS smoothing model was applied using the free software PAST 3
252 (Hammer *et al.*, 2001).

253

254 The occurrence of carbonate minerals was determined for 95 samples selected on the 9 master
255 cores (on average one sample every 5 years) with a Bernard calcimeter (University of Tours, France)
256 following the method described in NF ISO 10693 (1995). When detected, carbonates were
257 systematically removed from the sample aliquots before performing analyses to determine the total
258 organic carbon concentration (TOC) and stable C isotopes ratios of organic matter and clay-bound
259 organic matter. To reduce possible leaching effects on sedimentary organic matter during carbonate
260 removal, sediments were carefully treated with a 1N HCL solution during 96 hours under constant
261 agitation and pH control in order not to drop below 2.5. Powder X-ray diffraction controls were
262 conducted to ensure the full dissolution of all carbonate minerals in the sediment residue (calcite,
263 dolomite, ankerite, ...). All the samples were rinsed six times in deionized water to remove dissolved
264 salts. The solid residues were recovered by high velocity centrifugation (2500 rounds.min⁻¹) for 15
265 minutes each time (Huon *et al.*, 2002).

266 On these decarbonized sample aliquots, EA-IRMS analyses were conducted on dry sediment
267 for elemental concentration (TOC and total nitrogen concentration TN) and stable C and N isotopes
268 ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$) measurements. These measurements were performed with a continuous flow Elementar®
269 VarioPyro cube elemental analyzer (EA) coupled to a Micromass® Isoprime Isotope Ratios mass
270 Spectrometer (IRMS) at the Institute of Ecology and Environmental Science of Paris. Oxygen for
271 combustion was injected during 70 s (30 mL min⁻¹) and temperatures were set at 850 °C and 1120 °C
272 for the reduction and combustion furnaces, respectively (Agnihotri *et al.*, 2014). Analytical precision
273 and repeatability were controlled with tyrosine samples calibrated against international standards
274 (Coplen *et al.*, 1983). In the current research, the mean uncertainties were 0.2 ‰ for TN, 0.18 ‰ for
275 TOC, 0.1 ‰ for $\delta^{13}\text{C}$ and 0.2 ‰ for $\delta^{15}\text{N}$.

276

277 *2.3. Mass accumulation rate of organic material*

278 Organic matter Mass Accumulation Rates (MAR_{org}, expressed in gC cm⁻² yr⁻¹) were
279 estimated for each individual core using the Sediment Accumulation Rate (SAR, cm yr⁻¹, estimated
280 with the ²¹⁰Pb_{ex} decay), the Dry Bulk Density (DBD, expressed in g cm⁻³ and calculated by measuring
281 the amount of dry sediment in a known volume) and the total organic carbon concentration (TOC,
282 expressed in gC g⁻¹) following eq. 1. MAR_{org} corresponds to the amount of organic material deposited
283 at each individual coring site. This sediment may originate from terrestrial inputs (allochthonous
284 inputs) or may be directly produced in the water body (autochthonous production).

285

$$286 \quad \text{MAR}_{\text{org}} [\text{g cm}^{-2} \text{ yr}^{-1}] = \text{DBD} [\text{g cm}^{-3}] \times \text{SAR} [\text{cm yr}^{-1}] \times \text{TOC} [\text{gC g}^{-1}] \text{ (eq. 1)}$$

287

288 *2.4. Agricultural inputs*

289 Data from the CASSIS_N model (www.geosciences.univ-tours.fr/cassis) - (Poisvert *et al.*,
290 2017) was extracted for each individual catchment and for the entire Loire river basin. This model was
291 used for estimating the mineral and organic nitrogen (N) consumption expressed in kg.ha⁻¹. This
292 model based on data from agricultural census is available between 1955 and 2015 with an annual
293 resolution.

294

295 *2.5. Statistical analyses*

296 The Mann-Kendall non-parametric test (MK-test) was used for detecting monotonic trends in
297 temporal series (Warren & Gilbert, 1988). In this study, the temporal series correspond to the
298 evolution of production proxies ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$, TN, TOC, chlorophyll-a), MAR_{org}, MAR_{min} as well as
299 agricultural inputs throughout time. This test was applied to confirm the occurrence of monotonic
300 upward or downward trends of a given variable throughout time (P-value < 0.05). Trends can be
301 positive, negative or non-null.

302 The non-parametric Buishand test (BU-test) was used for detecting the occurrence of changes in
303 temporal series (Buishand, 1982). Buishand test with a P-value < 0.05 indicated a heterogeneous
304 temporal trend between two periods.

305 In addition, the correlation coefficient (denoted r) was used to quantify the direction (positive,
306 negative) and strength of the linear association between two variables.

307

308 3. Results

309

310 3.1. Chronology

311 According to the dating models, the deepest sediment layers in the cores were deposited before
312 1925 (n=8) except for a single core where it is assumed to have deposited after 1950 (Table 3). The
313 age controls provided by the detection of artificial radionuclides (^{137}Cs and ^{241}Am) were identified in
314 four reservoirs for the Chernobyl fallout and in eight sequences for the 1963 fallout (Fig 4, 5 and 6).
315 Errors between continuous $^{210}\text{Pb}_{\text{ex}}$ age models and those obtained with the radiocesium were in
316 average of 2 years for 1986 and 3 years for 1963. Chronology of two cores, respectively those of the
317 Prugnolas pond and Malaguet lake (Fig. 1) was detailed in Foucher *et al.* (2019b; a). Chronology of
318 the nine cores is detailed in Table 3.

319

320 3.2. Trends in paleo-production proxies

321 Organic material production has significantly changed in seven of the nine study sites with a
322 statistical increase of TOC concentrations in five of the reservoirs. For the two catchments dominated
323 by forests, TOC contents varied between 94 and 180 gC kg^{-1} (average value of $130 \pm 24 \text{ gC kg}^{-1}$) –
324 (Fig. 2). A non-significant trend in TOC content was recorded at both forested sites ($P > 0.05$ MK-
325 Test) – (Table 4). TOC content in catchments mainly occupied by grassland ranged between 18 and
326 160 gC kg^{-1} (average value of $50 \pm 29 \text{ gC kg}^{-1}$) – (Fig. 3). Content in organic material increased by

327 0.04, 1.6, 2.6 and 7.8 % after 1950 (for Loroux, Goule, Beaurepaire and Tazenat reservoirs,
328 respectively). The Loroux pond did not record any statistical change in TOC content. Nevertheless, a
329 peak of organic material was identified in 1965 (TOC = 75 gC kg⁻¹) – (Fig. 3). For the Malaguet lake,
330 this peak was identified in 1956 (TOC = 159 gC kg⁻¹) – (Fig. 2). During the same period, Tazenat lake
331 recorded an acceleration of organic material accumulation, with TOC contents increasing from 2.6 to
332 7.6 % (between 1950 and 1958). TOC concentrations measured in the three study sites dominated by
333 arable land ranged between 28 and 157 gC kg⁻¹ (average value of 65 ± 28 gC kg⁻¹) – (Fig. 4).
334 Concentration in organic material decreased by 0.1 and 1.1% (for Brosse and Passavant ponds,
335 respectively) although it increased by 1.3 % for the Boisvinet pond during the last century. Brosse and
336 Passavant ponds recorded an acceleration of organic material accumulation in the middle of the 20th
337 century (respectively between 1952 and 1960 (TOC = 157 gC kg⁻¹) and between 1955 and 1966 (TOC
338 = 95 gC kg⁻¹) – (Fig. 4).

