

1 Sediment flow-paths and associated organic carbon dynamics across a 2 Mediterranean catchment

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12

13 Abstract

14
15 Terrestrial sedimentation buries large amounts of organic carbon (OC) annually,
16 contributing to the terrestrial carbon sink. The temporal significance of this sink will
17 strongly depend on the attributes of the depositional environment, but also on the
18 characteristics of the OC reaching these sites and its stability upon deposition. The goal of
19 this study was to characterise the OC during transport and stored in the depositional
20 settings of a medium sized catchment (111 km²) in SE Spain, to better understand how soil
21 erosion and sediment transport processes determine catchment scale OC redistribution.
22 Total Organic Carbon (TOC), Mineral-Associated Organic Carbon (MOC), Particulate
23 Organic Carbon (POC), Total Nitrogen (N) and particle size distributions were determined
24 for soils (i), suspended sediments (ii) and sediments stored in a variety of sinks such as
25 sediment wedges behind check-dams (iii), channel bars (iv), a small delta in the conjunction
26 of the channel and a reservoir downstream (v) and the reservoir at the outlet of the
27 catchment (vi). The data show that the OC content of sediments was approximately half
28 of that in soils (9.42±9.01 g kg⁻¹ versus 20.45±7.71 g kg⁻¹, respectively) with important
29 variation between sediment deposits. Selectivity of mineral and organic material during
30 transport and deposition increased in a downstream direction. The mineralisation, burial or
31 *in situ* incorporation of OC in deposited sediments depended on their transport processes
32 and on their post-sedimentary conditions. Upstream sediments (alluvial wedges) showed
33 low OC contents because they were partially mobilised by non-selective erosion processes
34 affecting deeper soil layers and with low selectivity of grain sizes (e.g. gully and bank
35 erosion). We hypothesise that the relatively short transport distances, the effective
36 preservation of OC in microaggregates and the burial of sediments in the alluvial wedges
37 gave rise to low OC mineralisation, as is arguably indicated by C:N ratios similar to those in
38 soils. Deposits in middle stream areas (fluvial bars) were enriched in sand, selected upon
39 deposition and had low OC concentrations. Downstream, sediment transported over
40 longer distances was more selected, poorly microaggregated, with a prevalence of silt and
41 clay fractions and MOC pool. Overall, the study shows that OC redistribution in the
42 studied catchment is highly complex, and that the results obtained at finer scales cannot be
43 extrapolated at catchment scale. Selectivity of particles during detachment and transport,
44 and protection of OC during transport and deposition are key for the concentration and
45 quality of OC found at different depositional settings. Hence, eco-geomorphological
46 processes during the different phases of the erosion cycle have important consequences for
47 the temporal stability and preservation of the buried OC and in turn for the OC budget.
48

49 1. Introduction

50
51 Terrestrial ecosystems have captured up to 28% (2.6±0.8 Pg y⁻¹) of the CO₂ emitted
52 annually over the last decade (Le Quéré et al. 2013). Among the processes involved in this

53 terrestrial carbon (C) sink, terrestrial sedimentation of eroded soil and replacement of soil
54 organic carbon at eroding sites have been regarded as active components (Stallard, 1998)
55 (Harden et al., 1999; Van Oost et al., 2007) The magnitude of its contribution to the sink
56 has been estimated by some to be between 0.6 and 1.5 Pg of C annually (Stallard, 1998;
57 Aufdenkampe et al., 2011) through the burial of large quantities of laterally transported
58 organic carbon (OC). The significance of this contribution, however, will depend on the
59 long-term preservation of the buried OC, an issue that remains under debate (Van Oost et
60 al., 2012 and references therein). The fate of the redistributed OC will ultimately depend
61 on the mechanisms of its physical and chemical protection against decomposition, its
62 turnover rates and the conditions under which the OC is stored in sedimentary settings
63 (Van Hemelryck et al., 2011; Berhe and Kleber, 2013).

64 The study of the temporal evolution of buried OC at depositional sites can be approached
65 from different and complementary perspectives. It has been observed that organic matter
66 exported from rivers into the sea is not necessarily identical to the organic matter of the
67 plants and soils upstream in the river catchments (Raymond et al., 2001). This indicates that
68 tracing sediment from source areas and the processes taking place during transport and
69 deposition of eroded OC can also provide information on the quality and dynamics of the
70 eroded OC (Nadeu et al., 2012 and refs therein). Actually, more than 90% of the sediment
71 generated annually in uplands is not exported from catchments (Trimble, 1983; Meade et
72 al., 1990; Walling and Fang, 2003) but remains in transitory depositional sites such as lakes
73 and reservoirs, colluvial deposits at the bases of hillslopes, alluvium in floodplains and
74 channel bars (Meade et al., 1990). In fact, flood plains are expected to represent a key
75 storage site for OC within the catchment C balance, and increasingly this function is
76 fulfilled by reservoirs (Verstraeten et al., 2006; Wissler et al., 2013; Ran et al., 2014).
77 Although large efforts are being made to understand the flowpaths of OC at the
78 catchment scale, most of the abovementioned C sinks remain unquantified. Recently Ran
79 et al. (2014) have estimated an OC budget for the Yellow River, concluding that over a
80 period of 60 years, 49.5 % of the OC was buried in different sinks within the river system,
81 27 % was mineralized during the erosion and transport phases and 23.5% was delivered
82 into the ocean. However, there is still large uncertainty over the stability and residence
83 times of OC in many of these sinks, which are affected greatly by geomorphological and
84 hydrological dynamics (Hoffmann et al., 2013). Thus, characterising the OC at these
85 transitory settings and acquiring knowledge on the processes and factors that influence OC
86 stability at these sites contribute to the assessment of the significance of terrestrial
87 deposition in the C cycle.

88 Understanding how OC moves along with sediments through the different phases and
89 types of erosion and transport processes is crucial to explain partially the large variation in
90 C contents found in depositional sites. Along these lines, progressively the geomorphic
91 factors that control the redistribution of OC within watersheds are being defined (Berhe
92 and Kleber, 2013; Evans et al., 2013; Hoffmann et al., 2013; Nadeu et al., 2011; Nadeu et
93 al., 2015). The study of OC transport from erosion sites to depositional settings implies the
94 consideration of a large number of factors and processes taking place. Several studies have
95 described the impact of variation of transport and deposition of different OC size-
96 fractions and the role of OC mineralisation, as well as the breakdown of soil aggregates or
97 re-aggregation at depositional sites (Wang et al., 2010; Van Hemelryck et al., 2011;
98 Martínez-Mena et al., 2012) and the contribution of new OC formation from vegetation at
99 depositional settings. Altogether, these factors are considered responsible for the
100 transformations undergone by OC from source to sink. Yet, comprehensive studies of
101 source to sink processes are, to the best of our knowledge, lacking.

