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1 **Reconstructing the impact of nickel mining activities on sediment supply to**
2 **the rivers and the lagoon of South Pacific Islands: lessons learnt from the Thio**
3 **early mining site (New Caledonia)**

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15

16 **Abstract**

17 Opencast mining has exacerbated land degradation in New Caledonia, a French archipelago
18 located in the south-west Pacific Ocean. Developed since the 1880s, mining has become the major
19 economic activity in some catchments, which strongly disrupted sediment dynamics. Reconstructing
20 the temporal changes of sediment source contributions is essential to understand the driving factors
21 of soil erosion in response to i) the occurrence of cyclones, ii) the changes in mining practices during
22 the last several decades, and iii) other soil degradation processes such as extensive soil erosion
23 induced by fires, overgrazing and trampling of invasive species, and landslides. Accordingly, a multi-
24 parameter analysis including gamma spectrometry, color and X-ray fluorescence measurements was
25 conducted on a sediment core collected in a deltaic floodplain at the outlet of one of the first areas
26 exploited for nickel mining, the Thio River catchment (397-km²). One geochemical tracer (*i.e.* K) has
27 been used to quantify changes in sediment sources in the successive sediment layers deposited since
28 the beginning of mining activity. The results showed that the contribution of mining tributaries
29 largely dominated, with a mean contribution of 74 % (SD 13 %) of material sampled in the sediment
30 core. This contribution notably increased after the mechanization of mining activities (*i.e.* from
31 1950s; increase of 18 %). The occurrence of Cyclone Alison in 1975 triggered the progressive transfer
32 of mining waste accumulated on the foothills over 25 years into the river system. This tipping point
33 could be identified in the sediment sequence, which demonstrates that over the last 41 years (*i.e.*
34 1975-2016), a ~84-cm deep sediment deposit has accumulated in the alluvial floodplain of the Thio
35 River catchment (mean annual deposition rate of 2 cm yr⁻¹). Currently, the progressive release and
36 downstream transfer of this mining waste is still ongoing ~45 years after Cyclone Alison. Although
37 environmental legislation was introduced in 1975, mining tributary contributions to sediment still
38 dominate (80 %, SD 5 %). Overall, this multi-proxy approach to examining the cumulative effects of
39 mining activities on downstream sediment dynamics could be implemented in other mining
40 catchments of New Caledonia and around the world to compare the respective mining source
41 contributions to sediment and their evolution throughout time in these contrasted areas.

42 **Keywords:** Soil erosion • sediment fingerprinting • nickel mining • mechanization period

43

44 **1. Introduction**

45 The ongoing globalization and the growing demand from the increasing world population for
46 food, raw materials and services are causing ever-increasing pressures on the environment. Soil
47 erosion, one of the major global environmental threats, has accelerated in particular in recent
48 decades as a result of changes in landscapes induced by human activities: vegetation clearing and
49 deforestation (Restrepo et al., 2015), crop cultivation (Schmidt et al., 2018), over-grazing,
50 industrialization (Stinchcomb et al., 2013) and urbanization (Russell et al., 2017). Non-renewable soil
51 resources are being depleted (Amundson et al., 2015; Borrelli et al., 2017) while river and coastal
52 systems are being degraded by the excess supply of fine sediment (Syvitski et al., 2005). These
53 impacts are particularly exacerbated in tropical regions exposed to heavy rainfall (Lal, 1983; Restrepo
54 and Syvitski, 2006; Zhu et al., 2008). Tropical depression and cyclone-induced-overbank floods are
55 responsible for most of the sediment transfers observed annually in these river systems (Baltzer and
56 Trescases, 1971; Danloux and Laganier, 1991; Terry et al., 2008). An understanding of the sources,
57 transfer and storage of fine sediment in these river systems is required to guide the implementation
58 of effective management measures. However, the scientific diagnosis of human-induced soil erosion
59 is a challenging problem (García-Ruiz et al., 2017; Muchena et al., 2005). Disentangling the impacts of
60 anthropogenic activities on soil erosion from climatic factors is challenging (Walling and Webb 1996;
61 Restrepo and Syvitski 2006; Raclot et al. 2016). A second factor that may complicate the diagnosis is
62 that multiple anthropogenic activities generally occur during the same timeframe and on the same
63 landscape. Accordingly, it is generally difficult to clearly identify the main human activity controlling
64 soil erosion in a given catchment (Collins and Walling, 2004). Finally, the chronology of
65 anthropogenic activities at the decadal or century scale needs to be considered in order to fully
66 interpret the variability of the current suspended sediment loads in river systems (Dearing et al.,
67 2006; Hoffmann et al., 2010). Suspended sediment fluxes monitored at the outlet can be attributed
68 to current human activities. However, they can also be attributed to the erosion and/or
69 remobilization of legacy sediment mobilized by past human activities, for example agro-pastoralism

70 (Bajard et al., 2017) or mining activities (Balamurugan, 1991; Coulthard and Macklin, 2003; Navarro
71 et al., 2008).

72 All these complexities and questions can be found especially in New Caledonia. Indeed, this
73 archipelago, located in the south-west Pacific Ocean holds 25 % of the world nickel resources, making
74 it the sixth largest nickel producer. Started in 1880, mining activities have continued to intensify,
75 particularly with the mechanization of extractive industries from 1950s. This rapid increase of nickel
76 ore production was accompanied by a sharp increase in bare soil areas (*i.e.* mining sites, mining
77 prospection areas and mining roads) and mining waste (Iltis, 1992). In the absence of environmental
78 legislation, 200 million tons of mining waste have been stored on the foothills exposed to rainfall.
79 During the 1970s-1980s, a series of cyclones (*i.e.* Cyclone Colleen in 1969, Cyclone Gyan in 1981 and
80 Cyclone Anne in 1988) hit the archipelago leading to the dumping of this mining waste into the river
81 systems. Since then, the archipelago is confronted with an unprecedented problem of sediment
82 pollution of these river networks: hyper-sedimentation threatening both the inhabitants (*e.g.*
83 increased flooding risk, water pollution by sediment-bound heavy metals) and the island's
84 ecosystems (*e.g.* degradation of coral reef systems). The mining sector, although having contributed
85 largely to New Caledonia's economic growth, is considered to be the main driver of environmental
86 degradation (Bird et al., 1984; Iltis, 1992). This awareness of the environmental impacts of mining
87 activities led to the implementation of the first environmental regulations as early as in 1975 (*e.g.*
88 prohibition of direct dumping of mining waste rocks on the hillslopes) and the adoption of a Mining
89 Code in 2009 (*e.g.* implementation of mitigation hydraulic infrastructures such as retention ponds)
90 aimed at reducing sediment supply to the river network. However, other sources of sediment
91 independent of mining activities may also supply a significant quantity of sediment to New
92 Caledonian river systems and they should be distinguished from mining-related inputs. For instance,
93 bushfires and land clearing fires are regularly triggered by the local population to increase the area of
94 cultivable land (Dumas, 2010). These fires can lead to shallow landslides and/or extensive soil erosion
95 of surface material on hillslopes (Blake et al., 2009; Nyman et al., 2011; Smith et al., 2011). Moreover,

96 the deer population, an invasive species introduced in the early 20th century in New Caledonia,
97 significantly increased during the last 40 years. The associated trampling may have further
98 accelerated surface soil degradation (Shellberg et al., 2010).

99 A recent pilot sediment tracing study was conducted in the Thio River (397 km²) catchment
100 draining the archipelago's first mine (Sellier et al., 2020) to estimate the contributions of sediment
101 sources during two recent cyclonic events (*i.e.* Cyclone Marcia in 2015 and Cyclone Cook in 2017).
102 The authors showed that the spatial distribution of rainfall generated by these two events may
103 locally modify the respective contributions of non-mining and mining tributaries. However,
104 tributaries draining mining sources dominated the sediment supply to the main stem of the Thio
105 River during these two events: 68 % (SD 28 %) in 2015 and 86 % (SD 7 %) in 2017. However, several
106 research questions remained after this first investigation: Do the recently observed mining
107 contributions correspond to the past and/or current mining source contributions? Are these
108 contributions from mining sources significantly lower than those potentially observed at the time of
109 the intensification of mining activities (*i.e.* from 1950s)? In other words, did environmental
110 regulations lead to a reduction of sediment inputs? To answer these questions, the temporal changes
111 of sediment source contributions should be reconstructed since the beginning of mining activities.
112 Indeed, reconstructing these contributions is essential to understand the driving factors of soil
113 erosion in early mining catchments of the South Pacific Islands in response to i) the occurrence of
114 cyclones, ii) the changes in mining practices during the last several decades, and iii) the occurrence of
115 other soil erosion processes. This ultimate goal of our research is to understand the environmental
116 impacts of the current mining activities into a broader historical context of sediment erosion, and
117 thus to better understand the sediment legacy of the past mining activities. In a post-colonial context
118 of empowerment, this country cannot make its economy sustainable without extractive industries
119 that represent ~5 % of the GDP and ~88 % of the exports in 2016; it has therefore to face challenges
120 related to their development, although it is associated with major socio-environmental concerns:
121 public health, conciliation with other economic activities (*e.g.* agriculture, fishery and tourism),

122 respect of cultural rights of local communities (Horowitz et al., 2018), and the preservation of
123 biodiversity (Fernandez et al., 2006). This knowledge is therefore essential to inform the choices of a
124 development policy for the mining sector and its management in New Caledonia for the coming
125 decades.

126 To this end, a multi-parameter analysis based on gamma spectrometry, color and X-ray
127 fluorescence measurements was conducted to a sediment core collected in the alluvial plain of the
128 Thio River catchment. This sediment core recorded the overbank floods and the intensive soil erosion
129 events that occurred over the last several decades. The objective of the current research is to i)
130 identify the flood deposits in this sediment core, ii) estimate by sediment fingerprinting methods the
131 sediment source contribution, iii) provide a dating of the sediment core in order to (iv) discuss the
132 evolution of these contributions in relation to the history of the Thio River catchment and its mining
133 activity.