339 During the 20th century, δ¹³C results showed a significant positive trend in six of the studied
340 reservoirs with a switch around 1973 ± 11 yr in grassland environments, in 1969 in catchments
341 dominated by forest and in 1984 ± 1 yr in areas mainly occupied by arable land (Table 4). For the
342 three remaining water bodies (Brosse, Boisvinet and Prugnolas pond), no statistical trend was
343 observed (Table 4). In forested catchments, δ¹³C ranged between -27.4 ‰ and -29 ‰ (average value of
344 -28 ± 0.4 ‰) – (Fig. 2). For the Malaguet lake, the δ¹³C decreased by 0.3 ‰ during the covered period
345 while it decreased by 0.6 ‰ during the last century in the Prugnolas pond. For the pond located at the
346 outlet of catchments dominated by grassland, the δ¹³C signature of organic matter ranged between -
347 26.7 ‰ and -30.5 ‰ (average value of -28.6 ± 0.95 ‰) – (Fig. 3). For these four catchments, δ¹³C
348 values decreased by 0.5, 0.6, 1.7 ‰ and increased by 0.54 ‰ during the 20th century (for the
349 Beaurepaire, Goule, Tazenat and Loroux ponds, respectively). The δ¹³C of organic matter measured in
350 sediment cores collected in reservoirs draining catchments under arable land ranged between -26.7 ‰
351 and -32.3 ‰ (average value of -29.3 ± 1.4 ‰) – (Fig. 4). Values prior to 1950 were in average of -28.6
352 ± 1.7 ‰ compared to -29.5 ± 1.1 ‰ afterwards. During this period, δ¹³C values decreased by 1.6 ‰
353 and 0.32 ‰ (for Passavant and Boisvinet ponds, respectively) and increased by 0.2 ‰ in Brosse Pond.

354 The $\delta^{15}\text{N}$ signature of organic matter displayed a significant positive trend in three reservoirs
355 (Beaurepaire, Passavant and Boisvinet) and a negative trend in two other water bodies (Loroux and
356 Prugnolas ponds) – (Table 4). Regarding the observed positive trend, the change in trajectory was
357 recorded to occur around 1977 ± 11 yr while it started in 1976 ± 26 yr for negative trends. For the four
358 remaining sites located under various land covers, no statistical trend was recorded (Table 4). For the
359 forested catchments, $\delta^{15}\text{N}$ decreased by 0.2 ‰ and 1.3 ‰ (for Malaguet and Prugnolas reservoirs,
360 respectively) during the documented periods (Fig. 2). Under grassland environments, these values
361 ranged between 4.1 ‰ and 8.8 ‰ (average $7 \pm 1.4\%$). $\delta^{15}\text{N}$ increased by 0.3‰ in the Beaurepaire
362 pond and decreased by 0.4, 0.6 and 2.2 ‰ during the last century (for Goule, Loroux and Tazenat
363 study sites, respectively) – (Fig. 3). In catchments dominated by arable land, $\delta^{15}\text{N}$ ranged between 4.4
364 ‰ and 8.1 ‰ (± 5.7 ‰). When comparing values recorded prior to and after 1950, $\delta^{15}\text{N}$ increased by
365 0.5, 1 and 1.1 ‰ (for Brosse, Boisvinet and Passavant ponds, respectively) – (Fig. 4).

366 C/N ratios showed a significant negative trend in only two ponds (Loroux and Goule ponds)
367 with a switch in this trend occurring around 1969 ± 6 yr. For these two reservoirs, C/N ratio increased
368 from 7.3 to 9.5 for the Goule pond and from 8.6 ± 0.2 to 8.9 ± 0.6 for the Loroux pond, in 1950 and
369 2010, respectively (Fig. 3)

370 Statistical positive trends in chlorophyll-a were recorded in seven reservoirs. Only the
371 Prugnolas pond and the Tazenat lake did not show any statistical trend (Table 4). In the forested
372 catchment, this increase reached around 160 ± 205 % (Fig. 2). In catchments dominated by grassland,
373 chlorophyll-a values increased by 136 ± 87 % after 1950 (Fig. 3). During this period, an increase of
374 190 ± 160 % was recorded in arable land environments (Fig. 4). Changes in these trends were recorded
375 around 1996 ± 5 yr 1963 ± 7 yr and 1990 – for catchments dominated by arable land, grassland, and
376 forest, respectively.

377

378 *3.3. Mass accumulation rate of organic material*

379 Accumulation rate of sedimentary organic material (originating from soil erosion and
380 autochthonous production) has significantly changed during the last century. In the two catchments

381 occupied by forest, negative trends were recorded (decrease of $160 \pm 100\%$ – Table 4). In these
382 catchments, the deposition rates started to decrease around 1963 ± 23 yr (Fig. 5). A significant positive
383 trend in organic production was recorded in the four catchments occupied by grassland (MK test P
384 < 0.05) – (Table 4). In this context, organic matter deposition rate increased on average by 58 ± 23 %
385 between 1950 and 2015 (Fig. 5). In catchments dominated by arable land, a positive trend was only
386 recorded in one catchment (Boisvinet catchment) with an acceleration of organic accumulation around
387 1970. For the five catchments recording a positive trend, the average rate of deposition raised from 67
388 ± 35 gC m⁻² yr⁻¹ before 1950 to 134 ± 66 gC m⁻² yr⁻¹ after 1950 (Fig. 5). In the two reservoirs where no
389 statistical trend was observed (MK test $P < 0.05$) - (Brosse and Passavant reservoirs), a significant
390 increase of productivity was recorded between 1940 and 1975 (Fig. 5). On average, rates of organic
391 deposits reached 260 ± 44 gC m⁻² year⁻¹ before 1950, 350 ± 302 gC m⁻² year⁻¹ between 1950 and 1975
392 and 318 ± 126 gC m⁻² year⁻¹ after 1975.