102 The objective of this study is to characterise the OC in transit and at a range of
103 depositional settings in a medium size catchment and to associate our observations with

104 the catchment sediment dynamics. We aimed to: (i) characterise the OC concentrations in
105 the main sedimentary deposits along the catchment's drainage system; (ii) assess the main
106 processes involved in sediment redistribution; (iii) establish links between these processes
107 and the OC concentration and quality.

108 2. Study area

109 The study area is located in the headwaters of the Segura catchment (Murcia, SE Spain),
110 which drains to the Taibilla reservoir (Turrilla catchment) and is formed by three adjacent
111 subcatchments (Rogativa, Arroyo Blanco and Arroyo Tercero) covering a total area of
112 $\sim 111 \text{ km}^2$ (Figure 1). The Taibilla reservoir, built in 1974, provides water to more than 2
113 million people. The dominant lithology of the catchment consists of marls, limestones,
114 marly limestones and sandstones of the Cretaceous, Oligocene and Miocene. The
115 mountains are mainly constituted by limestones, while the middle and bottom valley
116 sections are dominated by marls (IGME, 1978).

117 The average annual rainfall for the period 1933-2004 was 583 mm and the average annual
118 temperature 13.3°C , at a station located in the centre of the basin at 1200 m above sea
119 level. Snow in the mountains, especially above 1700 m, is not abundant but is frequent in
120 winter. The dominant soils in the area are Lithosols, Regosols and Cambisols (IUSS-WRB,
121 2006). They have an average OC concentration in the first 10 cm between 3.2 and 1 %
122 depending on the land use, being the lowest for agricultural use mainly on marl lithology
123 (Nadeu et al., 2014). A previous study at the site showed that OC concentration in soil
124 profiles down to 1 meter located in forest and shrub areas in a subcatchment was $1.5 \pm 1.4\%$
125 and $2.2 \pm 1\%$ in profiles located in forest areas in another Rogativa subcatchment. In both
126 subcatchments average OC concentration in channel sediments down to 80 cm were
127 $1.1 \pm 0\%$ and $1.4 \pm 0.1\%$ respectively (Nadeu et al., 2012). Boix-Fayos et al. (2009) attributed
128 variation of OC concentration in depth in sediment profiles down to 120 cm located in
129 the main channel of 7 subcatchments of Rogativa to changes in the land use pattern of the
130 drainage area over the last decades.

131 The landscape is a mix of dryland farming (mainly barley), plantations of walnuts (*Junglans*
132 *regia* L.), forests and shrublands. The forest is dominated by *Pinus nigra* Arn. subsp.
133 *salzmannii*, although some individuals and masses of *Pinus pinaster* Ait. and *Pinus halepensis*
134 Mill. are located in the lowest parts of the basin. A relevant proportion of the pine forest
135 was planted in the reforestation works associated with dam construction in the 1970s.
136 Nowadays, masses of *Quercus rotundifolia* Lam. are isolated or associated with *P. nigra* subsp.
137 *salzmannii*.

138 The catchment has been affected by important land use changes since the second half of
139 the twentieth century. These changes consisted mainly of a progressive abandonment of
140 the dryland farming activities and an increase of the forest cover. In the 1970s, a network
141 of check-dams was installed. Previous studies highlighted the important impact of land use
142 changes and check dams on the catchment's sedimentary dynamics (Boix-Fayos et al., 2008;
143 Quiñonero-Rubio et al., 2013; Quiñonero-Rubio et al., 2014), causing important
144 morphological changes in the river bed and accelerated bank erosion processes (Boix-Fayos
145 et al., 2007). Land use changes have been estimated to be responsible for about 50% of the
146 reduction in catchment sediment yield (Boix-Fayos et al., 2008) and have had an important
147 impact on the soil carbon stocks of the catchment (Boix-Fayos et al., 2009).

148 The Turrilla catchment shows a dendritic channel pattern. The main channel has an average
149 slope of 7.7° and a total longitude of 22 km. The stream is discontinuous upstream and
150 continuous downstream. Geomorphological characterisations of channel reaches along the
151 main and tributary streams of the Rogativa catchment indicated dominance of non-
152 selective erosion processes affecting deeper soil layers and with no selectivity of grain sizes,
153
154

155 such as gully, bank and river bed erosion (Nadeu et al., 2012), often activated as a
156 consequence of the decrease in sediment input from the adjacent slopes caused by a
157 generalised recovery of the vegetation, following agricultural land abandonment and
158 reforestations (Nadeu et al., 2014a). Furthermore land use and morphological
159 characteristics of the drainage area were identified to be important driving factors
160 determining the concentration and organic carbon yield exported by lateral fluxes in
161 smaller subcatchments of the Rogativa watershed (Nadeu et al., 2015). In general terms,
162 the main channel of Rogativa moved from an aggradation period with large sediment
163 volumes coming from a well-connected agricultural catchment (1950's-1980's), to an
164 incision and degradation phase after afforestation, land abandonment and hydrological
165 control-works (Boix-Fayos et al., 2007). Nowadays the Rogativa catchment is under a
166 transition phase with an armoured main channel and sediments being incorporated in the
167 channel through gullies and bank erosion (Boix-Fayos et al., 2007; Nadeu et al., 2011).

168 169 **3. Methods**

170
171 The field experimental design was based on the main pathways of sediment and soil OC
172 during their transport through the catchment. Therefore, soils and different sedimentary
173 deposits within the catchment were sampled as shown in Figure 2.

174 175 **3.1. Soil data**

176
177 Topsoil (0-10 cm) samples were taken from 109 locations distributed from upstream to
178 downstream in the catchment, representing all land uses and their spatial extent. Of these
179 samples, 20 % were from high density forest soils, 30 % from low density forest soils, 20 %
180 from shrubland soils, 13 % from pasture soils and 20 % from agricultural soils. Disturbed
181 samples were taken for laboratory analyses and undisturbed samples (rings of 100 cm³) for
182 estimating soil bulk density.