134 **2. Materials and methods**

135 **2.1. Study area**

136 Located on the eastern coast of the New Caledonian archipelago's main island, called *la*
137 *Grande Terre* (ca. 17 000 km²), the Thio River catchment (397 km²) has been mined for nickel since
138 the 1880s. This catchment, like the rest of the archipelago, owes its abundance of minable nickel ores
139 to the widespread occurrence of peridotite massifs (*e.g.* harzburgite, dunite). Indeed, the weathering
140 of the peridotite results in the formation of Ni- and Fe-rich smectite, serpentine, goethite and
141 hematite. The migration of these transition metals through the soil profile results in higher Ni
142 concentrations in the saprolite (Ni: 0.5-3.5 %) and laterite layers (Ni: 0.9-1.8 %) (Trescases, 1973).
143 Peridotite massifs composed of laterites (18 % of the Thio River catchment area), peridotites (17 %),
144 serpentines (10 %) and harzburgites (1 %) are mainly located on the eastern part of this catchment
145 (Garcin et al., 2017) (Figure 1- a). The mountainous relief of the peridotite massifs (*i.e.* with a mean

146 altitude of 477 m asl, above sea level) contrast with the flatter western part of the catchment of
147 volcano-sedimentary formation rocks (*i.e* with a mean altitude of 368 m asl). These rocks are
148 composed of cherts (22 %), sandstone (9 %), a formation of basalt, dolerite and gabbro (6 %),
149 polymetamorphic rocks (6 %) and alluvia (4 %) (Figures 1-a and 1-b). The relief and the geology
150 control the spatial distribution of permanent vegetation covering 96 % of the catchment surface
151 area. Shrubland (35 %) and savannah (8 %) are found on peridotite massifs whereas the flatter
152 volcano-sedimentary formations are mainly covered with forests (37 %) and bushland (17 %) (Alric,
153 2009). Nevertheless, this permanent vegetation cover, which represents the main land use of the
154 Thio River catchment, can be degraded *inter alia* by the development of nickel mining activities
155 (Barselo, 1988). These open-mining casts are either active (*i.e.* Thio Plateau, Camp des Sapins since
156 1920) or abandoned (~12 sites, from the end of the 19th century to the 1970s, 21% of the catchment
157 area) (Figure 1-c).

158 The removal of vegetation related to these mining activities increase soil erosion already
159 exacerbated by the island's tropical climate, which is characterized by the succession of a hot wet
160 season (November-April; mean temperature of 27 °C) and a cooler dry season (May-October; mean
161 temperature of 20 °C). Furthermore, cyclones and tropical depressions occur frequently (*i.e.* every
162 2.7 years on average). These extreme rainfall events may supply more than 20 % of the annual
163 rainfall (average: 1620 mm; Météo France) and may cause overbank peak flow. The Thio River
164 catchment's river network is particularly reactive to heavy rainfall as most of the 12 tributaries
165 flowing into the main stem of the Thio River (28 km long, Figure 1-b) are governed by a torrential
166 regime (Dubreuil, 1974). In addition, the occurrence of 6 km² of mining roads (Garcin et al., 2017)
167 plays a paramount role in connecting mining zones devoid of vegetation to the river network (Alric,
168 2009). In general, extensive erosion processes are evident across the Thio River catchment, including
169 rills, gullies, landslides and channel bank erosion (Danloux, 1991). The drainage area of the mining
170 tributaries covers 60% of the Thio River catchment surface area compared to 40% for the non-mining
171 tributaries.

172 2.2. Sediment sampling

173 2.2.1 *Sediment sources*

174 Source samples were collected after two major floods (characterized by a ~10 year return
175 period): (1) the tropical depression of February 25, 2015 with lag deposits sampled between April 30
176 and May 5, 2015 (n= 21) and (2) Cyclone Cook on April 10, 2017, sampled between May 16 and 17,
177 2017 (n= 4). At each sampling site, between five to ten subsamples of fine sediment were collected
178 across a 10-m² surface with a plastic trowel at exposed subaerial sites free of vegetation on channel
179 bars. The subsamples were composited into one sample representative of the fine sediment
180 deposited on the channel bars. In total, 16 samples were collected in tributaries with mining
181 activities (*i.e.* mining tributaries), and eight samples were collected in tributaries draining areas
182 devoid of mining activities (*i.e.* non-mining tributaries) (Figure 1-c). Source samples were oven-dried
183 at 40°C for ~48 hours and sieved successively to 1 mm and 63 µm.

184 2.2.2 *Sediment core*

185 A 1.62 m depth sediment core was collected with an auger in the floodplain (latitude: 21.6357
186 (South); longitude: 166.1967 (East)) in April 2016 (Figure 1-c). The sediment core was sampled on
187 private land where the owners confirmed the systematic overflow of the river at this location during
188 the largest floods. There was no evidence of bank erosion in the vicinity of the coring site with field
189 observations or aerial imagery taken between 1943 and 2019. The core was cut into sections of
190 variable thickness (n = 32, between 2.5-7 cm, S1 to S32, from top to bottom) to best coincide with
191 the contrasted sediment layers characterized by different colours and textures. Sediment core
192 samples were also oven-dried at 40°C for ~48 hours and sieved successively to 1mm and 63 µm.

193 2.3. Sediment analyses

194 2.3.1 *Particle size*

195 Laser particle size measurements were conducted on the <1 mm fraction both source and
196 sediment core samples (n= 57) with a Hydro 2000G particle sizer (Malvern) at the Laboratoire
197 Environnement Ville et Société (EVS, Lyon, France). Destruction of the sediment organic matter was
198 performed with a H₂O₂ solution. Laser particle-size measurements were carried out using water as a
199 dispersant. The degree of obscuration ranged between 12–30%. To control the dispersion of
200 samples, stirrer and pump settings were fixed to 750 rpm and 1875 rpm, respectively.

201

202 2.3.2 *Spectrocolorimetry*

203 Spectrocolorimetric measurements were conducted at the Institut des Géosciences de
204 l'Environnement (IGE, Grenoble, France). The <63µm fraction of all sediment samples were stored in
205 60 mL polystyrene tubes and analyzed by a portable diffuse reflectance spectrophotometer (Konica
206 Minolta 2600d). Sample quantity varied between 0.1 g and 4 g. Owing to the rather small measuring
207 area (i.e. 3-mm radius circle), and to take into account the possible heterogeneity within the
208 samples, three measurements were taken on each sample. Spectral reflectance was measured
209 between 360 and 740 nm with a 10-nm resolution. All measurements were taken with the D65
210 standard illuminant, the 10° angle observer and with the specular component excluded. Raw data
211 collected were the spectral reflectance percentage for each of the 39-wavelength classes. From these
212 raw data, three components of various colorimetry models were also derived (Rossel et al., 2006).
213 The standardized tri-stimuli were then converted into CIEL*a*b* cartesian coordinate systems using
214 the equations provided by CIE (1994). The spectrophotometer was calibrated before each set of
215 measurements by making a zero and a white calibration. Control measurements were also taken
216 regularly during and at the end of each set of measurements; the latter consisted of measurements

217 on red, green and yellow panels as well as six contrasting soil samples. Among the colorimetric
218 parameters analyzed, one of them, a^* , was preferentially used to describe the sediment core: when
219 $a^* < 0$, colour of the sediment tends to be green whereas when $a^* > 0$, it tends to be red. First
220 Derivative reflectance of the Visible Spectra(FDVS) was also carried out in order to identify potential
221 differences in the sediment mineralogical composition (Debret et al., 2011).

222 2.3.3 *Elemental geochemistry*

223 The contents of 11 geochemical elements (*i.e.* Mg, Al, Si, K, Ca, Ti, Cr, Mn, Fe, Ni and Zn) were
224 measured by energy dispersive X-ray fluorescence spectrometry (Epsilon 3, Malvern PANalytical)
225 in source samples (n=16) and sediment sections (n=32) at the Laboratoire des Sciences, du Climat et
226 de l'Environnement (LSCE, Gif-sur-Yvette, France). Calibration curves for the geochemical elements of
227 interest have been established based on 16 certified reference samples including Internal Atomic
228 Energy Agency (IAEA) standards. To conduct these analyses, small mass holder (SMH) cells
229 characterized by an air double X-ray Mylar film were prepared with a sample quantity between 0.2
230 and 0.5 g of the previously sieved (<63 μm) material. This analysis consisted of irradiating samples
231 with a primary beam generated by an Rh anode X-ray tube emitting electromagnetic waves between
232 100eV and 1MeV with a maximum power, typical current and voltage fixed to 15 W, 3mA and 50 kV
233 respectively. The associated Si-drift detector had a Be window thickness of 8 μm and recorded the
234 sample spectrum in a 2D optical geometry configuration. X-ray intensities were converted into
235 concentrations using the Epsilon 3 software program through the application of the fundamental
236 parameters method. Correlations between the determined and the standard elemental contents
237 were comprised between 0.90 And 0.99. To validate the calibration, 3 certified standards different
238 from those used for the calibration were analysed, and the associated mean relative error was 17%
239 (SD 5%).

240 2.4. Identification of sediment layers

241 Sediment deposits were identified from visual observations of the colour shades and particle
242 size. In addition, four colorimetric, geochemical and particle size parameters were used to
243 corroborate the visual observations and identify sediment deposits that were not visually observable:
244 a^* , FDVS, Ni content and the silt-sized fraction.

245 2.5. Methodological framework for sediment fingerprinting.

246 Fallout radionuclides (Evrard et al., 2020; Evrard et al., 2015; Wallbrink et al., 1998),
247 geochemical (Collins et al., 1997; Laceby and Olley, 2015) and mineral properties (Klages and Hsieh,
248 1975; Walden et al., 1997) are the most frequently used tracers to quantify sediment source
249 contributions. More recently, alternative sediment fingerprinting techniques have been developed
250 based on the analysis of properties such as sediment color (Legout et al., 2013; Poulenard et al.,
251 2009), Compound-Specific Stable Isotopes – CSSI signatures (Reiffarth et al., 2016) or environmental
252 DNA (Evrard et al., 2019; Ficetola et al., 2018).