393

394 *3.4. Agricultural inputs*

395 The analysis of N fertilizer inputs (mineral + organic) of the nine selected catchments shows
396 the occurrence of three main trends. In the six catchments occupied by arable land and grassland, a
397 significant increase in N fertilizer use was observed during the last century (MK test $P < 0.05$) - (Table
398 4) In these sites, a major acceleration occurred around 1966 ± 5 yr (BU test) – (Fig. 6). On average,
399 this consumption started to stabilize or to decrease in 1994 ± 5 yr. For these catchments, consumption
400 of N fertilizers increased on average by $60 \pm 10\%$ for the 1955-2015 period. No statistical trend (MK
401 test $P > 0.05$) was observed for the Boisvinet catchment (site 5, Fig. 1) located in lowland
402 environments dominated by arable land (Table 2).

403 Finally, the two catchments currently occupied by forest displayed a negative or the absence of any
404 significant statistical trend in N fertilizer inputs (for the Malaguet and the Prugnolas catchment,
405 respectively) – (Table 4). For the Malaguet site, a significant decrease in consumption was recorded
406 around 1975. The use of N fertilizer inputs decreased by 20 % at this site (Fig. 6).

407

408

409 4. Discussion

410

411 *Local records*

412

413 The non-linear decrease and the independent variation of paleo-production proxies through
414 time (chlorophyll-a, sedimentary bulk organic matter $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ and C/N concentration) does not
415 suggest the post-depositional degradation of organic matter as no typical increasing logarithmic trend
416 of TOC and TN concentrations was observed in the upper part of the sediment cores (Fig. 2, 3 and 4) -
417 (Emerson *et al.*, 1985; Arndt *et al.*, 2013). Variation in individual proxies can therefore be attributed to
418 a change in organic matter sources.

419 Results obtained on the nine sedimentary sequences collected at the outlet of contrasted
420 catchments showed variable contributions of terrestrial versus autochthonous organic matter during
421 the 20th century. These changes in organic matter sources were attributed to anthropogenic pressures
422 observed in the landscape, with the intensification of anthropogenic activities and a greater use of
423 agricultural inputs (e.g. Schindler Wildhaber *et al.*, 2012).

424 In the two forested catchments, the period prior to 1960 was characterized by the highest C/N
425 ratios recorded in this study (ranging between 17 and 24), suggesting the dominant delivery of
426 vascular debris (C/N >20 from vascular plants (Meyers, 1994)). These results were in agreement with
427 our historical knowledge on the processes occurring in these catchments. During the 20th century, both
428 catchments were affected by land abandonment, evolving from an open landscape, mostly occupied by
429 grassland, to forested catchments. Between 1950 and 1960, major afforestation works were conducted
430 in the Malaguet catchment inducing a significant acceleration of soil erosion (Foucher *et al.*, 2019b).
431 The forest regrew spontaneously in the Prugnolas catchment and, as a consequence, soil erosion
432 decreased progressively during the 20th century. The increase of values recorded by terrigenous
433 organic matter proxies (C/N, TOC concentration) during the 1950-1960 period was recorded during
434 this period of afforestation, soil disturbance and soil erosion. After this period, organic sediment
435 sources have drastically changed as reflected by an increase of TOC contents in sediment (from 130 to
436 170 gC kg⁻¹ and from 110 to 150 gC kg⁻¹) as well as a decrease in the C/N ratios (from 24 to 13 and

437 from 16 to 11, for Prugnotas and Malaguet reservoirs, respectively) – (Fig. 2). In contrast, during this
438 period, chlorophyll-a increased (450% and 160% for Prugnotas and Malaguet reservoirs, respectively).
439 The strong negative relationship observed between chlorophyll-a and $\delta^{13}\text{C}$ ($r = 0.99$ and 0.87 for
440 Prugnotas and Malaguet reservoirs, respectively) suggests a greater contribution of autochthonous
441 production during the second half of the 20th century. During this period, agricultural inputs remained
442 constant or even decreased (Fig. 6), and they were not correlated to paleo production proxies (e.g.
443 correlation between chlorophyll-a vs N inputs $r = 0.02$ and 0.5 for Prugnotas and Malaguet lakes). In
444 the Prugnotas catchment, the increase of ^{137}Cs activities recorded in sediment deposited after 1999 was
445 attributed to a greater contribution of surface soil erosion during a landscape management period
446 following the major 1999 windstorm (Foucher *et al.*, 2019a). During this period both TN
447 concentration and $\delta^{15}\text{N}$ increased as well as chlorophyll-a values ($r = 0.98$ and 0.99 , respectively) –
448 (Fig. 2). Acceleration of autochthonous production during the last 25 years can be therefore attributed
449 to a release of soil nutrients.

450 In grassland areas (drained by the Beaurepaire, Tazenat and Loroux-Bottereau reservoirs),
451 changes in sedimentary matter signatures were also recorded during the 20th century. Between 1950
452 and 1970, a general increase in TOC contents (on average 120 ± 110 %) associated with higher C/N
453 organic ratios (average value of 9.5 ± 1), an increase of MARorg rate (140 ± 70 %) and changes in $\delta^{13}\text{C}$
454 and $\delta^{15}\text{N}$ proxies were observed (Fig. 3). A similar observation was made in catchments dominated by
455 arable land (Passavant and Brosse basins). During this same period, TOC contents increased on
456 average by 63 ± 1 % and were associated with an acceleration of MARorg deposition (84 ± 40 %) and
457 high C/N (11.5 ± 0.5). In addition, this period corresponds to an increase of ^{137}Cs activity suggesting a
458 mobilization of material originating from surface soil sources (Olley *et al.*, 2013) for le Loroux,
459 Beaurepaire, Tazenat and Passavant catchments (Fig. 3 and 4).

460 In both contexts of arable land and grassland, this post Second World War (WWII) period corresponds
461 to a phase of intensification of agricultural practices across the Loire river basin, with the
462 implementation of major landscape modifications (e.g. land consolidation corresponding to the
463 reorganization and the increase of plot size, stream redesign, tile drain creation) - (Vandromme *et al.*,
464 2016; Chartin *et al.*, 2013; Grangeon *et al.*, 2017). The change in proxy values observed in the current

465 research together with this increase of accumulation of organic matter characterized by high C/N ratios
466 very likely occurred in response to these landscape modifications (Mackie *et al.*, 2005).
467 After this period of more intense management (after 1970) the MARorg, TOC and chlorophyll-a
468 proxies increased following a statistically significant trend (Table 4). These results suggest a greater
469 accumulation of autochthonous organic matter. MARorg values obtained in agricultural and
470 abandoned catchments during the 2010-2017 period (255 ± 170 and 78 ± 22 gC m⁻² yr⁻¹, respectively)
471 are in agreement with those obtained in previous studies (Fig. 5). For example, a median value of 350
472 gC m⁻² yr⁻¹ and 56 gC m⁻² yr⁻¹ were respectively recorded in reservoirs draining cultivated and
473 abandoned catchments - (Schlesinger & Bernhardt, 2013).