183
184 Figure 1 and 2

185 186 **3.2. Sediment data**

187 188 **a) Alluvial wedges (behind check-dams)**

189
190 The sediment deposited behind the network of check-dams installed in the 1970s was used
191 as representative of material mobilised by erosion processes and fluvial transport from the
192 upper catchment areas (Figure 1 and 2). Nineteen (sub) catchments, evenly distributed,
193 were sampled. The sediment wedges deposited behind each check-dam were sampled at the
194 front (close to the check-dam) to a maximum depth of 1.25 m. In addition, 14 of the
195 sediment wedges were sampled also upstream, at the back of the sediment wedge, to a
196 maximum depth of 96 cm. At all points, bulk samples of 100 cm³ were taken at intervals
197 of 7 cm depth until the maximum depth was reached. Moreover, for 14 wedges, replicate
198 samples of the first layers were taken at 7-cm intervals, to 35 cm depth. A total of 537
199 undisturbed samples were collected.

200 201 **b) Fluvial bars**

202
203 Sedimentary fluvial bars located at two different reaches in the Rogativa main stream, 2 km
204 apart in the middle section of the catchment, were sampled. One of the reaches is a
205 permanent-flow reach where four different bars were sampled during three seasons

206 (autumn 2009, winter and spring 2010), and the other is an intermittent reach where four
207 bars were sampled in autumn 2009 and winter 2010 (the same dates as the bars of the
208 permanent-flow reach). A disturbed sample of the first 5 cm of the bar was taken at each
209 sampling period, as no different layers corresponding to different events could be
210 distinguished in these bars.

211 Two more bars, also in an intermittent reach in the middle section of the catchment and in
212 a confluence of a small subcatchment (11 ha, barranco Escalerica 2) and the Rogativa main
213 stream, were sampled. The bars in the stream bed were incised during the last runoff event
214 (winter 2010) that took place a few days before the sampling. The deposited layers
215 corresponding to different runoff events could be identified (Figure 1 and 2) and were
216 sampled to a maximum depth of 30 cm (bar 1, seven layers, average depth of each layer 4.2
217 cm) or 20 cm (bar 2, six layers, average depth of each layer 3.3 cm). These bars represented
218 a mixture of sediments coming from several erosion processes and were considered
219 representative of the type of sediment being transported along the stream bed from
220 further upstream (Nadeu et al., 2011). A total of 46 samples were collected.

221

222 **c) Suspended sediment**

223

224 Two devices for the sampling of suspended sediments were installed in the main stream of
225 the Rogativa channel in October 2010: one was installed in the downstream area, draining a
226 catchment area of 54.4 km², and the other was installed below the confluence with the
227 perennial stream of Arroyo Blanco, draining a total catchment area of 78.1 km².

228 The sampling devices consisted of a column of 6 bottles located at an averaged height
229 difference of 7.5 cm from each other (Figure 1 and 2). At both locations the upper bottle
230 represented bank-full conditions of the incised stream. A total of 69 samples,
231 corresponding to 13 events and collected over a 2.5-year period, were collected.

232

233 **d) Delta sediments**

234

235 Sediments in the confluence of the river channel and the reservoir were sampled. This
236 delta area is characterised by a very gentle slope and point bars formed by the meandering
237 contact between the river and the reservoir. A sampling scheme following the convex
238 depositional areas of the meanders of the river towards the reservoir was implemented: a
239 total of six positions with two replicates and two depths (0-5, 5-10 cm) were sampled,
240 collecting a total of 24 samples.

241

242 **e) Reservoir sediments**

243

244 Sediments in the Taibilla reservoir were sampled in March 2010. Water height in the
245 Taibilla reservoir is highly variable between years and during the year. Samples were taken
246 at a distance of 500 m from the confluence of the main stream of Rogativa and the
247 reservoir, in exposed sediments forming a terrace 20 cm above the water level at the
248 moment of sampling. Sampling was done with a Cobra TT hydraulic hammer to a depth of
249 1 m. The 1-m-deep core was divided at 5-cm intervals to 20 cm depth and then at 10-cm
250 intervals. A total of 23 samples were analysed.

251

252

253 **3.3. Laboratory analysis**

254

255 All soil and sediment samples (from erosion deposits and deposition bars) were air-dried or
256 dried in an oven at a low temperature (<60°) and then sieved at 2 mm. Primary particle size

257 distribution was measured using a combination of wet sieving (particles $>63\ \mu\text{m}$) and laser
258 diffractometry (particles $<63\ \mu\text{m}$) using a Coulter LS, for the sand fraction and the silt and
259 clay fractions, respectively. The organic matter in these samples was oxidised with hydrogen
260 peroxide and chemically dispersed with a mixture of sodium hexametaphosphate and
261 sodium carbonate (anhydrous) for 18–24 h. The fractions obtained were classified as:
262 coarse sand (2000–250 μm), fine sand (250–63 μm), coarse silt (63–20 μm), fine silt (20–2
263 μm) and clay ($<2\ \mu\text{m}$). The effective particle size distribution was measured by introducing
264 air-dried samples into a Coulter LS, without previous physical or chemical dispersion. From
265 these two measurements, micro-aggregation indices were derived.
266 The percentage of micro-aggregated particles of fine sand, coarse silt and fine silt sizes and
267 two aggregation indices that give an indication of the total percentage of micro-aggregated
268 particles were used: IA (Index of Aggregation), as the sum of the differences of the
269 dispersed and non-dispersed material in each size group (Wang et al., 2010), and ASC
270 (Aggregation Silt and Clay) as the difference between dispersed clay and silt and non-
271 dispersed clay and silt (Igwe, 2000).
272 Given that soil carbon pools might have differences in turnover times, it is of high interest
273 to evaluate the behaviour of different carbon pools separately, as this will give us insight in
274 the long term stability of the mobilized carbon by lateral fluxes. For this purpose the OC
275 was divided into physical fractions by wet sieving: particulate organic carbon ($>53\ \mu\text{m}$)
276 (POC) was separated from mineral associated organic carbon ($<53\ \mu\text{m}$) (MOC) after
277 shaking 10 g of air-dried soil sieved at 2 mm with 50 ml of sodium hexametaphosphate for
278 18 h (Cambardella and Elliot, 1992). Fractions were oven-dried at 60°C for water
279 evaporation and the dry material was weighed prior to OC determination. The OC and
280 nitrogen contents were determined by dry combustion in an elemental analyser (FLASH
281 EA 1112 Series Thermo). The total organic carbon (TOC) was assumed to be the sum of
282 the POC and MOC. The MOC accounted for micro-aggregate and intra-aggregate OC in
283 the silt and clay size fractions. Duplicate or triplicate soil samples were used for laboratory
284 analysis.