253 In New Caledonia, a sediment fingerprinting approach had previously been developed to
254 quantify sediment source contributions over two cyclonic events (Sellier et al., 2020). In this
255 approach, the use of geochemical elements such as K to trace mining versus non-mining tributary
256 contributions is supported by the characteristics of the lithological material where both source types
257 are found. Indeed, peridotite massifs where nickel ores are extracted are depleted in K. On the
258 contrary, volcano-sedimentary formations devoid of mining exploitation sites are naturally enriched
259 in K (Sevin, 2014). As mining sites are concentrated in peridotite massifs, the application of this tracer
260 for identifying the mining basin source contribution is straightforward. However, this technique was
261 only applied to sediment collected after cyclones that occurred in 2015 and 2017, and the mining
262 source contributions estimated based on their analysis cannot be extrapolated to the entire period of
263 mining exploitation that started in the 19th century. A sediment tracing approach was therefore

264 carried out to select the optimal suite of tracers that discriminate mining and non-mining tributary
265 contributions among 11 geochemical properties.

266 2.5.1. Source discrimination

267 The sediment fingerprinting technique relies on the ability of tracers to discriminate between
268 sediment sources while remaining conservative (Collins et al., 1996). A range test was applied to
269 ensure that the sediment core samples plotted within the source range (*i.e.* between minimum and
270 maximum values found in source samples). Thereafter, the Mann-Whitney U-test ($\alpha= 0.05$) selected
271 parameters that significantly discriminated between the two potential sediment sources (*i.e.* mining
272 and non-mining tributaries) for each group of potential tracers. A stepwise discriminant function
273 analysis (DFA) was then used to select the optimal number of potential tracers through the selection
274 of those that collectively maximize the discrimination between both sediment sources for each set of
275 tracers. The optimal tracers for modelling were selected by minimizing the Wilks' lambda values in
276 the backwards mode, with a $p>0.05$ used to select a tracer and $p<0.05$ used to remove a tracer.

277 2.5.2. Distribution modelling

278 A distribution modelling approach was used to model the source contributions from the
279 mining and non-mining tributaries to target sediment core samples (Lacey et al., 2015; Lacey and
280 Olley, 2015). This approach incorporates distributions throughout the entire modelling framework,
281 including the source contribution terms and the target sediment. For this two-source model, it is
282 assumed that sediment samples constitute a discrete mixture of their sources, with the contribution
283 from Source A being x , and the contribution of Source B being $(1-x)$ where:

$$284 \quad Ax + B(1 - x) = C \quad \text{Equation 1}$$

285 Where C is the sediment core sample distribution, A and B are again the two source
286 distributions (*i.e.* mining and non-mining tributaries), and x is modelled as a truncated normal

287 distribution ($0 \leq x \leq 1$) with a mixture mean (μ_m) and standard deviation (σ_m) (Laceby and Olley,
288 2015).

289 The model is solved by minimizing the median difference between the distributions of both
290 sides of Equation 1 (C and $Ax + B(1 - x)$) with the Optquest algorithm in Oracle's Crystal Ball software
291 with a randomly generated mixture mean (μ_m) and standard deviation (σ_m). Source contributions (x)
292 were determined by simulating all distributions in Equation 1 with 2500 Latin Hypercube samples
293 from 500 bins and varying the mixture mean (μ_m) and standard deviation (σ_m). This simulation and
294 solving procedure repeated 2500 times and the median proportional source contribution from these
295 2500 additional simulations being reported as the source contribution to the target sediment.
296 Uncertainty was determined through summing three quantifiable model uncertainties: (1) the
297 median absolute deviation of the individual source median contribution for the additional 2500
298 simulations; (2) the modelled standard deviation; and (3) the median absolute deviation of this
299 modelled standard deviation for the 2500 model additional simulations (Laceby et al., 2015).

300 2.6. Source contribution variability and definition of sediment core units

301 A statistical analysis based on cumulative deviations, developed by Craddock (1979) and
302 Buishand (1982), was applied to identify the main changes in mining source contributions within the
303 sediment core. This is more specifically a test of data homogeneity based on rescaled adjusted partial
304 sums S_{k}^{**} :

$$305 S_{k}^{**} = \frac{\sum_{i=1}^{n=32} (Y_i - Y)}{\sigma}$$

306 Where Y_i is the mining source contribution of the core section i (S_i), Y is the mean mining
307 source contribution over the entire sediment core and σ is the standard deviation associated with Y .
308 The confidence bound was set at 99 % which determined the critical value of 1.46 and a threshold
309 value (*i.e.* $1.46 \times \sqrt{n}$) of 8.46. If S_{k}^{**} is lower than the threshold value (*i.e.* 8.46) then mining source

310 contributions are considered to remain homogenous over the entire sediment core section. On the
311 contrary, if S_k^{**} exceeds the threshold value then mining source contributions are considered
312 heterogeneous and consequently, several sediment units can be defined.

313 2.7. Establishing core chronology

314 2.7.1. ^{137}Cs and $^{210}\text{Pb}_{\text{xs}}$ dating models

315 Fallout radionuclides (*i.e.* ^{137}Cs , $^{210}\text{Pb}_{\text{xs}}$) were measured in source (n=24) and sediment core
316 (n=32) samples by gamma spectrometry using coaxial HPGe detectors (Canberra/Ortec). For the
317 measurement of radionuclides in each sample, approximately 1 g of material packed in 15 mL
318 polyethylene containers was analyzed. Indeed, the potential application of fallout radionuclides (*i.e.*
319 ^{137}Cs and $^{210}\text{Pb}_{\text{xs}}$) for dating overbank sedimentation on the floodplain was demonstrated in
320 numerous studies (Collins et al., 1997; Foucher et al., 2014; Owens et al., 1999). The depth at which
321 the ^{137}Cs peak concentration occurs can be related to the period of peak fallout (*i.e.* in 1965 in the
322 Southern Hemisphere (Turney et al., 2018)) while He and Walling (1996) showed that the exponential
323 radioactive decay of $^{210}\text{Pb}_{\text{xs}}$ with depth may be used to calculate the mean sedimentation rates over
324 the last 100 years.

325 2.7.2. Rainfall-runoff analysis

326 Eight rainfall stations managed by Météo France (*i.e.* Thio Plateau, Thio village, Camps des
327 Sapins) and by Direction des Affaires Vétérinaires Alimentaires et Rurales (DAVAR) (*i.e.* Kouaré, Bel-
328 Air, Ningua, Kuenthio, Mont Do) have daily precipitation records since 1952. Daily discharge has been
329 monitored at a river gauging station located on the main stem of the Thio River section (at Saint-
330 Michel) since 1981 by the DAVAR (Figure 1-b). Overbank flood events at the coring site were
331 assessed by Schneider et al. (2018) based on based on 1) the estimation of bankfull discharge (*i.e.*
332 $790 \text{ m}^3 \text{ s}^{-1}$) at the coring location according to the discharge time serie and recent flood observations
333 (*i.e.* no overflow observed during cyclone Edna in 2014 ($788 \text{ m}^3 \text{ s}^{-1}$) versus occurrence of overflow

334 after cyclone Cook in 2017 ($796 \text{ m}^3 \text{ s}^{-1}$); 2) the discharge time-series analysis from the DAVAR
335 database (from 1981 to present) to reconstruct the flood chronology over the last 40 years; and 3) an
336 intensity-duration rainfall statistical analysis (from 1952 to 1980) to extrapolate this reconstruction
337 to the previous period between 1952 and 1981. Schneider et al. (2018) estimated the occurrence of
338 flooding at the coring site and cross-validated these results with a historical database of cyclone and
339 tropical depression events provided by Météo France. Moreover, this flood event chronology was
340 cross-validated with the results of a survey conducted among Thio's inhabitants.

341 **3. Results**

342 **3.1. Stratigraphy**

343 Sediment deposits were clearly identified from 72 to 87 cm depth ($n= 4$, sediment layers: S16,
344 S17, S18, S19) and from 102 to 111.5 cm depth ($n= 2$, S23, S24) (Figure 2-a). The thickness of these
345 deposits varied between 2.5 and 5 cm. Two types of sediment deposits were observed (*i.e.* fine red-
346 orange deposits and brown deposits). Although no other clearly identifiable sediment layers were
347 found in the rest of the sediment core, clear trends in the colour properties were detected. From 0 to
348 72 cm depth, the sediment had a light red-brown colour. A second sediment unit was observed from
349 72 to 111.5 cm depth. It comprised both the sediment deposits mentioned above ($n= 6$, S16, S17,
350 S18, S19, S23 and S24) and a layer of 15 cm of sediment (*i.e.* from 87 to 102 cm depth) where a
351 mixture between fine red-orange and brown sediment was observed. In contrast, the lower part of
352 the sediment core (*i.e.* from 111.5 to 162 cm depth) was dark brown (Figure 2-a).

353 All sediment core sections showed a^* values exceeding 0 (*i.e.* between 7.9 – 16.0), thus
354 indicating a red coloration of the sediment (Figure 2-b). This red coloration was particularly marked
355 from 45 to 93 cm depth with the highest a^* values (*i.e.* between 9.3-16). The remainder of the
356 sediment core (*i.e.* from the top to 45 cm depth and from 93 cm depth to the bottom) is
357 characterized by lower values in a^* between 7.9-13.2. Ni contents comprised between 2.9 and 15.5 g

358 kg^{-1} varied in a similar way as a^* in the sediment core (Figure 2-c). Ni contents were highest (*i.e.*
359 between $3.9 - 15.5 \text{ g kg}^{-1}$) from 45 to 93 cm depth. The lower part of the sediment core (*i.e.* from 93
360 to 160 cm depth) showed the lowest Ni contents, comprised between $2.9 - 6.9 \text{ g kg}^{-1}$. In the upper
361 part of the sediment core (*i.e.* from 0 to 45 cm depth), Ni contents was nearly constant, in the range
362 between $5.9 - 9.2 \text{ g kg}^{-1}$.

363 This red coloration of sediment associated with high Ni contents likely reflects the occurrence
364 of iron oxides (*i.e.* hematite) and oxyhydroxides (*i.e.* goethite), as indicated by Schwertmann and
365 Latham (1986). The FDVS method used to identify the sediment mineralogical composition confirmed
366 this finding by showing that the main differences were found when examining the intensity of
367 spectral signatures of goethite (*i.e.* at 525 nm) and hematite (*i.e.* at 555, 565 and 575 nm) in the
368 sediment core. Figure 2-d showed that the spectral intensities of goethite and hematite suddenly
369 increased from 45 to 93 cm depth. The lower part of the sediment core (*i.e.* from 93 to 160 cm
370 depth) showed low spectral intensities of goethite and hematite compared to those found in the rest
371 of the sediment core. These three parameters showed a similar trend, for instance a modification of
372 sediment composition from 93 cm depth resulting in the appearance of red-coloured, nickel and
373 goethite/hematite-enriched sediment from 93 cm depth to the uppermost sections of the sediment
374 core.