474

475 *Regional trend*

476

477 The compilation of results obtained in the seven agricultural catchments demonstrates the
478 continuous acceleration in organic material accumulation in ponds across the Loire River basin during
479 the 20th century (MK-test P <0.05). Similar observations were made in agricultural catchments of the
480 northern United-States (Heathcote *et al.*, 2013). In this area, the regional acceleration started around
481 1950 and was accompanied by an increase of C/N ratios in sediment (Fig. 7). In previous
482 paleolimnological studies, these increases were interpreted as historical periods characterized by the
483 delivery of a high proportion of organic matter derived by soil erosion particularly during periods of
484 greater human-induced environmental modifications (Guilizzoni *et al.*, 1996; Routh *et al.*, 2004). In
485 addition to this change, $\delta^{15}\text{N}$ signature recorded a general decrease suggesting a major change in
486 organic matter sources. The beginning of acceleration of organic matter deposits (1950) recorded in
487 these reservoirs occurred concomitantly with the major landscape changes that took place in the Loire
488 river basin. These changes were associated with an acceleration of erosion processes and a higher
489 sediment delivery to the reservoirs (Foucher *et al.*, 2014). This period of soil erosion was observed in
490 various catchments across Europe. For example, a two to ten-fold acceleration of soil erosion rates was
491 recorded in various catchments of the UK after 1950 in response to land-use change (Foster *et al.*,
492 2011). In addition, the greater contribution of phytoplanktonic production was also observed from

493 1955 onwards: chlorophyll-a started to increase following the regional trends of N fertilizer inputs ($r =$
494 0.9 between chlorophyll-a contents and N inputs during the 1955-1990 period). From 1970 onwards,
495 the terrigenous contribution decreased sharply following the trend of MARorg. After 1970, human
496 pressure on landscape started to decrease in France (land consolidation programs mainly took place
497 between 1955 and 1975, Andre & Polombo, 2013). After a period dominated by the terrigenous
498 contribution to sediment deposited in the reservoirs, the MARorg showed a progressive acceleration
499 from 1970 to 2015 (+40%). At the regional scale, $\delta^{15}\text{N}$ is well correlated to N inputs ($r = 0.76$), C/N
500 ratio ($r = 0.8$), to MARorg after 1970 ($r = 0.72$) as well as to chlorophyll-a ($r = 0.68$). The increase of
501 organic matter deposition associated with a good correlation between phytoplanktonic proxies
502 demonstrates a shift in the paleo productivity evolving from a system dominated by terrigenous inputs
503 just after WWII to a period of dominant autochthonous production after 1970.

504 Between 1990 and 2015, the European legislation induced a slight reduction in fertilizer use in the
505 Loire river basin (-13% on average) - (Poisvert *et al.*, 2017). Nevertheless, organic matter continues to
506 accumulate in reservoirs (+17% in 15 years). The MARorg remained highly correlated to
507 phytoplanktonic proxies during this period ($r = 0.87$ between chlorophyll-a and MARorg). The
508 negative correlation between N inputs and phytoplanktonic occurrence ($r = 0.95$ between chlorophyll-a
509 and N inputs and $r = 0.97$ between MARorg and N inputs) highlights the effects of other driving
510 factors and not only the trends in agricultural inputs. Past studies have demonstrated that in addition to
511 human activities, climate warming or again secondary release of nutrients (Shayo & Limbu, 2018)
512 may have a major impact on organic production and eutrophication phenomena.

513 Nowadays, although eutrophication represents a major threat for a large number of reservoirs
514 across the world (Le *et al.*, 2010; Bhagowati & Ahamad, 2019), this process remains insufficiently
515 studied in small headwater basins. The general positive trend of paleo production proxies observed in
516 study could be improved by the collection of additional samples in various catchments of the Loire
517 basin. Nevertheless, the results obtained in the current research emphasized the occurrence of a
518 massive acceleration of organic matter deposition after 1950 in these reservoirs. Moreover, the
519 sediment recorded in ponds draining contrasting land uses also showed a transition between a system
520 dominated by terrigenous inputs to a system dominated by the autochthonous production during the

521 1950-2015 period. Although the paleo production proxies were highly correlated to the use of
522 agricultural inputs until the 1990s, this relationship was no longer observed since then. Accordingly,
523 questions remain to identify the factor(s) driving the recent autochthonous production in these
524 reservoirs. Overall, soil erosion is classically considered to be the main factor inducing siltation of
525 water bodies. As the excess of organic matter production can also lead to the filling of water bodies,
526 the contribution of these processes and their respective contribution to reservoir siltation need to be
527 further quantified. This will allow the design of effective control measures such as the systematic
528 sowing of cover crops in winter to improve nutrient absorption by plants and better protect soils
529 against erosion.

530

531 5. Conclusions

532

533 The collection and analysis of sediment accumulated in freshwater bodies provides a powerful
534 technique for reconstructing changes in organic matter sources and for investigating their relationship
535 with landscape management. Although these changes were studied during the 20th century in coastal
536 areas, great lakes or during the last decades in riverine systems, there is currently a lack of records to
537 support a regional analysis of these changes in continental areas over the last century. Results obtained
538 in the current research underline an acceleration of organic matter deposition rates in cultivated areas
539 during the second part of the 20th century. Between 1950 and 1965, organic matter that accumulated in
540 water bodies draining agricultural catchments was mainly supplied by terrigenous sources. These
541 deposits occurred during the implementation of major landscape management programs such as land
542 consolidation, soil drainage or stream re-design. The increasing use of synthetic fertilizers which
543 started in the 1970s induced a shift in organic matter accumulation with the acceleration of primary
544 production in water bodies. This autochthonous production was highly correlated to the use of N
545 fertilizers until the early 1990s. Nevertheless, the current research contributed to improve our
546 understanding of long-term trends in eutrophication through the identification of a switch in
547 production and the associated driving factors during the 1960-1970 period. Since then, although the

548 primary production continues to increase, this change cannot be exclusively explained by the enhanced
549 use of fertilizers. The other factors driving this change should therefore be identified and quantified.
550 This identification is of prime importance in order to reduce primary production in these water bodies
551 and to improve the water quality of freshwater systems.

552

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559

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806 Figures:

807

808 Fig.1 Localization of the nine studied catchments within the Loire river basin. (1) Le Loroux-
809 Bottereau, (2) Beaurepaire, (3) Passavant-sur-Layon, (4) Brosse, (5) Boisvinet, (6) Prugnolas,
810 (7) Goule, (8) Tazenat, (9) Malaguet

811

812 Fig. 2 Evolution of sediment properties throughout time in reservoirs draining forested
813 catchments: (6) Prugnolas and (9) Malaguet. Grey lines correspond to the identification of
814 ^{137}Cs peaks associated to the 1963 and 1986 fallouts.