285
286 Table 1

287 288 **3.4. Statistical analysis**

289
290 Significant differences between averages were tested using the non-parametric Kruskal-
291 Wallis test at $p < 0.05$. Spearman correlations were performed to explore the relationships
292 between the TOC and pools, as well as C:N ratios and the percentages of primary and
293 micro-aggregated soil particles, together with different aggregation indexes, for all cases or
294 each sediment reservoir type separately. All statistical analyses were performed with the
295 software SPSS 19.0 (SPSS, Chicago, IL).

296 297 **4. Results**

298 299 300 **4.1. Primary particle size distribution of soils and sediments**

301
302 The particle size distribution of soils differed from that of most of the sediments (Figure
303 3). The alluvial bars showed, on average, higher sand contents and lower contents in the
304 fine fractions than the soils (Figure 3). This enrichment in coarse fractions in the alluvial
305 bars was accompanied by the lowest TOC (Figure 4). In contrast, the suspended solids and
306 the delta and reservoir sediments had higher percentages of silt and clay than the soils. The

307 high sand contents in the reservoir and delta sediments were similar to the sand content
308 (around 20%) of the wedges. The particle size distributions of the alluvial wedges were
309 more similar to those of the soils of the catchment than to those of the rest of the
310 deposits (Figure 3).

311
312 Figure 3

313 314 **4.2. Microaggregated particles in soils and sediments**

315
316 Based on the microaggregation data, two groups can be distinguished. Soils and sediment
317 wedges with similar IA and ASC values (Table 2) showed a significantly higher
318 microaggregation level than suspended sediment and reservoir sediments. The sediments in
319 alluvial wedges had 10-times more large aggregates (250-63 μm) than the suspended
320 sediment and 6-times more than the reservoir sediments. This was the dominant class in
321 soils and alluvial wedges, while no differences between size classes occurred in suspended
322 and reservoir sediments. The percentages of medium-sized microaggregates (63-20 μm) in
323 the alluvial wedges were around 3- and 7-times greater than in the suspended sediment and
324 reservoir sediments, respectively. No significant differences were found between the
325 percentages of microaggregated particles in the suspended sediment and reservoir
326 sediments, regarding the total microaggregated material and size classes.

327
328 Table 2

329 330 **4.3. Organic carbon and nitrogen in soils and sediments**

331
332 The TOC concentrations of sediments were, on average, around half ($9.42 \pm 9.01 \text{ g kg}^{-1}$) of
333 the average TOC concentrations of surface soils (0-10 cm) ($20.45 \pm 7.71 \text{ g kg}^{-1}$), being
334 higher in alluvial wedges behind dams, in the suspended solids, in the delta and in the
335 reservoir and lower in alluvial bars in the main channel ($4.44 \pm 1.98 \text{ g kg}^{-1}$) (Figure 4).

336 The highest particulate organic carbon (POC) content was observed in sediment samples
337 taken from the alluvial wedges, while suspended solids and reservoir profiles contained the
338 highest contents of mineral associated organic carbon (MOC). The lowest MOC content
339 was found in the bars (Figure 4). Similar POC:MOC ratios were found on soils, wedges and
340 bars (ranging from 0.5 in bars to 0.8 in soils), whereas much lower POC:MOC ratios were
341 found for the delta and reservoir (~ 0.2), reflecting the prevalence of the MOC pool
342 transported downstream (Figure 4 and 5). Furthermore, while C:N ratios of the MOC and
343 POC fractions of soils sampled under different land uses in the catchment did not show
344 significant differences (data not shown, K-W test, $p > 0.05$), most of the sediment deposits
345 (except for alluvial wedges) showed significantly lower C:N ratios than soils (Figure 5). In
346 general a decrease of C:N ratios of sediment deposits along the fluvial path was
347 accompanied by a simultaneous decrease in N, with the exception of suspended load, in
348 which a higher N content was found than in the other deposits (data not shown, K-W test,
349 $p < 0.05$).

350
351 Figure 4 and Figure 5

352 353 **4.4. Correlations between primary and micro-aggregated particles, OC pools and** 354 **C:N ratios**

355
356 The Spearman correlation coefficients showed different patterns in the relationships
357 between the percentages of primary and micro-aggregated particles and the OC pools and

358 C:N ratios across the deposit types (Table 3). The Index of Aggregation (total amount of
359 micro-aggregated particles) was positively correlated with TOC (for all data) and for the
360 reservoir ($p < 0.06$), POC (for all data and alluvial wedges) and C:N ratio (in soils).
361 Furthermore, the percentages of micro-aggregates of 250-63 μm were positively correlated
362 with POC for all data and alluvial wedges and negatively with C:N ratio (for wedges and
363 suspended sediments). Percentages of microaggregates of 63-20 μm were positively
364 correlated to TOC and MOC of all deposits data and to TOC and MOC of alluvial
365 wedges. Furthermore this microaggregate size was positively correlated to C:N of all data,
366 soils and wedges. The smallest microaggregated particles (20-2 μm) were positively
367 correlated to TOC and MOC of the reservoir and to POC of all data. In contrast, the
368 ASC index (micro-aggregated material of silt and clay size fractions) was negatively
369 correlated with the OC pools, only in the cases that not correlation with fine-sized
370 microaggregates (20-2 μm) was found. ASC correlated also negatively with C:N ratios for
371 all data, soils and alluvial wedges. In general, it seemed that the TOC content and its
372 fractions correlated positively with micro-aggregated material (IA). Some microaggregated
373 sizes showed significant positive correlations with OC of some deposits. Larger
374 microaggregated sizes correlated with TOC and POC of alluvial wedges and the finest
375 microaggregated size with TOC and MOC in the reservoir downstream. No consistent
376 correlation patterns were found between the percentages of primary particles and the OC
377 pools and C:N ratio across deposit types. OC (total and/or different pools) was associated
378 positively with the sand fraction and negatively with the clay fraction, for all the data
379 considered together, soils and the reservoir. By contrast, the clay fraction was associated
380 positively with TOC in wedges and suspended sediment, and with MOC in alluvial wedges.
381 Overall, as may be expected, the OC content was positively correlated with the clay fraction
382 in some deposits (e.g., alluvial wedges and suspended sediments), indicating selectivity
383 during detachment and transport. However, OC was also positively correlated with the
384 content of sand particles in the soils and reservoir, probably indicating the entrance of
385 organic material from other processes (e.g. in situ formation of C in lakes (Tranvik et al.,
386 2009)) in the reservoir and OC formation in the upper layers of sediment. Positive
387 correlations of the sand and OC in soils are probably due to the presence of several
388 samples of sandy soils covered by dense forest (25% of dense forest soil samples had 45-
389 70 % sand).