375 3.2. Identification of sediment layers

376 Seven sediment sections (*i.e.* at S7, S13, S16, S18, S20, S24 and S29) were more specifically
377 distinguished with maximum a^* values, maximum Ni contents and maximum spectral intensities of
378 goethite and hematite (Figures 2-b, c, d). Among these seven sediment layers, three (*i.e.* S16, S18 and
379 S24) were visually detected and corresponded to fine red-orange sediment deposits (Figure 2-a). By
380 combining visual observations with geochemical and colorimetric analysis results, ten sediment
381 sections (*i.e.* seven fine red-orange and three brown sediment deposits (*i.e.* S17, S19, S23) were
382 more specifically distinguished from the rest of the sediment core (Figures 2-a, b, c).

383 Among the ten sediment layers identified visually and based on their different colorimetric and
384 geochemical properties, six showed a sudden increase in their silt fraction (*i.e.* 26 %, SD 9 %), the
385 second dominant grain size fraction after the fine sand fraction (61 %, SD 12 %) in the sediment core:
386 S7, S13, S16, S18, S20 and S24 (Figure 2-e, Table 1). These six core sections all corresponded to red-
387 coloured, nickel and goethite/hematite-enriched sediment deposits (Figures 2-a, b, c, d). Conversely,
388 the three brown sediment deposits visually observed in the sediment core were characterized by a
389 minimum silt fraction (Figures 2-a, e). The variations of the silt fraction (*i.e.* extremum) therefore
390 provided a flood layer indicator. As a result, 19 extrema were found in the sediment core (Figure 2-
391 e).

392 3.3. Sediment tracing

393 3.3.1 *Source conservation and discrimination*

394 Particle size analysis confirmed the occurrence of particle size sorting during erosion and
395 sediment transfer processes. Indeed, sediment core sections had a finer grain size composition than
396 both sources (D_{10} , D_{50} and D_{90} ; Table 1). Particle size sorting may be at the origin of the non-
397 conservation of some tracer properties (Lacey et al., 2017). Nevertheless, the previous results
398 obtained in Sellier et al. (2020) demonstrated that 11 geochemical tracers analyzed in the current
399 research were not significantly impacted by particle size effect. According to the range test results, all
400 properties (except Fe) were found to be conservative. Fe contents measured in the sediment core
401 layers were higher than the highest source values (Table 2). Nine of the remaining potential tracers
402 (*i.e.* Al, Ca, Cr, K, Mg, Mn, Ni, Si, Ti) significantly discriminated between the potential sediment
403 sources (*i.e.* Mann-Whitney U-test, $\alpha= 0.05$, p-value <0.1 , Table 3). Among them, only K was selected
404 by the DFA to model sediment source contributions from mining and non-mining sources,
405 respectively, with a Wilk's lambda value of 0.1691. As the Wilk's lambda value is the proportion of
406 the total variance due to the error of the source discrimination, K provided an error of 16.9 % and
407 explained approximately 83.1 % of the differences between the sources (Table 3).

408 3.3.2 Source contribution variability and definition of sediment core units

409 The results of the mixing model showed that mining tributaries dominated the sediment inputs
410 along the entire sediment core with a mean contribution of 74 % (SD 13 %) (Figure 3). According to
411 the test of data homogeneity, three distinct sediment core units were identified. The first sediment
412 unit extended from 111.5 cm depth to the bottom of the sediment core. This phase was
413 characterized by a mean mining tributary contribution of 59 % (SD 5 %). Except one maximum of
414 mining tributary contributions reached in layer S29 (*i.e.* 136-141 cm depth, 68 %) cm depth, a low
415 variability in terms of source contributions was observed along the first unit (*i.e.* SD 5 %).

416 The second unit, from 65 to 111.5 cm depth, corresponded to a transitional phase. Indeed, a
417 sudden increase in mining tributary contributions was observed with a mean contribution of 77 %
418 (SD 14 %) (Figure 3). This phase also showed a higher variability in terms of source contributions (*i.e.*
419 SD 14 %). Overall, the mining tributary contribution reached its maximum along the core (97 %) in
420 S18 (*i.e.* 81-84.5 cm depth). Finally, three other secondary maxima of mining tributary contributions
421 were reached in S16 (*i.e.* 72-77 cm depth, 94 %), S20 (*i.e.* 87-93 cm depth, 82 %) and S24 (*i.e.* 107-
422 111.5 cm depth, 69 %) in this second unit.

423 In the third unit (*i.e.* from 0 to 65 cm depth), mining tributary contributions stagnated with a
424 mean contribution of 80 % (SD 5 %). They therefore remained largely dominant. Although three
425 maxima of mining tributary contributions were reached in S7 (*i.e.* 31-36 cm depth, 83 %), S11 (*i.e.* 50-
426 57 cm depth, 90 %) and S13 (*i.e.* 60-65 cm depth, 92 %), the variability of source contributions
427 remained rather low (*i.e.* SD 5 %).

428 3.4. Core dating

429 Dating of the sequence of sediment deposits could not rely on fallout radionuclide
430 measurements as in most previous studies (Collins et al. (1996) and Foucher et al. (2014)). ¹³⁷Cs
431 activities were lower than the detection limits in 24 sediment sections. The remaining samples

432 showed ^{137}Cs activities ($<1.7 \text{ Bq kg}^{-1}$) were too low to be used for dating because of the absence of a
433 peak or visible trend in the core profile. Similar results were observed for $^{210}\text{Pb}_{\text{xs}}$ activities with results
434 below the detection limits in 19 core sections and activities between $3\text{-}38 \text{ Bq kg}^{-1}$ in the remainder of
435 the core, without any visible trend with core depth (data not shown).

436 Therefore, only a relative deposition chronology based on the correspondence between the
437 floods identified by the rainfall-runoff analysis carried out by Schneider et al. (2018) and the
438 successive layers found in the sediment core was conducted. The rainfall-runoff analysis carried out
439 by Schneider et al. (2018) led to the identification of 14 overflowing flood events that occurred at the
440 coring site since 1952 to 2017 (Table 4). As the core was collected in 2016, 13 flood events were
441 estimated to have deposited sediment in the floodplain at this location since 1950. This analysis is
442 fraught with uncertainty since it is based on extrapolated data (*i.e.* no discharge time-series over the
443 period 1952-1980, only rainfall information, testimonies of local inhabitants and Météo France
444 archives) and recent flood observations to define bankfull discharge ($790 \text{ m}^3 \text{ s}^{-1}$).

445 According to the rainfall-runoff analysis and Thio's inhabitants testimonies, four flood events
446 occurred between 2009 and 2015 (floods 1 to 4; Table 4), which would locate the year 2009 in layer
447 S7 (*i.e.* 31 - 36 cm depth). Dry sediment deposits with a thickness comprised between 4 and 6 cm
448 have been observed during 2015 and 2017 flood events at the coring location. Moreover, the
449 inhabitant mentioned in particular one of 2013 flood events (*i.e.* July 1st, exceptional event because it
450 did not occur during the cyclone season) as having contributed a mud thickness of *ca.* 20 cm.
451 Estimating the deposition of 7 cm-thick sediment layers during individual flood events or a mean
452 deposition rate of 2 cm yr^{-1} appears therefore to be relevant (Figure 4).

453 The limit between units 2 and 3 (*i.e.* at 65 cm depth) was estimated to occur in 1990 according
454 to the four next overbank floods (Table 4; floods 5 to 8, Figure 4). Among the 13 flood events
455 identified (Table 4) by the rainfall-runoff analysis, Cyclone Alison (1975) triggered massive mining
456 waste transfers through the river systems (Garcin et al., 2017; Gosset, 2016). Based here again only

457 on the relative deposition chronology, this tipping point should correspond to S15 (*i.e.* from 69 to 72
458 cm depth), in unit 2 (Figure 4). This hypothesis remains consistent because it leads this exceptional
459 event to be comprised in unit 2 which is also a transition phase and where a significant increase in
460 the contribution of mining tributaries was observed. However, all the results originating from the
461 sediment tracing approach, colorimetric and geochemical analyses converged and indicated that this
462 flood likely corresponds to S18 (*i.e.* from 81 to 84.5 cm depth), also located in unit 2. Indeed, the
463 maximum of mining tributary contributions was reached in this sediment section (S18, 97 %, Figure
464 4). S18 is also characterized by the stronger red coloration of sediment (*i.e.* highest a^* values),
465 highest Ni contents and highest spectral intensity of iron oxides and oxy-hydroxides (Figures 3-b, c,
466 d). It is very likely that several floods have not been recorded in this study due to uncertainties in the
467 analyses (*i.e.* extrapolated data, parameter estimation based on recent flood event observations,
468 core cutting), thus explaining this light temporal shift. Nevertheless, the location of this flood in the
469 transition phase (*i.e.* unit 2) suggests that the relative deposition chronology is consistent. Moreover,
470 we obtained a 1957 deposition year for S22 (*i.e.* from 97 to 102 cm depth) which is again consistent
471 with the assumption that the 1950s sediment deposits would correspond to those layers located
472 below 111.5 cm depth (*i.e.* a change between units 1 and 2) (Figure 4).

473 **4. Discussion**

474 4.1. Sediment sources variability and the mining history of the Thio River 475 catchment (1880 – 2016)

476 The sediment core has been divided into three main units in terms of sediment source
477 contributions (Figure 4): (1) unit 1 (before ~1950), the contributions of the mining tributaries are
478 high, and remain relatively constant with time: 59 % (SD 5 %); (2) unit 2 (~ 1950 – 1990)
479 demonstrates a high variability and overall a rapid increase of these latter (*i.e.* mean of 77 %, SD 14
480 %); (3) unit 3 (~1990 – 2016), the mining tributary contributions are 20 % higher than in unit 1 with

481 low variations (*i.e.* 80 %, SD 5 %). These sediment deposit units can now be compared with the
482 mining history of New Caledonia in general, and the Thio River catchment in particular.