815

816 Fig. 3 Evolution of sediment properties throughout time in reservoirs draining catchments
817 dominated by grassland: (1) Le Loroux-Bottereau, (2) Beaurepaire, (7) Goule and (8) Tazenat.
818 Grey lines correspond to the identification of ^{137}Cs peaks associated to the 1963 and 1986
819 fallouts.

820

821 Fig. 4 Evolution of sediment properties throughout time in reservoirs draining catchments
822 dominated by arable land: (3) Passavant-sur-Layon, (4) Brosse and (5) Boisvinet. Grey line
823 correspond to the identification of ^{137}Cs peak associated to the 1963 fallouts.

824

825 Fig. 5 Evolution of Nitrogen consumption in the nine selected catchments under arable land,
826 grassland or dominated by forest (1) Le Loroux-Bottereau, (2) Beaurepaire, (3) Passavant-sur-
827 Layon, (4) Brosse, (5) Boisvinet, (6) Prugnolas, (7) Goule, (8) Tazenat, (9) Malaguet

828

829 Fig. 6 Evolution of Mass Accumulation Rates of organic material (MARorg) during the last
830 century in reservoirs located at the outlet of the nine studied catchments: (1) Le Loroux-
831 Bottereau, (2) Beaurepaire, (3) Passavant-sur-Layon, (4) Brosse, (5) Boisvinet, (6) Prugnolas,
832 (7) Goule, (8) Tazenat, (9) Malaguet. Grey area correspond to a significant acceleration of
833 MARorg.

834

835 Fig. 7 Evolution of sediment properties throughout time in reservoirs draining the seven
836 cultivated catchments. The light grey part corresponds to the period dominated by terrigenous
837 inputs whereas the dark grey colour corresponds to the period dominated by autochthonous
838 primary productivity.

839

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841 Tables:

842

843 Table 1. Characteristics of the Loire Basin sub-catchments

844

845 Table 2. Characteristics of the studied catchments

846

847 Table 3. Chronology of sediment cores and modern and historic Mass Accumulation Rates of

848 organic matter (MAR_{org} expressed in $\text{gC m}^{-2} \text{yr}^{-1}$: modern MAR_{org} correspond to average

849 value of MAR_{org} for the 2017-1950 period and the historical MAR_{org} to the average value

850 before 1950). ND = non detected.

851

852 Table 4. Results of the Mann Kendal test with the significant p value <0.05 . NT = No trend, +

853 = positive trend and - = negative trend

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855

	Sub catchment area (km ²)	Average elevation (m)	Average slope (%)	Land use
Lower Loire	36,780	108	1.7	Arable land (60%), Grassland (24%), Forest (11%), Urban (5%)
Middle Loire	48,471	216	2.1	Arable land (48%), Forest (25%), Grassland (24%), Urban (4%)
Upper Loire	32,630	570	5.5	Grassland (43%), Forest (35%), Arable land (17%), Urban (4%)

Tables

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Site ID	Reservoir name	Core location	Core length (cm)	Water depth (m)	Lake/Pond area (ha)	Catchment area (km ²)	Average elevation (m)	Average slope (%)	Catchment location	Main land use	Land use	Landscape management
1	Loroux-Botterau	47.2379N, -1.3440E	71	1.2	1.7	4.5	68	2.7	Lower Loire	Grassland (65%)	Arable land (34%). urban area (1%)	Hedge removal during the 1965-1975 period
2	Beurepaire	47.0726N, -0.4737E	108	2.5	38	14.3	122	1.7	Lower Loire	Grassland (52%)	Arable land (45%). water bodies (3%). woodland (0.1%)	tile drain implementation and land consolidation in 1977
3	Passavant-sur-Layon	47.1013N, -0.3941E	116	1.4	14	108	121	2.4	Lower Loire	Arable land (70.5%)	Grassland (21%). woodland (6.36%). water bodies (0.9%). mining (0.6%). urban areas (0.3%)	Major land consolidation during the 1960's
4	Brosse	47.2761N, 1.0558E	127	2.4	11	24.5	105	2.3	Middle Loire	Arable land (83%)	Woodland (8%). grassland (7%). water bodies (1%). urban areas (1%)	Major land consolidation in 1959
5	Boisvinet	48.0733N, 0.8832E	97	2.1	39	9.5	189	2.5	Lower Loire	Arable land (63%)	Grassland (23%). woodland (9.5%). water bodies (4.3%)	
6	Prugnotas	45.8689N, 1.9032E	82	0.8	1.8	7.8	727	12	Middle Loire	Woodland (82%)	Grassland (16%). arable land 0.4%)	Afforestation after 1950, major windstorm in 1999 inducing damage on forest
7	Goule	46.7277N, 2.8004E	152	4.3	52	34	242	3	Middle Loire	Grassland (65.9%)	Woodland (18.4%). arable land (13.3%). water bodies (1.5%). urban areas (0.8%)	
8	Tazenat	45.9796N, 2.9900E	115	67	30	2.3	690	9.3	Upper Loire	Grassland (68%)	Woodland (16%). water bodies (15%)	Land consolidation in 1990
9	Malaguet	45.2512N, 3.7103E	69	3.1	20.5	3.7	1073	7.5	Upper Loire	Woodland (71%)	Grassland (13%). water bodies (7%). urban areas (6%). arable land (2%)	Afforestation works between 1950 and 1965

Site ID	Max $^{210}\text{Pb}_{\text{ex}}$ date / depth	1986 ^{137}Cs fallout	1963 ^{137}Cs fallout	Modern MARorg	Historic MARorg
1	1921 (71 cm)	ND	± 3 year (40 cm)	133 ± 32	46 ± 21
2	1925 (105 cm)	ND	± 2 year (70 cm)	219 ± 71	76 ± 12
3	<100 (74 cm)	ND	± 2 year (46 cm)	229 ± 50	133 ± 11
4	1922 (130 cm)	ND	± 2 year (92 cm)	460 ± 117	342 ± 126
5	1898 (90 cm)	ND	± 3 year (50 cm)	111 ± 28	76 ± 2
6	1918 (140 cm)	± 1 year (55 cm)	± 1 year (85 cm)	167 ± 12	114 ± 25
7	1901 (65 cm)	± 1 year (11.5 cm)	ND	141 ± 46	450 ± 330
8	1902 (44 cm)	± 1 year (11 cm)	± 3 year (19 cm)	41 ± 11	21 ± 12
9	1948 (76 cm)	± 4 year (16 cm)	ND	163 ± 73	-