390
391 Table 3

394 5. Discussion

395
396 Hoffman et al. (2013) pointed out that accounting for the non-steady state of C dynamics
397 along flow-paths from hillslopes through river channels and into oceans is crucial to
398 understand the overall C budget. The redistribution of OC through soil erosion and
399 sediment transport is determined by processes that affect the mineral component of
400 sediment (e.g. selectivity and non-selectivity of material during the detachment, transport
401 and deposition phases) and by processes directly affecting the OC fraction within the
402 sediments (mineralisation, fixation, protection, new OC formation). To better understand
403 the role of soil erosion and sediment transport in the overall C budget, based on the data
404 reported in this study, the following paragraphs discuss the importance of spatial scale and
405 particle size selectivity as well as the C dynamics of eroded sediments during detachment,
406 transport and deposition phases.

407 408 5.1. OC redistribution: effect of scale of observation

409

410 All sediments studied within the Turrilla catchment showed, on average, OC
411 concentrations ($9.42 \pm 9.01 \text{ g kg}^{-1}$) less than half those of soils ($20.45 \pm 7.71 \text{ g kg}^{-1}$), indicating
412 depletion of OC in sediment at this scale. This clearly contrasts with experimental data
413 from erosion plots that usually show higher concentrations of OC in sediments than in the
414 original source-soils (Owens et al., 2002; Girmay et al., 2009; Martinez-Mena et al., 2008).
415 These relatively low OC concentrations found in sediments at the catchment scale raise the
416 question: do the erosion and sediment transport processes lead to C losses to the
417 atmosphere, or is there another explanation for this difference in C concentration between
418 soils and sediments?

419 Previous studies have also found a depletion of OC in sediments measured at a catchment
420 scale (Avnimelech and McHenry, 1984; Haregeweyn et al., 2008; Chaplot and Poesen,
421 2012). For a small catchment (22 ha), Fiener et al. (2005) observed that sediment deposited
422 in ponds was depleted in OC and clay relative to the source soils, while the sediment
423 collected at the outlet of the pond was enriched in OC and clay compared to soils. This
424 demonstrates the important changes in OC content that may occur during different
425 transport and deposition phases due to preferential deposition. Similar results were
426 reported by Wang et al. (2010) and Rhoton et al. (2006), who found OC impoverishment in
427 deposited sediments within their study catchments, while the sediment transported in
428 suspension and exported out of the catchment was enriched in OC (1.2-3 times) compared
429 to the OC concentration in the source soils. Ran et al. (2014) attributes also lower values of
430 OC than soils for different sedimentary settings, based on several methods, to estimate the
431 OC carbon budget of the Yellow River. In our case, the suspended sediments as well as the
432 reservoir sediments showed significantly higher contents of MOC compared to the other
433 sediment deposits, while the MOC contents in soils was similar to that in reservoir
434 sediments and slightly higher than in suspended sediments. This was also found by
435 Amegashi et al. (2011) and can be probably attributed partially to new, in-situ formation of
436 OC in the reservoir (Einsele et al., 2001). We suggest that at these larger scales the decrease
437 of OC in sediments compared to soils is due to a combination of factors related to the
438 spatial scale of observation, namely; (i) variety of sediment sources, that dilute the source
439 effect; (ii) interaction of multiple erosion processes, both selective and unselective; (iii) long
440 transport distances that favour continuous remobilisation of sediment facilitating aggregate
441 breakdown and a reduced physical protection of OC; and (iv) the ample time lapse (from
442 hours to several years) that occurred between sediment detachment and its sampling. This
443 contrasts with sediments collected at the plot and hillslope scales, which are collected close
444 to their sources during the detachment and transport phases, and often shortly after the
445 erosion event, with little opportunity for OC mineralization.

446

447

448 **5.2. Mechanisms of OC loss or gain in sediments**

449

450 Among all the factors and processes abovementioned that condition a general decrease of
451 OC in sediments in our studied catchment compared to soils, three of them closely
452 interact: (i) sources of sediment linked to specific erosion processes; (ii) size selectivity; and
453 (iii) processes affecting the organic components of sediments during the erosion and
454 fluvial transport pathway such as burial, mineralisation or new OC formation.

455

456 **5.2.1. Size selectivity and sources of sediment**

457

458 Wang et al. (2010) explained how selectivity of soil material determines the OC
459 concentration during transport. They concluded that there was very little mineralisation of

460 TOC during the erosion process, since they found higher C:N ratios in sediments than in
461 source soils and similar enrichment ratios of clay and C in deposited sediments. Based on
462 multiple indicators our data show evidence for the contrary in the middle and lower
463 catchment deposits (suspended sediments, delta and reservoir): higher clay contents, lower
464 OC and low C:N ratios in sediments than in soils. These findings could indicate C losses by
465 mineralisation during transport over longer distances. However, the interpretation of
466 narrowed C:N ratios as indicator of mineralization must be done cautiously and in
467 combination with other indicators (Wang et al., 2010), given the complexity of N behaviour
468 along the fluvial path (Robertson and Groffman, 2007). This is even more complex in
469 sediments of intermittent rivers (as in our case) because nitrification and denitrification
470 processes are also dependent on the drying and rewetting cycles of sediments (Gómez et
471 al., 2012; Arce et al., 2013). In our study, total TOC and N were lower in all sediment
472 deposits compared to soils, with the exception of suspended load. Increase of N in
473 sediments rich in clay (as suspended load) has been attributed to the presence of
474 ammonium or to the contribution of fresh-water algae (Sánchez-Vidal et al., 2013).
475 The low OC concentration of sediments deposited in the upper catchment may have a
476 different explanation. The sediments in the alluvial wedges showed textural classes and C:N
477 ratios similar to those of the soils, but lower OC concentrations. The low OC
478 concentration may be explained by the fact that the sediments originate from deeper soil
479 layers, mobilised by non-selective erosion processes such as bank, gully and channel
480 erosion, with lower OC concentrations than the surface soil. Geomorphological field
481 assessments in the area indicated that these deeper operating erosion processes are indeed
482 important sources of sediments in the catchment (Boix-Fayos et al., 2007). Yet, sediments
483 derived from deep soil layers not only have lower OC concentration but also different OC
484 pool composition (Nadeu et al. 2011) and different turnover rates (Nadeu et al. 2012) than
485 those derived from topsoil. Differences in OC pools between sediment deposits can also
486 be the result of transport conditions due to higher or lower rainfall intensity (Martínez-
487 Mena et al., 2011; Zhang et al., 2013) or to the discharge. Smith et al. (2013) related high
488 discharge rates with transport of modern C, associated with erosion and the release of
489 organic matter (Sanchez-Vidal et al., 2013), and C from vascular sources (Goñi et al., 2013).
490 Low flows were associated with export of fossil OC (Smith et al., 2013), from biogenic
491 sources dominated by non-vascular plants (Goñi et al., 2013) or from fresh water primary
492 producers (Sanchez-Vidal et al., 2013).
493 A comparison of the particle sizes of the studied sediments and soils of the catchment
494 points to a selection of transported material in a downstream direction. Finer micro-
495 aggregated and single particles were present in the suspended load and in the reservoir. In
496 the soils and in the alluvial wedges behind check-dams, the material was much more
497 heterogeneous, having similar particle size distribution and larger microaggregates,
498 indicating again its transport by non-selective erosion processes and over short distances.