483 *4.1.1. Period 1, from 1880 to 1950: the manual handling mining without any*
484 *environmental legislation*

485 The first period (*i.e.* 1880s-1950s) corresponds to the beginning of mining activities in New
486 Caledonia. As shown in Figure 5, nickel ore production by manual handling mining was very low (*i.e.*
487 around mean 30 000 tons per year) during this period. The recent discovery of nickel ores in 1864
488 and the associated need to find suitable smelting processes as well as the employed extraction
489 process (*i.e.* manual extraction) may explain this low nickel ore production. Furthermore, at this time,
490 mining colonial companies focused mainly on the extraction of the most nickel-rich ores (*i.e.*
491 saprolites), which delineated the extent of mining sites.

492 This period would correspond to unit 1 (*i.e.* from 111.5 cm depth to the bottom) observed
493 within the sediment core. Despite the limited mining development and the manual extraction, the
494 contribution of mining tributaries was high (*i.e.* 59 %, SD 5 %) (Figures 4 and 5). Accordingly, this past
495 activity left a strong imprint in the landscape and has driven and probably still significantly drives the
496 exports of suspended sediment to the delta and the lagoon. Mining contributions were expected to
497 dominate because peridotite massifs, once exploited, represent the most erosion-sensitive structures
498 (Dumas, 2010). It is only with the increasing scarcity of saprolite ores that the mining industries
499 began to change their methods of extraction. The 1920s marked open-cast nickel mining
500 implementation to facilitate extraction of saprolite ores located at depth (Figure 5).

501 *4.1.2. Periods 2 and 3, from 1950 to 1989: the mechanization of mining activities, from a*
502 *non-regulated extraction to the first environmental regulations*

503 For the second period initiated in 1950s, the mechanization of mining activities began with
504 the excavators, bulldozers and caterpillars left in New Caledonia by the US Army Forces after the

505 Second World War which helped make it possible to increase nickel ore production by a factor of 7.
506 The huge development of mining industry in New Caledonia occurred mainly in the Thio River
507 catchment. In the absence of environmental legislation, the sensitivity of soils to erosion increased
508 extensively in the Thio River catchment during this period, as a consequence to (1) the increasing
509 bare soil surface area (233 ha to 775 ha, factor 13 from ~ 1950 to 1975), (2) the rapid extent of
510 mining prospect roads (44 ha to 613 ha, factor ~ 4 from ~ 1950 to 1975) and (3) the direct dumping
511 of mining waste rocks on the hillslopes (4 to 30 million tons from ~ 1950 to 1975) (Garcin et al.,
512 2017). Indeed, the rapid increase of nickel ore production was accompanied by a sharp increase in
513 mining waste (*i.e.* 2.1 tons per tons of nickel ore mined; Ittis (1992); Figure 5), resulting in an increase
514 of sediment inputs to the New Caledonian rivers (Bird et al., 1984; Ittis, 1992).

515 Cyclone Alison occurred in 1975, after the huge development of an unregulated mining
516 industry and an 8-year period of very low cyclonic activity (Table 4). Among the 13 flood events
517 identified (Table 4), Cyclone Alison (1975) was mentioned repeatedly by the inhabitants of Thio as
518 the event that triggered the first significant problems of overburden and hypersedimentation in the
519 river systems. Garcin et al. (2017) showed that overburden material was stored on the mine foothills
520 prior to 1975 and Cyclone Alison was a major contributor to the dumping of this mining waste into
521 the river systems. Indeed, Cyclone Alison began to connect the sedimentary sources (*e.g.* mining
522 areas, waste dumps, prospecting roads) to the creeks and the Thio delta.

523 Cyclone Alison was concomitant with the establishment of environmental legislation in France
524 and New Caledonia (environmental code, n°75-633/1975) that prohibited the disposal of waste
525 products and the recovery of materials, including the prohibition of direct dumping of mining waste
526 rocks on the hillslopes. The polluter pays principle was then applied for the mining extraction from
527 1975. However, in the case of the Thio River catchment, nearly a century of non-regulated mining
528 activity had already led to severe environmental impacts that an environmental law could hardly
529 regulate, especially during a huge event such as Cyclone Alison.

530 Regarding the sediment core analysis, periods 2 and 3 (1950 – 1989) would correspond to unit
531 2. During these periods, a sudden increase in the contribution of mining tributaries was observed (*i.e.*
532 mean of 77 %, SD 14 %). Moreover, visual observations, colorimetric and geochemical analyses
533 indicated a modification of sediment composition during this phase resulting in the dominance of
534 red-coloured, nickel and goethite/hematite-enriched sediment. This red coloration of sediment
535 described in the studies (Bird et al., 1984; Iltis, 1992) but also observed in the sediment core could
536 correspond to the release of laterites initially stored on the foothills.

537 Moreover, this variability of mining tributary contributions observed in unit 2 could be
538 explained by the gradual transformation of landscapes, for example the opening of new mining
539 concessions, the mining dumps on hillslopes, the exploration of new mining sites requiring the
540 creation of mining roads. The high variability of mining tributary contributions at the outlet could
541 also be attributed to 1) a low hydro-sedimentary connectivity and 2) a spatial variability of rainfall
542 that can locally modify the contributions of sediment sources already identified in Sellier et al.
543 (2020). As the mining activity areas increase, so does the probability that a tropical cyclone or
544 depression will affect one of these areas and connect these different sources to the creeks.

545 4.1.3. *Period 4, from 1989 to present: a period of industrial extraction with an increasing* 546 *concern of environmental issues by policies and mining industries*

547

548 Since 2009, a Mining Code in New Caledonia regulates the present mining activities (*i.e.* post-
549 1975) (Figure 5). Among other points, it focused on water pollution in the Thio River and its lagoon
550 with suspended sediment supplied by mining areas in activity. It requires the implementation of
551 mitigation infrastructures on mining areas and environmental monitoring, for instance the collection
552 of rainwater and suspended sediment on mining areas, the sedimentation of fine particles in

553 retention ponds, the stabilization of hillslopes and the planting of vegetation in abandoned mining
554 areas.

555 The relative deposition chronology made it possible to locate this period within the sediment
556 core. Unit 3 corresponds to period 4 (*i.e.* 1990-2016, with 8 flood events). Mining tributaries
557 dominated the sediment input with a mean contribution of 80 % (SD 5 %) (Figure 4). Since this date,
558 the hydro-sedimentary connectivity is very high. This could explain in particular the low variability of
559 mining tributary contributions in phase 3 (*i.e.* SD 5 %) where finally, present and past mining industry
560 drives most of the soil erosion in the entire catchment.

561 4.2. The tipping point concept applied to the Thio River hydro-sedimentary
562 system

563 Despite the efforts made to reduce the environmental impacts of mining activities since
564 1975, sediment transiting the river system still mainly originates from mining areas. The soil erosion
565 processes occurring in peridotite massifs, once initiated, are difficult to contain. The hydrosystem has
566 experienced a shift to another erosive state during the 1950-1989 period, and the year 1975, with
567 the occurrence of Cyclone Alison, can be considered as a tipping point. The concept of tipping point
568 reflects the idea of a change of state of a system with feedback loops that propel the system on a
569 completely new course (Scheffer et al., 2001). This concept seems to be well adapted to the hydro-
570 sedimentary system of the Thio River catchment.

571 The results derived from the sediment core analysis show that rehabilitation and soil erosion
572 mitigation infrastructures and initiatives did not allow a quick recovery to a pre-1950s state (*i.e.* unit
573 1). A slow trend in the decrease of the mining fraction has been recorded in unit 1 since 2009 (from
574 83 % to 70 %). However, the results obtained based on the analysis of the sediment deposits
575 collected after Cyclone Cook in 2017 in Sellier et al. (2020) show that these management structures
576 are not sufficiently effective during intense tropical cyclones (88 % of mining tributary contribution in

577 2017). This conclusion is consistent with measurements conducted downstream of retention ponds
578 during an extreme event in 2001 in another mining location in New Caledonia (Mathys et al., 2015).
579 Indeed, the impoundments and runoff control infrastructures are effective only for floods with
580 limited return periods, for instance hourly rainfall intensity less than 2 years return-period.

581 However, the control of erosion occurring in active mining tributaries is probably masked by
582 the contribution of the legacy mining sources that remain very active. In 1950, the mining
583 contribution was on average 60 %, for instance three quarters of the present mining tributary
584 contribution. Unfortunately, the sediment tracing approach did not discriminate between legacy and
585 current mining contributions. The destabilization of the hillslopes, the difficulties to reduce the
586 sediment loads during cyclonic events on active mining areas, and the presence of fine sediment in
587 all river compartments are so high that it is very likely that a recovery to a pre-1950s state will not be
588 possible without a strong human intervention, for example re-vegetation, runoff and waste water
589 management, along with hillslope stabilization. Moreover, long-term surveys of mining tributary
590 contributions and sediment loads during each cyclone is required to monitor the evolution of
591 sediment source contributions in the coming years and to assess the efficiency of re-habilitation
592 works.

593 4.3. Comparison with literature studies on legacy sediment

594 Similar cases have been observed in the literature around the world. The term 'legacy sediment' in
595 particular has appeared in the literature since the 1980s and is defined as sediment deposits
596 episodically produced by accelerated erosion associated with intensive anthropogenic activities (*e.g.*
597 agriculture, mining; (James, 2013). In a mining context, Macklin (1985) showed that the
598 intensification of mining activities (*i.e.* 1858-1908) had significantly contributed to sediment inputs in
599 the River Axe catchment (Britain). Indeed, floodplain sedimentation rates were four times higher
600 during this period (*i.e.* 1858-1908) compared to those recorded during the period when mining
601 activity had ceased (*i.e.* after 1908). This legacy sediment can be identified in particular by

602 significantly higher metal contents than those recorded in the pre-mining state in a sediment
603 sequence (Brunskill et al., 2004; Ciszewski et al., 2012; Lecce and Pavlowsky, 2001). However, a gap
604 between a period of increased mining activity and the resulting pollution may also occur several
605 years or even several decades later as observed by Nguyen et al. (2009). Indeed, the authors showed
606 that owing to the absence of environmental legislation to manage mining waste during rapid
607 industrialization periods (*i.e.* 1950s-1960s), high contents of heavy metal were recorded over the
608 period 1975-1990 in a lacustrine core in Hungary. Valette-Silver (1993) compiled surveys about heavy
609 metal contamination associated with the industrialization period (*i.e.* 1960s-1970s) and focused on
610 the effect of implementation of new discharge and emission regulations from 1980s on
611 concentration trends in sediment cores. Results showed that a decrease in heavy metal contents was
612 observed in sediment without returning to the pre-mining state. Indeed, this legacy sediment (*i.e.*
613 mining waste and mining alluvial sediment) is not permanently stored and is therefore likely to be
614 gradually remobilized (Coulthard and Macklin, 2003).