Tables

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Site ID	TOC		$\delta^{13}\text{C}$		$\delta^{15}\text{N}$		C/N		TN		chlorophyll-a		MARorg		N Inputs	
1	0.46	NT	<0.05	-	<0.05	-	<0.05	-	0.46	NT	<0.05	+	<0.05	+	<0.05	+
2	<0.05	+	<0.05	-	<0.05	+	0.3	NT	<0.05	+	<0.05	+	<0.05	+	<0.05	+
3	<0.05	+	<0.05	-	<0.05	+	0.27	NT	<0.05	-	<0.05	+	0.53	NT	<0.05	+
4	0.84	NT	0.82	NT	0.07	NT	0.11	NT	0.96	NT	<0.05	+	0.33	NT	<0.05	+
5	<0.05	+	0.6	NT	<0.05	+	0.46	NT	<0.05	+	<0.05	+	<0.05	+	0.43	NT
6	<0.05	+	<0.05	-	0.19	NT	<0.05	-	<0.05	+	<0.05	+	<0.05	+	<0.05	+
7	0.138	NT	0.408	NT	<0.05	-	0.5	NT	<0.05	+	0.06	NT	<0.05	-	0.38	NT
8	<0.05	+	<0.05	-	0.54	NT	0.46	NT	<0.05	+	0.38	NT	<0.05	+	<0.05	+
9	0.13	NT	<0.05	-	0.45	NT	0.23	NT	<0.05	+	<0.05	+	<0.05	-	<0.05	-

Figure 1
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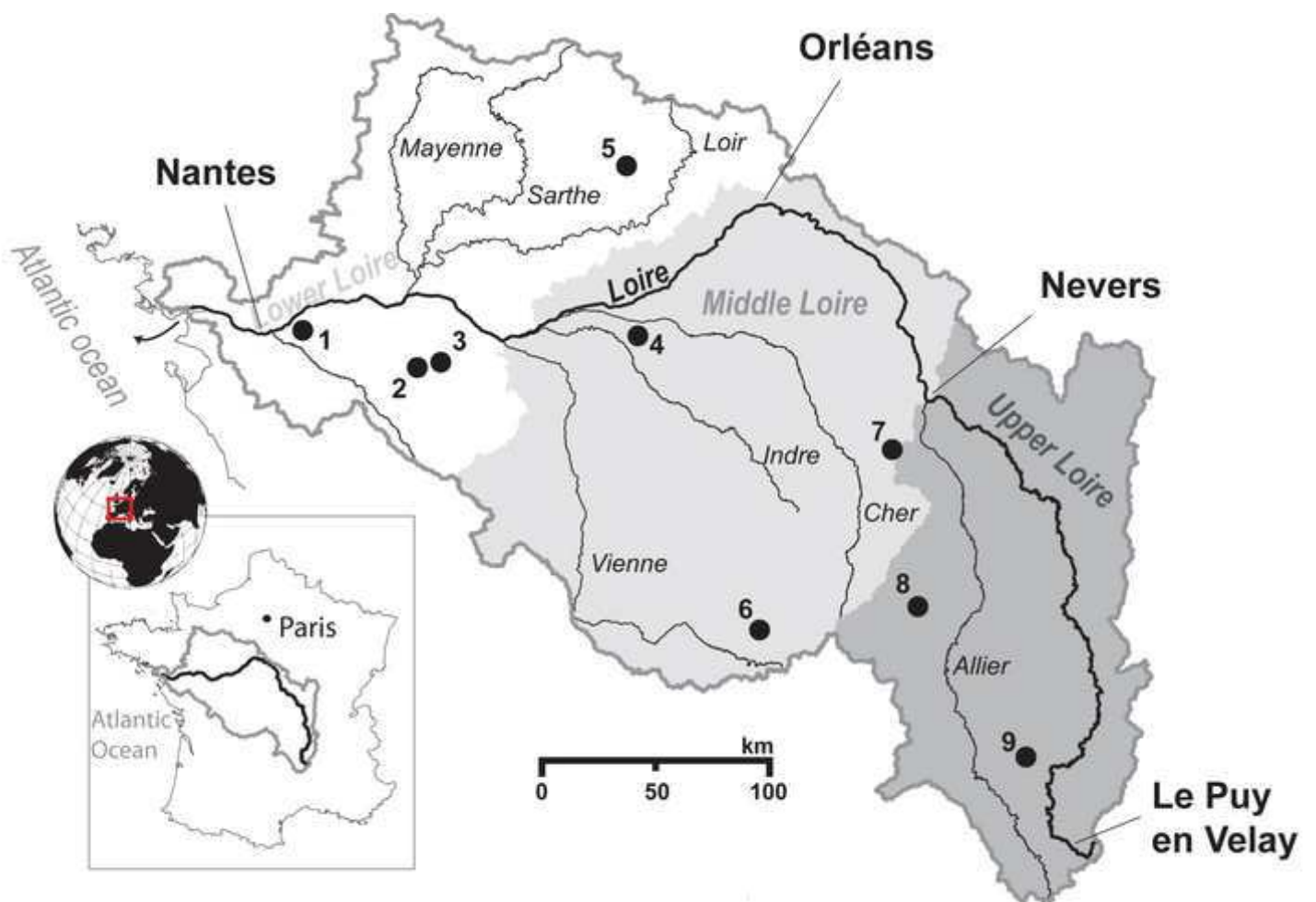