500 **5.2.2. Processes affecting organic carbon dynamics during transport and** 501 **deposition**

502
503 The OC concentration, the variation in C:N ratios and the different associations of OC
504 with textural classes in the studied deposits indicate different processes of loss or gain of
505 OC during the transport along the studied catchment.

506 The C:N ratios showed clearly two groups (Figure 5): lower values in suspended sediments,
507 delta and reservoir sediments, and higher values in soils and alluvial wedges. This suggests a
508 relatively low mineralisation of OC in the sediments of alluvial wedges due to efficient
509 burial or a short transport time, as reported also by Smith et al. (2013) and suggested by
510 Ran et al. (2014). Further downstream in the suspended sediments, delta and reservoir

511 sediments, the lower C:N ratios could indicate mineralisation of OC in sediment
512 transported over longer distances (Figure 5) (Bouchez et al., 2010; Hovius et al., 2011;
513 Raymond and Bauer, 2001). Ran et al. (2014) recently reported a mineralization of 27 %
514 during erosion and transport of sediment and associated OC through the Yellow River
515 catchment. Although, as previously stated, interpretation of C:N ratios for mineralization
516 must be done with caution. Nevertheless, and interestingly, the observed trend in the C:N
517 ratios in soil and among different sediment deposits was consistent with that found in the
518 POC:MOC ratios (Figure 5) which support our statement that the degree of OC
519 mineralisation is increased during transport along the flow-path.

520 Apart from the indications of OC mineralisation, the MOC concentration of suspended
521 sediments was low and very variable (3.8-0.1 %) which can be attributed to the diverse
522 characteristics of the events that mobilise material from different sources (Smith et al.,
523 2013; Goñi et al., 2013; Sanchez-Vidal et al., 2013) or to sediments mobilised by different
524 erosion processes (Nadeu et al., 2011; 2012).

525 Furthermore, the relatively high C content of the reservoir sediments could indicate *in situ*
526 organic matter formation from ecological lake processes stimulating primary production
527 (Einsele, 2001) or allochthonous OC input from the establishment of vegetation and soil
528 formation in the frequently exposed upper sediment layers. Moreover, while other deposits
529 (wedges and suspended sediment) showed the well-known relationship between clay and
530 OC (Rodriguez-Rodriguez, 2004; Rhoton et al., 2006; Martínez-Mena et al., 2008), reservoir
531 sediments presented a correlation between the sand fraction and concentration of OC in
532 the two pools, that could be indicative of *in situ* C formation. Autochthonous input of
533 organic matter in river and lakes can account around 50 % of the total organic matter in
534 aquatic ecosystems of tropical-semiarid and dryland areas (Kunz et al., 2011; Medeiros and
535 Arthington, 2011) (Kunz et al., 2011). The fluctuation in autochthonous organic matter
536 production in riverine ecosystems depends on the input from terrestrial land uses in the
537 drainage area, with thresholds in which terrestrially derived C is replaced by in-stream algal
538 productivity (Hagen et al., 2010). However, it can also be influenced by river
539 hydrodynamics (Cabezas and Comín, 2010; Devesa-Rey and Barral, 2012). In particular, in
540 Mediterranean river ecosystems there is an important seasonal shift between inputs of
541 allochthonous and autochthonous organic matter, with high primary production in spring
542 and allochthonous organic matter inputs in autumn (Romani et al., 2013). Given that C:N
543 ratios of allochthonous organic matter (more recalcitrant and resistant to degradation by
544 microorganisms) tend to be higher than *in situ* produced organic matter (Devesa-Rey and
545 Barral, 2012), shifts between one and the other will change OC decomposition rates and
546 dynamics.

547

548 The combined interpretation of OC pools, textural analysis and C:N ratios in the soils and
549 different sediment deposits indicates that catchment scale C redistribution by lateral fluxes
550 is controlled by both the organic and the mineral nature of sediments:

551 (i) Sediments in upstream depositional areas (alluvial wedges) showed significantly
552 lower C concentrations than soils, but sediment texture and C characteristics
553 were more similar to those in soils than to those in sediments transported
554 further downstream, showing little indication of mineralisation (similar C:N
555 than in soils) and low selectivity of particles (similar primary and aggregated
556 particle size distribution to soils). This is probably related to the non-selective
557 character of dominant erosion processes and to the proximity of sediment
558 sources, giving little time for aggregate breakdown or C mineralisation.

559 (ii) In middle stream areas, preferential deposition of coarse particles can be seen in
560 the channel bars, enriched in the sand fraction and showing the lowest
561 concentrations of all C fractions, among all deposits.

562 (iii) Downstream, the suspended sediment in transit and the sedimentary deposits (delta
563 and reservoir) showed higher contents of fine particles (clay and silt) -
564 accompanied by lower C:N ratios and a slightly higher C concentration, though
565 still lower than in soils. The sand contents of the delta and reservoir deposits
566 indicate also bedload contribution to the deposits downstream. The differences
567 in the C:N ratio combined with other indicators could indicate different degrees
568 of mineralisation of OC along the flow-path, the OC being protected more
569 when associated with large micro-aggregated particles in soils and in the
570 deposits of the upper catchment areas.