615 4.4. Implications for tracing the impact of mining activities on sediment supply in 616 the South Pacific

617 One objective of the current research was to apply a sediment fingerprinting approach that
618 had previously been used to trace sediment sources over two cyclonic events to a sediment core
619 covering several decades. The geochemical properties used in this approach were shown not to be
620 impacted by particle size effect (Sellier et al., 2020). This study showed that most of them (*i.e.* Mg, Al,
621 Si, K, Ca, Ti, Cr, Mn, Ni, and Zn) remained conservative over time. Among these latter, K offered the
622 best discrimination between sediment sources. It is a lithological tracer that discriminates the
623 sediment contributions of volcano-sedimentary formations that of peridotite massifs where mining
624 sites are located. The results obtained with the K-model showed that mining contributions dominate
625 sediment input are corroborated by the remote sensing study conducted by Garcin et al. (2017). The
626 authors showed in particular that natural erosion of peridotite massifs represented approximately 12

627 % of the sediment production areas in 1954 (*i.e.* beginning of mechanization of mining activity), 5 %
628 in 1976 and 2008. As a result, mining erosion is largely dominant on peridotite massifs (*i.e.* ~95 %).
629 Therefore, the results demonstrated that the contributions of mining tributaries provided the main
630 sediment source in the Thio River catchment. This tracer is therefore just as effective to quantify the
631 contributions of sources associated with recent events (Sellier et al., 2020) as it is over longer periods
632 of time.

633 4.5. Difficulties and limitations of sediment core sampling

634 The deposition rate determined in this study (*i.e.* mean annual deposition rate of 2 cm yr⁻¹)
635 cannot be considered as representative of the sedimentation rate observed across the entire
636 watershed. Lecce and Pavlowsky (2001) and Knox (2006) showed in particular that deposition rates
637 were higher in the floodplain than in upper catchment parts. Furthermore, successive sediment
638 deposits recorded at one location in the floodplain cannot be considered representative of the
639 sediment deposits across the entire deltaic floodplain. Lecce and Pavlowsky (2001) and Knox (2006)
640 insisted on the notion of topography: deposition rates were the highest on the lowest valley floor
641 surfaces. Calhoun and Fletcher III (1999) also showed in a subtropical, heavy rainfall, steep-sided
642 catchment (*i.e.* Hanalei, Kauai; Hawaiï Islands) that distance from the river is also a factor influencing
643 sediment deposition. They observed decreasing sediment deposition rates with increasing distance
644 from the main river channel. The more recent study conducted by Omengo et al. (2016) in a tropical
645 floodplain (*i.e.* Tana River, Kenya) confirmed these results and showed that during flood events,
646 sediment settles rapidly and its transport to remote parts of the floodplain remains very limited.

647 Future studies could be devoted to the analysis of several other sediment profiles coupled with
648 geophysical measurements (*e.g.* LIDAR) at the level of the deltaic floodplain of the Thio River
649 catchment in order to have a better spatial and temporal representativity of the sediment deposits.
650 However, given the location of the sediment core sampled in this study (*i.e.* 10 m from the river, at

651 the mouth of the deltaic floodplain), it is likely that the deposition rate determined is representative
652 of the upper limit of the sedimentation rates that may occur in this floodplain.

653 4.6. Fine and coarse sediment transport: distinct sources and dynamics

654 The observations made in this study only focused on the dynamics of the finest fraction of
655 sediment (i.e. <2 mm). Hence, these results cannot be extrapolated to coarse sediment transport (i.e.
656 >2 mm) as they are characterised by very different sediment sources and dynamics. Both type of
657 sediment are thus implying very different management.

658 Sources of coarse sediment are mainly limited to legacy sediment (i.e. inherited mining waste)
659 and landslides (i.e. triggered on mining or non-mining areas). Because of the sudden decrease of the
660 channel slope, this coarse sediment deposited mainly in the creeks upstream the confluences with
661 the Thio River. On the contrary, the sources of fine sediment are rather much more diffuse at the
662 catchment scale with the erosion of mining sites, mining prospection areas, mining roads, inherited
663 mining waste (mining sources) and erosion of large bare soil surfaces generated by fires, landslides or
664 trampling (non-mining sources). As a consequence, this sediment fraction can easily reach the
665 lagoon, even during low flood magnitude, and deposits all along the drainage network and the delta.

666 Severe aggradation of mining tributaries by coarse sediment occurred from the start of mining
667 activities and it intensified following the partial destocking of inherited mining waste generated by
668 the occurrence of cyclone Alison in 1975. Since this event and following the implementation of
669 mitigation infrastructures on the tributaries, the processes of creek aggradation tend to decrease
670 (Garcin et al., 2018). In contrast, fine sediment transport from mining sources does not show any
671 trend of decrease, despite the rehabilitation efforts and the construction of retention basins to
672 mitigate fine sediment transfer.

673

674 **5. Conclusions and perspectives**

675 The current study has reconstructed the temporal evolution of the sediment source
676 contributions based on the analysis of a sediment core collected in the alluvial plain at the outlet of
677 the Thio River catchment. Mining contributions have dominated and continue to dominate sediment
678 inputs to the river systems (74 %, SD 13 %). The past activity has left strong imprints across the
679 landscape. Despite the limited mining development and the manual extraction, the contribution of
680 mining tributaries during the pre-mechanization period (<1950) was found already very high (*i.e.* 59
681 %, SD 5 %). These contributions sharply increased during the period of mechanization (1950s-1975)
682 (*i.e.* increase of 18 %). The absence of environmental legislations has led to the storage of millions of
683 tons of mining waste on the foothills. It was only during a particularly intense rainfall event (*i.e.*
684 Cyclone Alison in 1975) that this mining waste, accumulated over 25 years, was dumped into the
685 river system. At present, this de-stocking is still ongoing almost 45 years after the triggering event.
686 Despite the entry into force of environmental protection legislation in 1975 that prohibited sediment
687 storage on the foothills and the more recent 2009 mining code that initiated the installation of
688 control measures to limit runoff and erosion on-site (*e.g.* retention ponds, waste water
689 management), mining tributary contributions remained very high (80 %, SD 5 %), raising questions
690 about the effectiveness of the expensive hydraulic infrastructures to deal with the extreme rainfall
691 events.

692 This study thus illustrates the challenging problem of environmental pollution legacy. This
693 issue is critical and raises many questions about the current and future responsibilities of the mining
694 industries since the polluter pays principle would imply that the mining industries should pay for the
695 pollution that they have generated since 1975. However, it remains difficult to quantify
696 unambiguously the fraction of pollution that is attributed to present and past activities. Furthermore,
697 a better distinction between the legacy sediment and the waste mining sediment would also help
698 guiding the implementation of effective mitigation measures. Many mining sites have been

699 abandoned since the 1970s and most of them have not been rehabilitated yet (*e.g.* re-vegetated);
700 they may therefore contribute significantly to the sediment inputs into the river systems. In such a
701 context, the increased mining extraction observed during the last decade should be accompanied by
702 huge efforts of rehabilitation and the installation of runoff mitigation infrastructures in both
703 abandoned and operational mining sites.

704 However, according to the tipping point theory (Scheffer *et al.*, 2001), a return to a pre-mining
705 state is unlikely, even illusory. Indeed mitigating efforts should be significantly greater than the
706 extractive forces that have created this situation during the last 150 years. Mining is, and will remain
707 part of the Thio River catchment's landscape for centuries. This perspective raises tensions between
708 the local population, the mining industries and the government. Local residents are suffering daily
709 from the deleterious impacts of floods, soil and water pollution. They are now waiting for effective
710 solutions to be implemented. A pragmatic approach should be defined in close consultation with the
711 population, the levels of pollution that would be acceptable locally according to local environmental,
712 economic and societal issues. At strategic locations, significant rehabilitation efforts should be made
713 for limiting pollution allowing for the development of alternative economic activities (*e.g.* agriculture,
714 tourism) and the well-being of inhabitants.

715 Since by definition, mineral resources are not renewable, this study finally addresses a major
716 socio-environmental issue for the post-mining period that will come one day. Hydraulic
717 infrastructures that limit runoff and sediment transfer runoff have limited lifetimes. If they are no
718 longer properly managed and maintained, important risks of reservoir failing and landslide can occur
719 with disastrous environmental, economic and social injuries during cyclonic events. Accordingly, in a
720 post-mining perspective, when the mining sites will close, who will control and maintain these
721 mitigation runoff infrastructures (*e.g.* roads drainage, retention and infiltration basins) day after day,
722 who will pay for it, and finally for how long (several decades at least, centuries)?

723 Hence, a thorough understanding of past, present and future sediment dynamics in the
724 mining catchments is required to guide decision-making in these mining environments. A study of the
725 biophysical processes (*e.g.* sediment transfer mechanism and vegetation recovery) and the social
726 processes (representation of mining and its impacts by the different actors) have to be conducted
727 together because all these aspects are strongly interconnected. This sediment tracing study
728 contributes to improve this understanding and could also contribute to provide a method to evaluate
729 the long-term efficiency of solutions for preventing erosion in mining areas. This multi-proxy
730 approach could be extended to other mining catchments of New Caledonia but also to other similar
731 mining catchments around the world (*e.g.* Australia, Brazil, Dominican Republic, Cuba).