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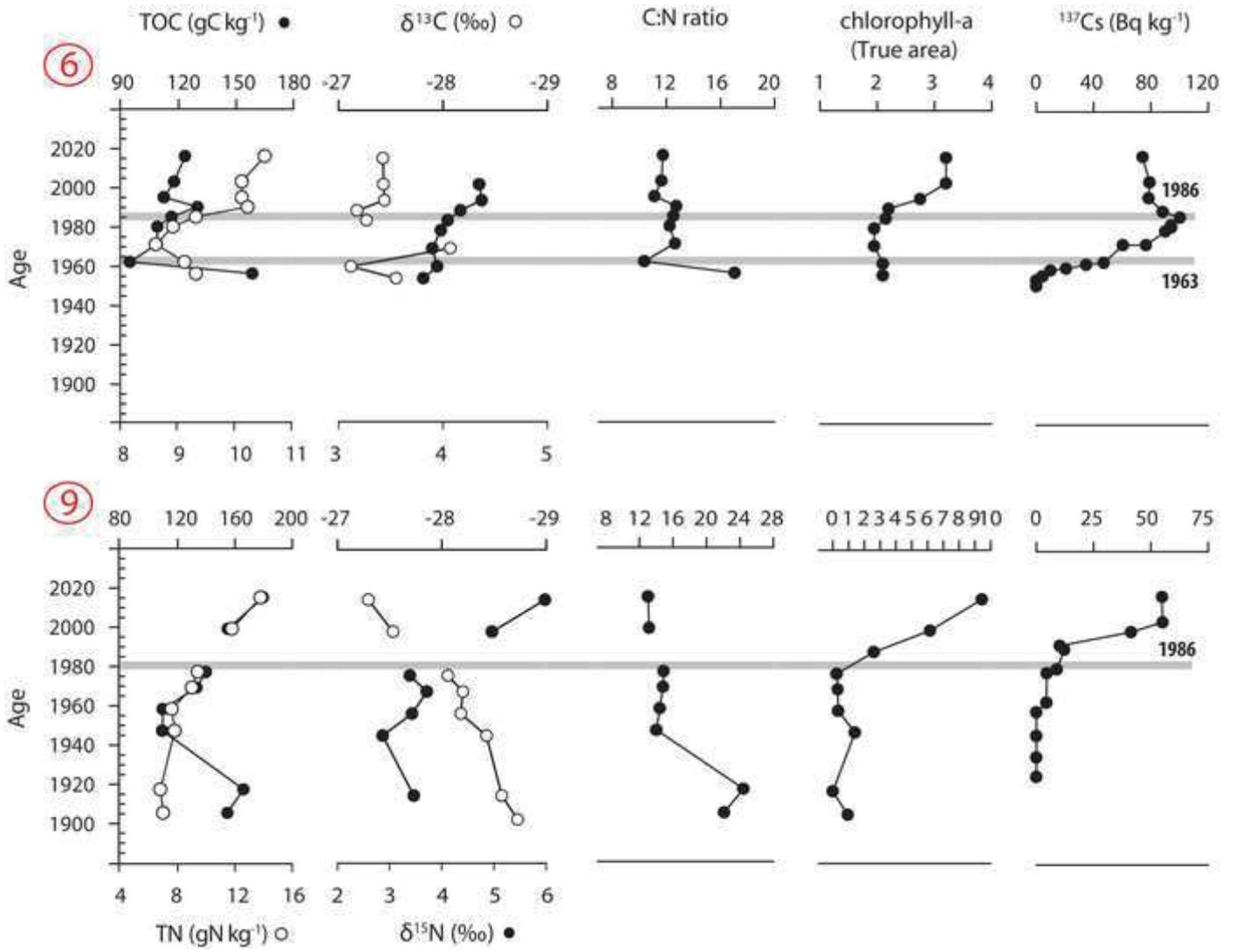


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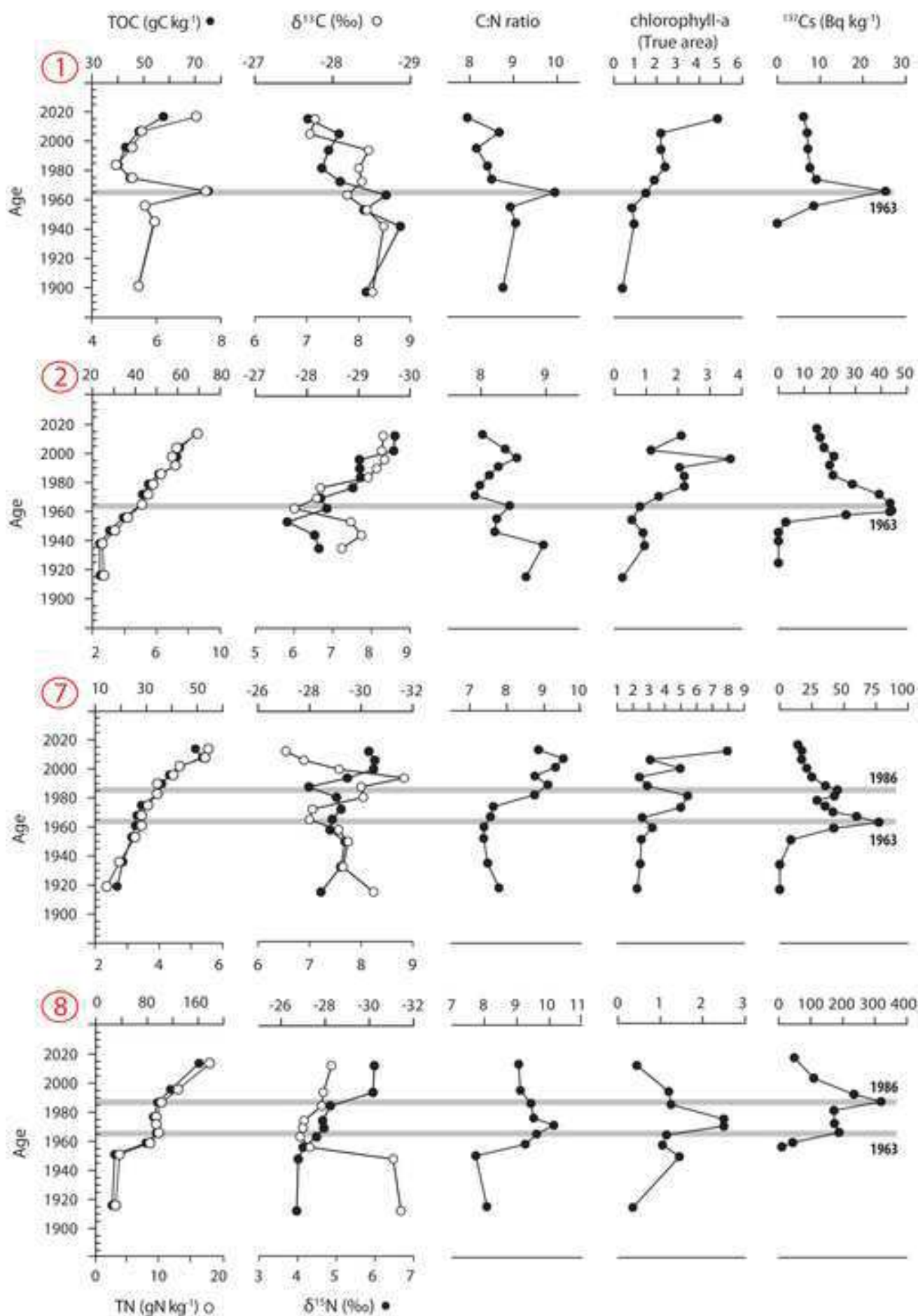