572 **5.3. Implications for the fate of eroded OC**

573
574 The results from this study suggest that sediment reaching depositional settings is
575 composed of a heterogeneous mixture of OC particles and different states of
576 decomposition. The role of the source area, sediment transport and post-deposition
577 processes were revealed as crucial to understanding the characteristics of the OC and
578 differences among the analysed deposits and distinct phases of the erosion process.
579 Although mineralisation fluxes were not addressed directly, the decrease in the level of
580 particle aggregation downstream suggests a potential increase in OC decomposition by
581 microorganisms, leading to higher potential mineralisation rates. Distance from source
582 areas, selective transport and deposition of sediments were identified as important factors
583 controlling the characteristics of the OC in sediments and its fate.

585 **6. Conclusions**

586
587 A non-homogeneous redistribution of OC by water flow takes place within catchments,
588 which can be associated with the geomorphological processes and dynamics of sediment
589 transport and deposition. The redistribution of OC in sediments at the catchment scale is
590 controlled by factors affecting their organic component (mineralisation, protection of OC
591 within micro-aggregates and new OC formation in some deposits) and by factors affecting
592 their mineral component (selectivity of sediment sizes during the detachment, transport
593 and deposition phases of erosion, and the type of erosion processes: selective versus non-
594 selective).

595
596 The processes that determine OC concentration at different pools are related also to the
597 different phases of erosion (detachment, transport and deposition): (i) during detachment:
598 size selectivity, type of erosion process and source of material; (ii) during transport: size
599 selectivity, protection of OC in micro-aggregates and transport distances, and (iii) during
600 deposition and in the post-deposition phase: size selectivity, protection of OC from
601 mineralisation by stabilisation of micro-aggregates and burial and new OC formation are
602 important.

603 The OC mobilised in catchments is associated very closely with the sediment dynamics and
604 can have long residence times, linked to the fate of the sediments. In addition, it can be
605 increased by ecological processes and by replacement in eroded areas, converting
606 catchments into relevant sinks for C budgets.

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621

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Table 1. Main location and characteristics of the sampling areas within the catchment

Location of soil and sediments	Group name	Drainage area (ha)	Deposition areas	Profiles /Subcatchments /Events	Maximum depth (cm)	Samples (n)
Soils						
Slopes	Soils	11000	-	109	10	109
Sediments						
Subcatchments in the third and fourth order channels	Wedges	8-146	Sediment wedges behind check-dams	19	125	537
Main channel and a tributary stream	Bars	5000	Channel bars	10	3-30	46
Main channel	Suspended sediment	5000-7800	Suspended load	13	7-42	69
Main channel	Delta	11000	Delta in the conjunction of the main channel and reservoir	6	10	24
Reservoir	Reservoir	32000	Reservoir sediments at the exit of the catchment	2	100	23

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Table 2. Percentages of microaggregated particles in different size classes and microaggregation indices (IA, Wang et al., 2010; ASC, Igwe et al., 2000).

Group name	250-63 μm	63-20 μm	20-2 μm	IA	ASC
Soils (N=16)	24.99 \pm 16.27aA*	3.91 \pm 6.99aB	7.34 \pm 16.06aB	73.91 \pm 21.03a	12.51 \pm 34.91a
Wedges (N=25)	36.49 \pm 17.88aA	9.59 \pm 8.87bB	0	92.18 \pm 23.50a	36.48 \pm 17.92a
Suspended sediment (N=41)	3.60 \pm 9.21bA	3.19 \pm 3.22aA	2.52 \pm 6.40aA	18.22 \pm 12.29b	0.98 \pm 2.26b
Reservoir (N=12)	6.65 \pm 7.87bA	1.46 \pm 2.70aA	2.44 \pm 3.46aA	24.94 \pm 14.30b	-0.41 \pm 9.71b

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*Different lower case letters mean significant differences among sediments and soil groups.
Different capital letters mean significant differences within sediments and soil groups, with regard to micro-aggregated class sizes
(Kruskal-Wallis test, $p < 0.05$)

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Table 3. Spearman correlation coefficients between primary and microaggregated particle indicators and organic carbon pools and C:N ratios.

Groups	Microaggregated particles				Primary particles			
	250-63 µm	63-20 µm	20-2 µm	IA	ASC	Sand	Silt	Clay
TOC					-			
All data		0.361	-	0.329	-	0.701	-0.264	-0.642
Soils	-	-	-	-	-0.512	0.574	-	-
Wedges	-	0.364	-	-	-	-0.494	-	0.497
Suspended sed	-	-	-	0.34 ^a	-	-	-0.480	0.474
Reservoir	-	-	0.667	-	-	0.636	-	-0.713
POC								
All data	0.586	-	0.515	0.584	-	-	-	-0.273
Soils	-	-	-	-	-0.618	-	-	-
Wedges	0.526	-	-	0.650	0.517	-0.720	0.450	-
Suspended sed	-	-	-	-	-	-	-	-
Reservoir	-	-	-	-	-0.674	0.825	-	-0.874
MOC								
All data	-	0.462	-	-	-	-	-	-
Soils	-	-	-	-	-	0.588	-	-
Wedges	-	0.579	-	-	-	-	-	0.724
Suspended sed	-	-	-	-	-	-	-	-
Reservoir	-	-	0.667	-	-	0.629	-	-0.727
C:N								
All data		0.498	0.333	0.274	-0.552	0.414	-0.269	-0.229
Soils		0.517			-0.703	-	-	-
Wedges	-0.559	0.750		-	-0.559	-	-	-
Suspended sed	-0.400	-	0.458	-	-	-	-0.259	-
Reservoir	-	-	-	-	-	-	-	-

* Only significant correlations are shown. Bold correlation indices are for $p < 0.005$, the rest have $p < 0.05$