732

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940

941 **Figure captions**

942 **Figure 1:** Main lithologies (a), location of the rainfall and river monitoring stations (b), location of the
943 sediment core and sediment samples collected along with tributary source classifications (c)
944 conducted in the Thio River catchment, New Caledonia

945 **Figure 2:** Sediment core description through visual observations (a), profiles of chromatic indicator a*
946 (b), Ni contents (c), FDVS (d) and grain size fractions (e)

947 **Figure 3:** Profile of mining source contributions (between 50-100%) and delineation of sediment core
948 units (*i.e.* test of data homogeneity) within the sediment core

949 **Figure 4:** Dating based on the silt size fraction (cyclonic events referenced by n°, Table 4) and
950 estimations of mining source contributions (between 50-100%) within the sediment core

951 **Figure 5:** Evolution of Ni ore and mining waste tonnage over the period 1900 – 2017 coupled to
952 chronology of cyclonic events over the period 1952-2017, mining activity intensification and
953 environmental legislation implementation

954

955 **Table captions**

956 **Table 1:** Grain size parameters and percentage of grain size fractions (*i.e.* clay, silt, fine, medium and
957 coarse-sized sand) in the sediment sources and the sediment core (<63 μm fraction, mean \pm standard
958 deviation)

959 **Table 2:** Geochemical element contents (mean \pm standard deviation, mean \pm SD; minimum, Min;
960 Maximum, Max) in the sediment sources and the sediment core (<63 μm fraction)

961 **Table 3:** Results of Mann-Whitney U test and the DFA used to identify the optimum geochemical
962 tracer combination to differentiate sediment sources in the sediment core (<63 μm fraction)

963 **Table 4:** Chronology of cyclonic events over the period 1952-2017. The number of flood refers to the
964 probable flood deposit recorded in the sediment core (top-to-down numbering). Cyclone Alyson (in
965 bold) corresponds to a tipping point in the sediment core

Table 1

Sediment size	Mining tributaries (n= 16)	Non-mining tributaries (n= 8)	Core sections (n= 32)
d10 (μm)	52 \pm 38	60 \pm 57	17 \pm 10
d50 (μm)	210 \pm 133	275 \pm 201	173 \pm 218
d90 (μm)	655 \pm 345	742 \pm 488	538 \pm 248
Clay <2 μm (%)	2 \pm 1	3 \pm 3	2 \pm 1
2 μm < Silt < 63 μm (%)	21 \pm 9	30 \pm 29	26 \pm 9
63 μm < Fine –sized sand <500 μm (%)	55 \pm 19	43 \pm 19	61 \pm 12
500 μm < Medium- sized sand < 1000 μm (%)	12 \pm 10	17 \pm 14	4 \pm 2
1000 μm < Coarse- sized sand <2000 μm (%)	10 \pm 20	7 \pm 9	6 \pm 14

Table 2

	Mining tributaries (n= 16)		Non-mining tributaries (n= 8)		Core sections (n= 32)	
	Mean \pm SD	Min-Max	Mean \pm SD	Min-Max	Mean \pm SD	Min-Max
<i>Geochemical elements</i>						
Al ($g\ kg^{-1}$)	21 \pm 21	4 – 71	67 \pm 8	51 – 78	38 \pm 10	19 – 53
Ca ($g\ kg^{-1}$)	3.7 \pm 0.5	0.3 – 15.0	9.3 \pm 5.2	4.5 – 21.1	3.0 \pm 2.8	1.1 – 4.4
Cr ($g\ kg^{-1}$)	7.5 \pm 4.6	1.8 – 17.7	0.7 \pm 1.0	0.05 – 2.9	6.1 \pm 1.4	4.0 – 9.0
Fe ($g\ kg^{-1}$)	144 \pm 70	63 – 256	62 \pm 24	48 – 119	187 \pm 43	120 – 288
K ($g\ kg^{-1}$)	1.6 \pm 2.2	0.1 – 7.0	14.0 \pm 3.7	5.8 – 17.4	4.6 \pm 1.8	1.2 – 7.3
Mg ($g\ kg^{-1}$)	99 \pm 59	16 – 202	16 \pm 13	5 – 37	67 \pm 7	53 – 81
Mn ($g\ kg^{-1}$)	2.5 \pm 1.3	1.2 – 4.8	1.4 \pm 0.6	1.0 – 2.5	2.9 \pm 0.5	1.9 – 3.8
Ni ($g\ kg^{-1}$)	6.6 \pm 5.1	0.6 – 17.2	0.3 \pm 0.3	0.1 – 0.9	7.6 \pm 3.3	2.9 – 15.5
Si ($g\ kg^{-1}$)	178 \pm 42	52 – 230	254 \pm 28	196 – 281	193 \pm 22	154 – 227
Ti ($mg\ kg^{-1}$)	1.4 \pm 2.1	0.02 – 5.9	5.4 \pm 0.8	4.5 – 7.0	2.6 \pm 0.9	0.9 – 4.1
Zn ($mg\ kg^{-1}$)	146 \pm 47	91 – 224	125 \pm 4	118 – 130	172 \pm 31	129 – 262

Table 3

Fingerprinting property	Mann-Whitney U test			DFA- backward mode		Correctly classified samples (%)
	U value	p value	Wilk's Lambda	Variance explained by the variables (%)	Squared Mahalanobis distance	
Al	8	0.000	-	-	-	-
Ca	21	0.007	-	-	-	-
Cr	124	< 0.0001	-	-	-	-
K	2	< 0.0001	0.1691	83.1	20.3	96
Mg	119	0.000	-	-	-	-
Mn	108	0.006	-	-	-	-
Ni	125	< 0.0001	-	-	-	-
Si	6	< 0.0001	-	-	-	-
Ti	13	0.001	-	-	-	-
Zn	68	0.834	-	-	-	-