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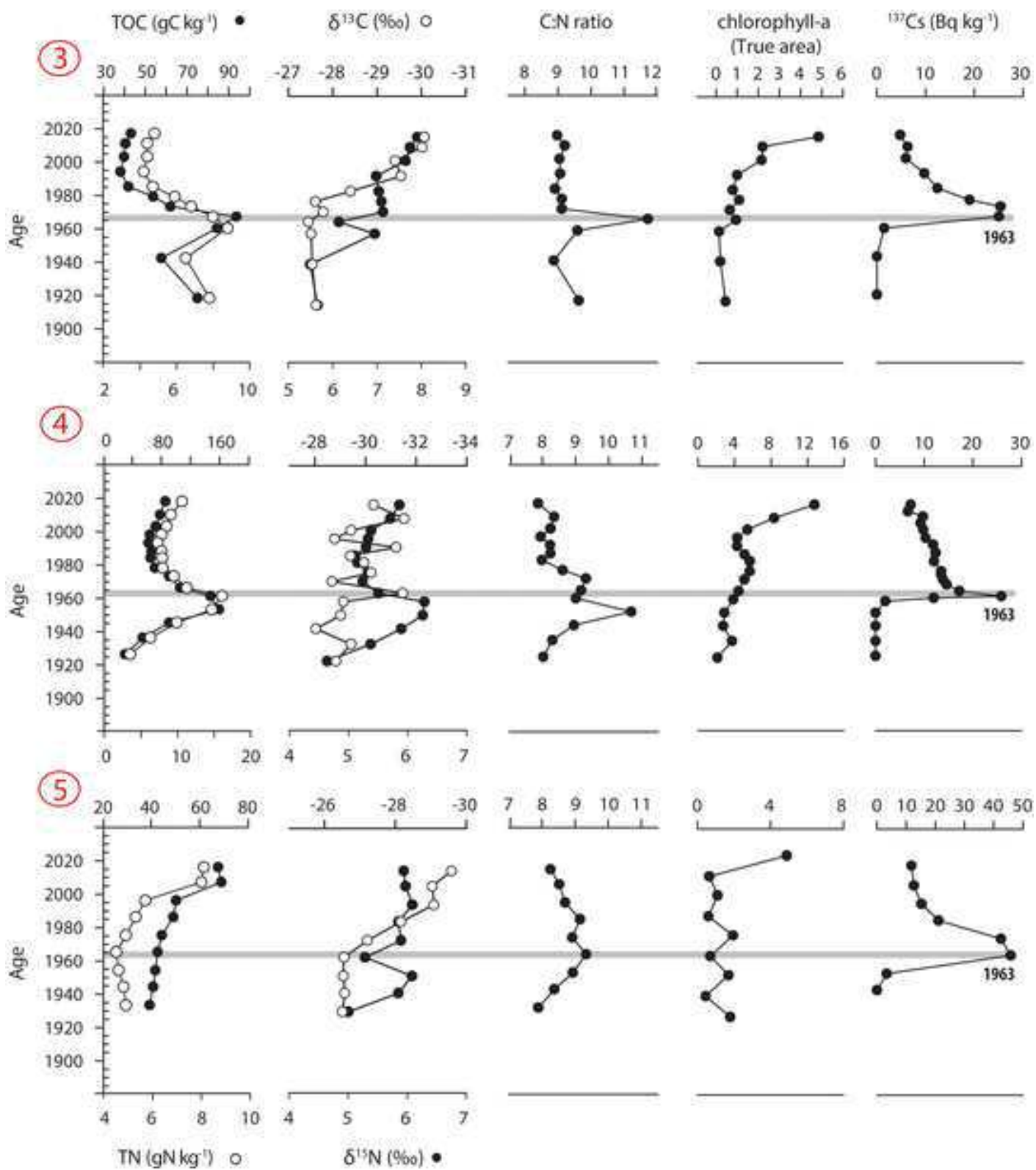


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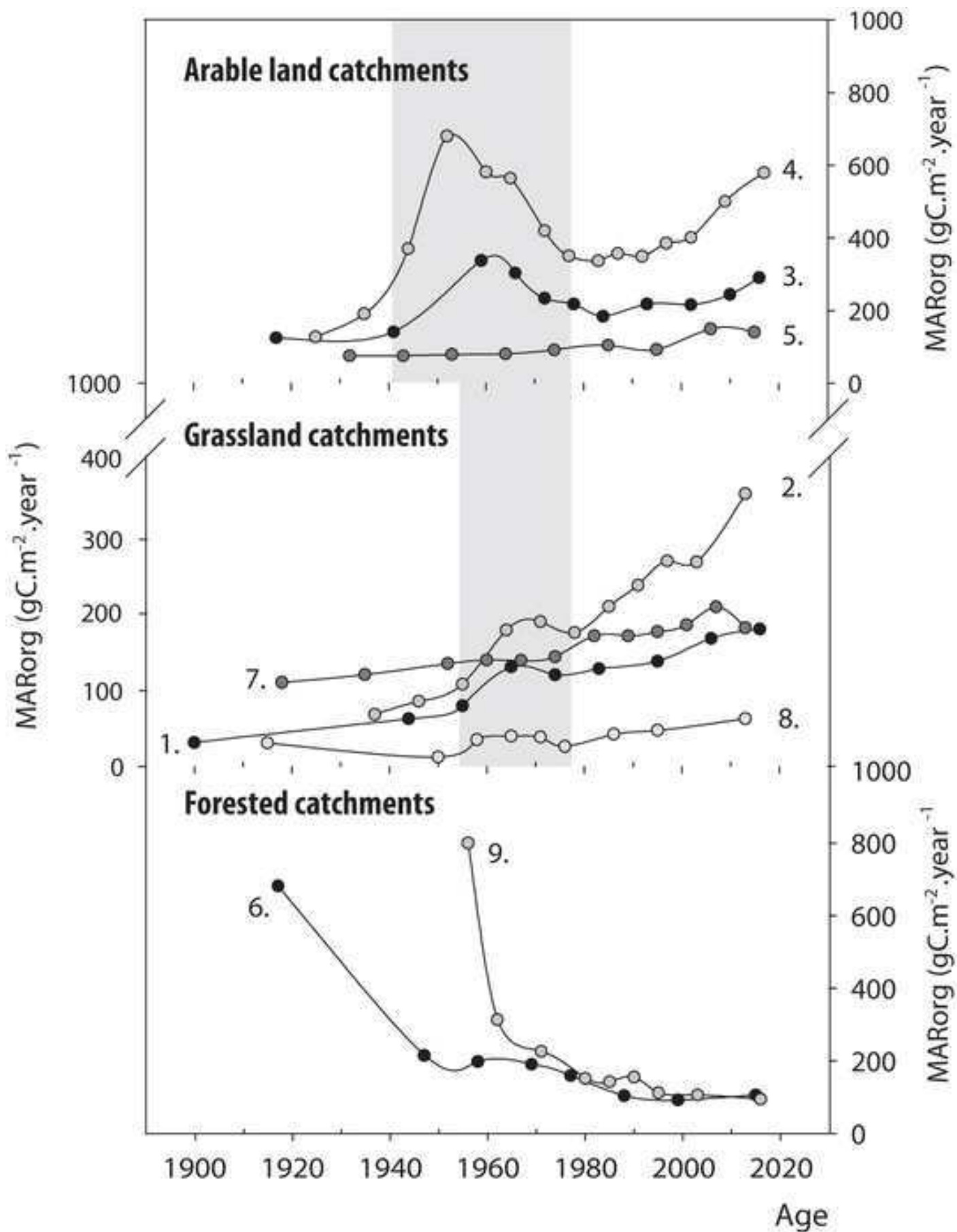


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