Figure 1

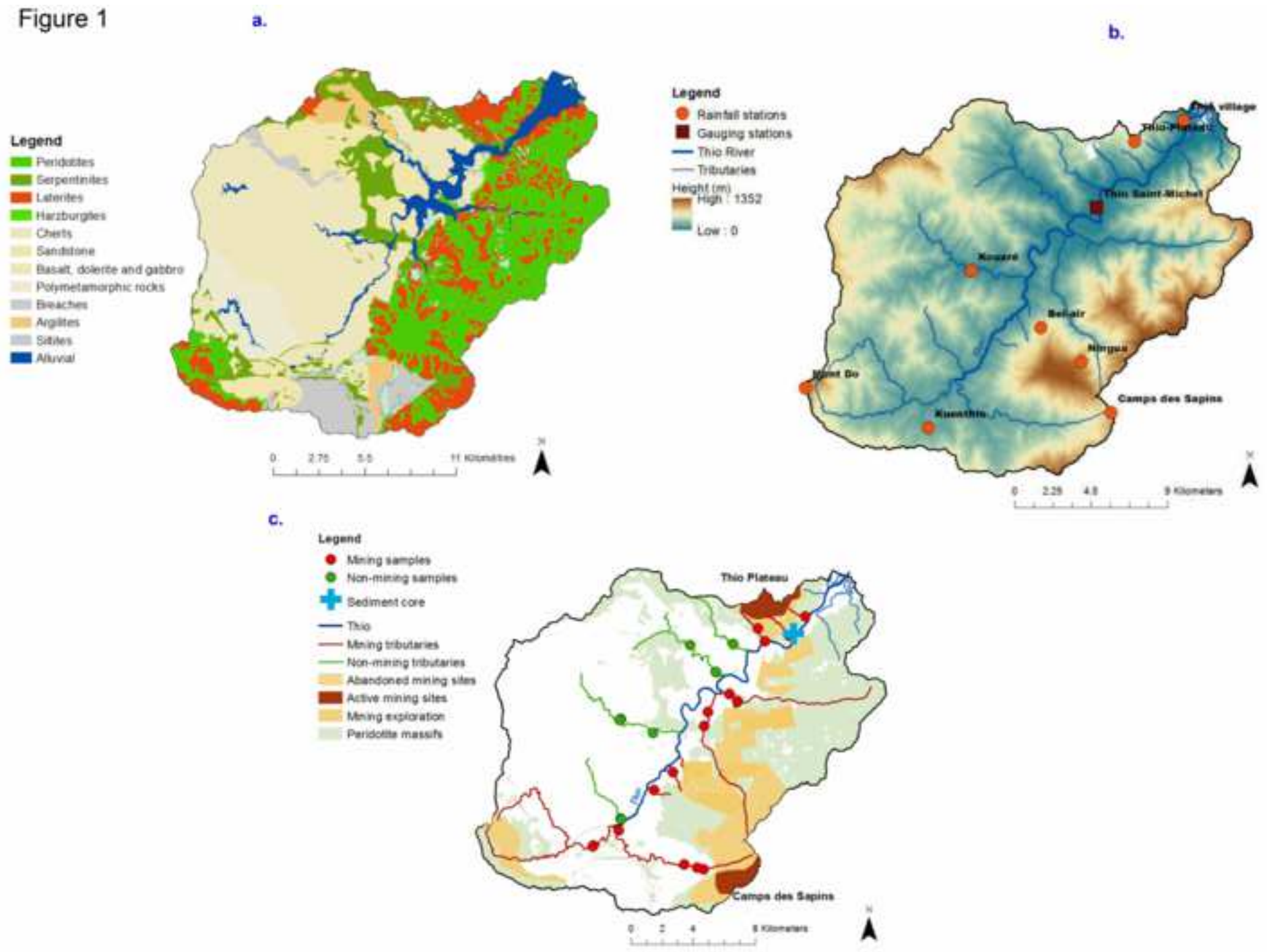


Figure 2

Figure 2

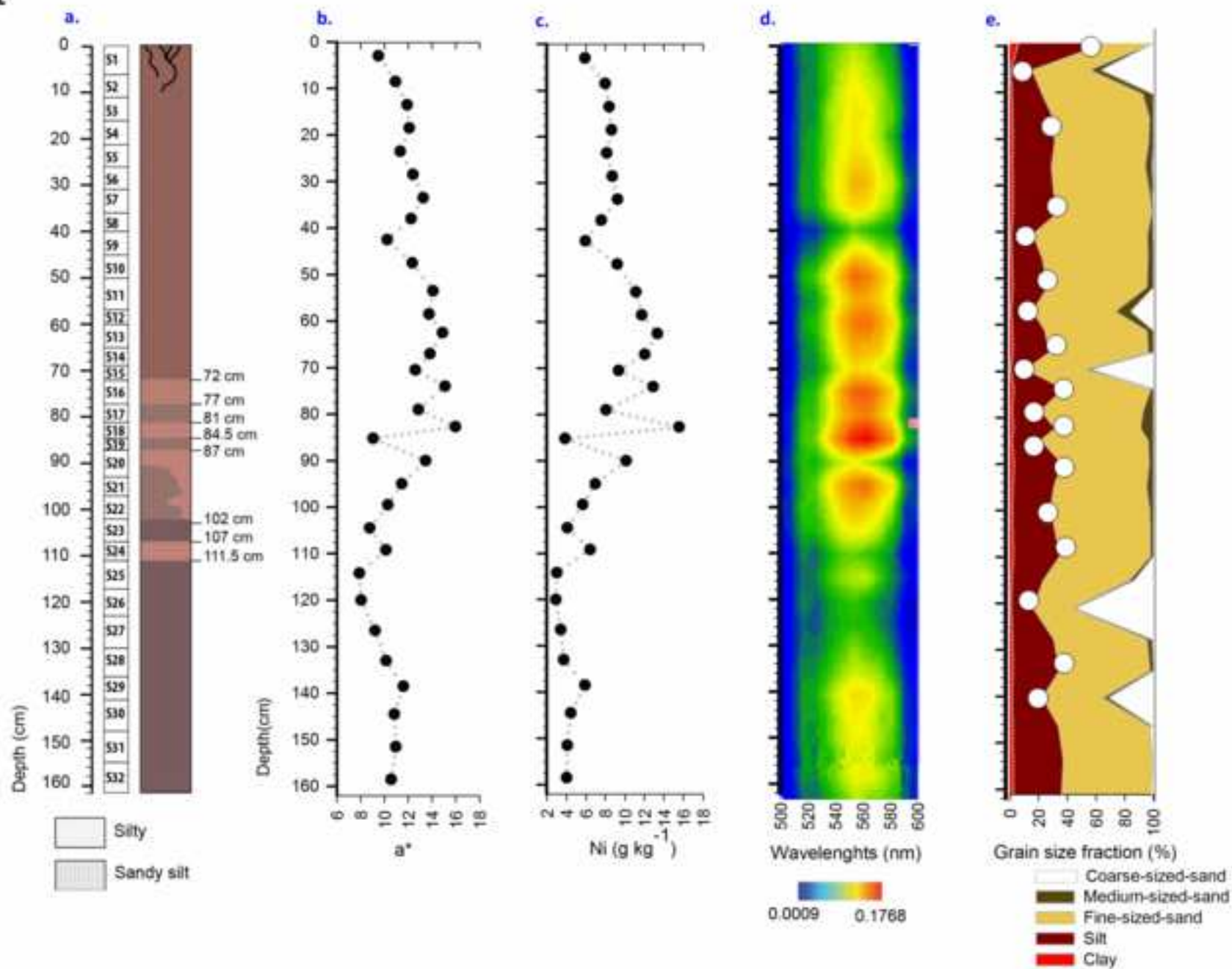


Figure 3

Figure 3

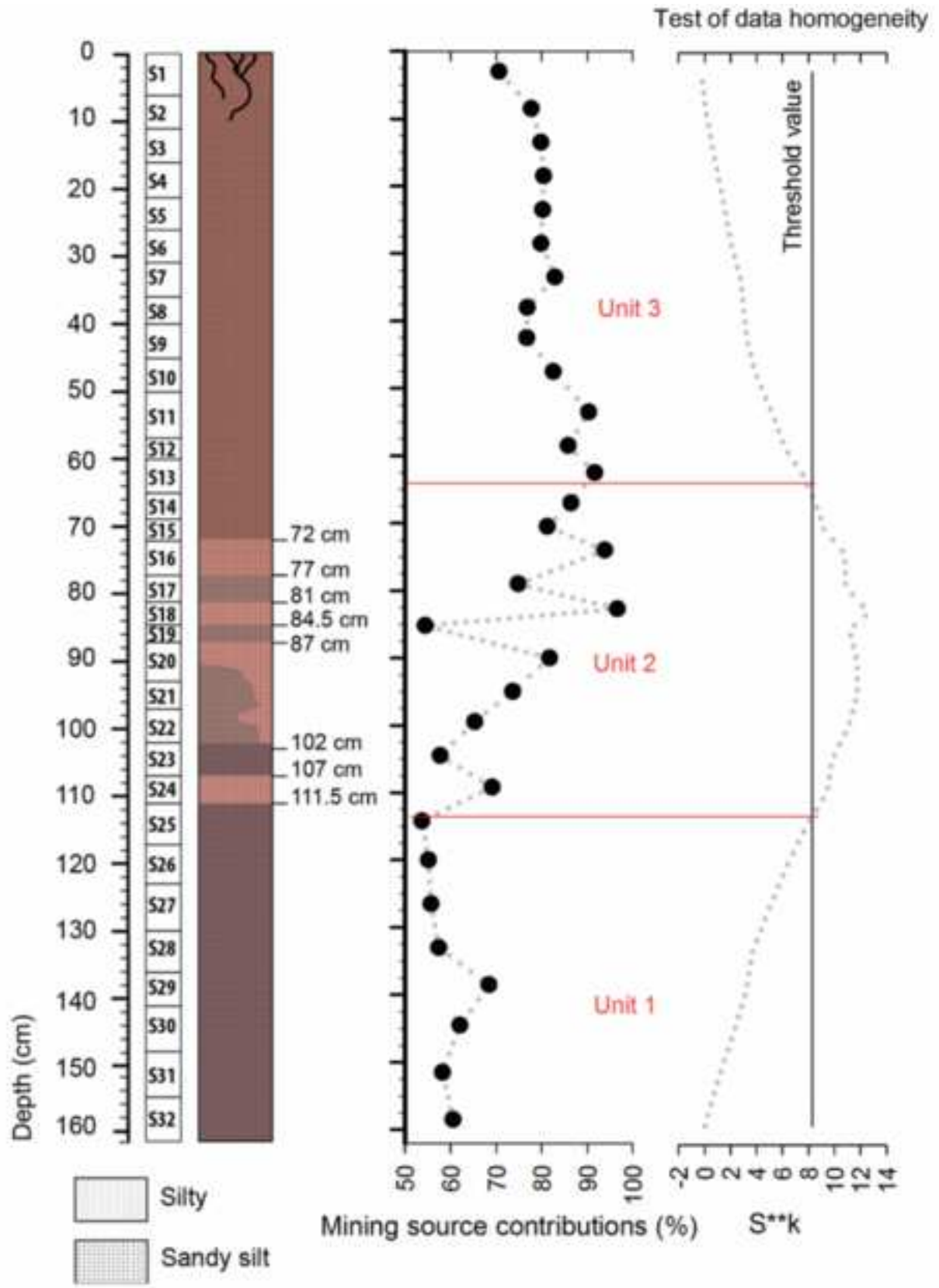


Figure 4

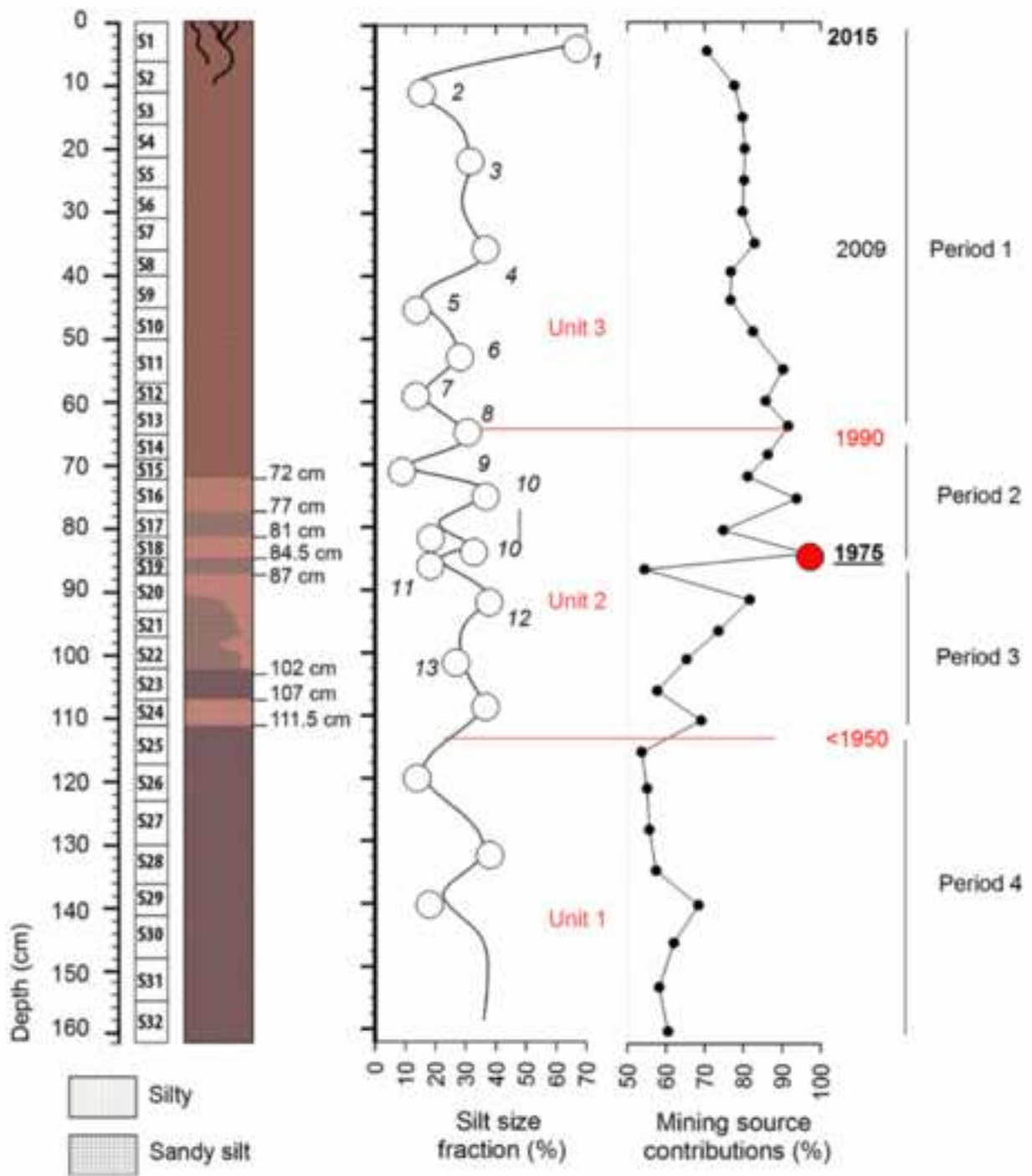


Figure 5

Figure 5