^a This correlation has $p < 0.06$

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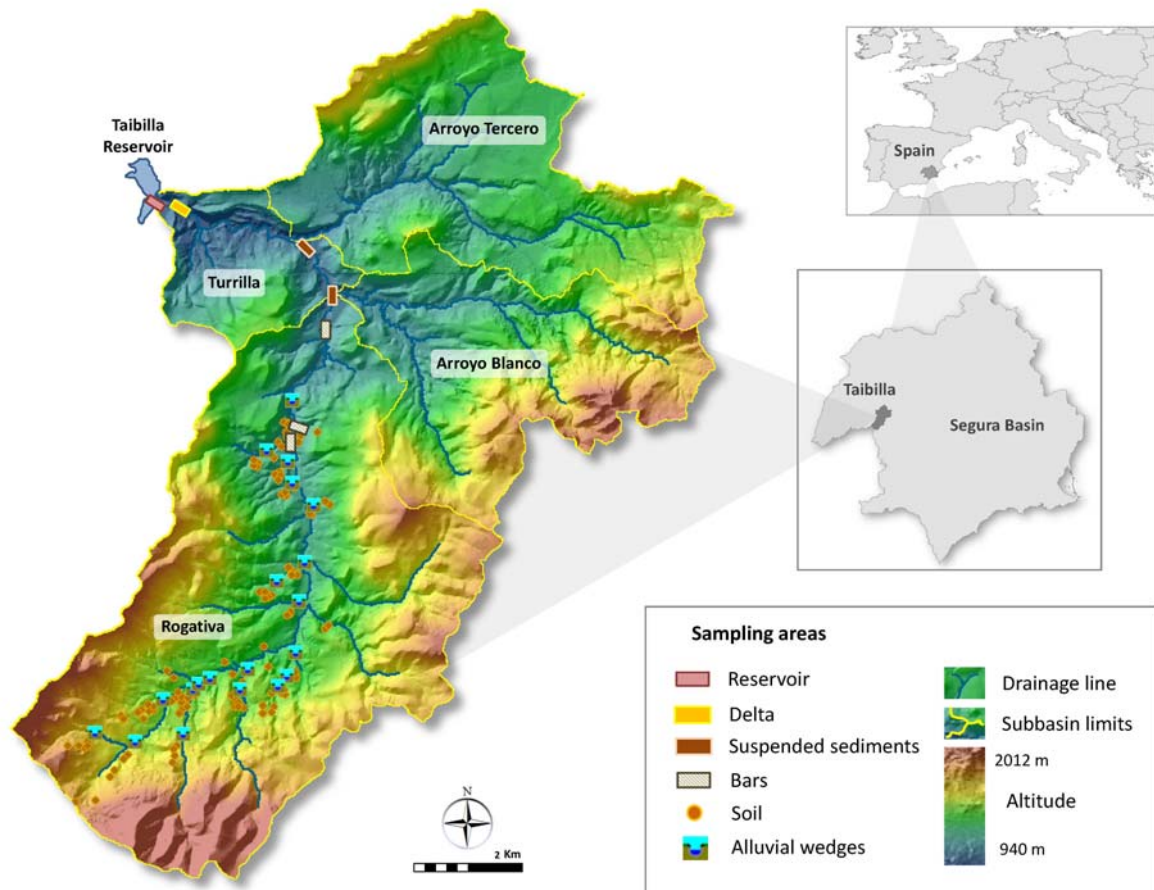
Figure 1. Location of the study area and sampling scheme

Figure 2. Sketch representing the morphological positions selected for sampling of soil and sediments. Soils were sampled all around the catchment in all the different land use classes, alluvial wedges behind check-dams predominantly in the upper and middle parts of the catchment, while alluvial bars and suspended solids were sampled in the middle and downstream areas. Delta and reservoir represent sampling areas downstream.

Figure 3. Median, percentiles (10th, 25th, 75th and 90th) and error of the main textural classes of the different types of sedimentary deposits within the Turrilla catchment. Different letters mean significant differences according to the Kruskal-Wallis test ($p < 0.05$).

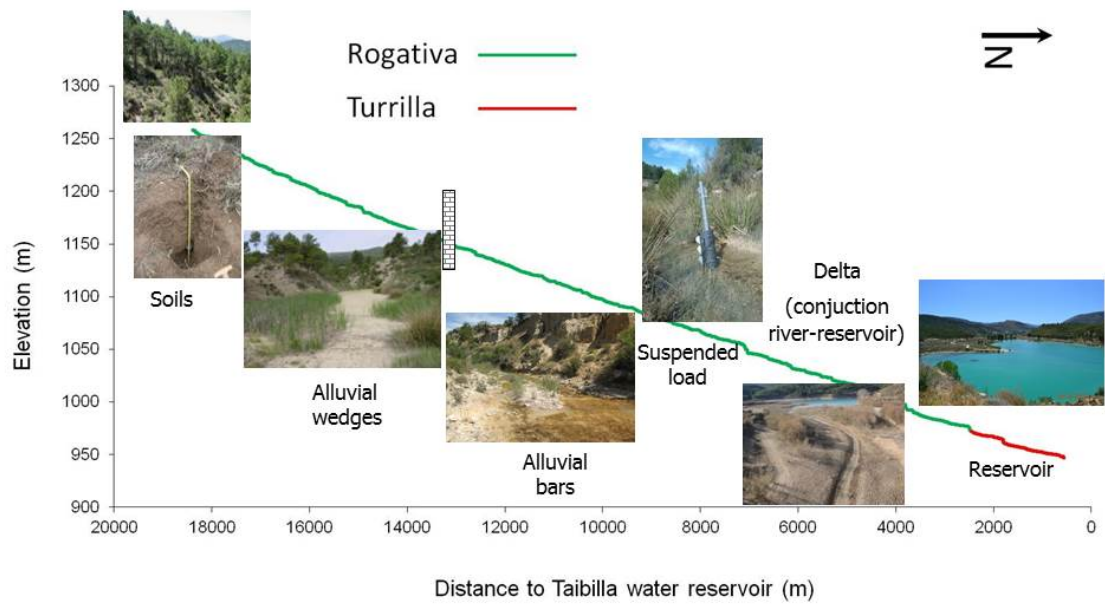
Figure 4. Median, percentiles (10th, 25th, 75th and 90th) and error of the total organic carbon concentration (TOC), particulate organic carbon (POC), mineral associated organic carbon (MOC) of soils and sediments within the Turrilla catchment. Different letters (a-e) indicate significant differences according to the Kruskal-Wallis test ($p < 0.05$). * = suspended solids measured contained only the MOC fraction.

Figure 5. Median, percentiles (10th, 25th, 75th and 90th) and error of C:N ratios and POC:MOC ratios in soils and sediments within the Turrilla catchment. Different letters (a-e) indicate significant differences according to the Kruskal-Wallis test ($p < 0.05$). ND= No data available, * = suspended solids measured contained only the MOC fraction.

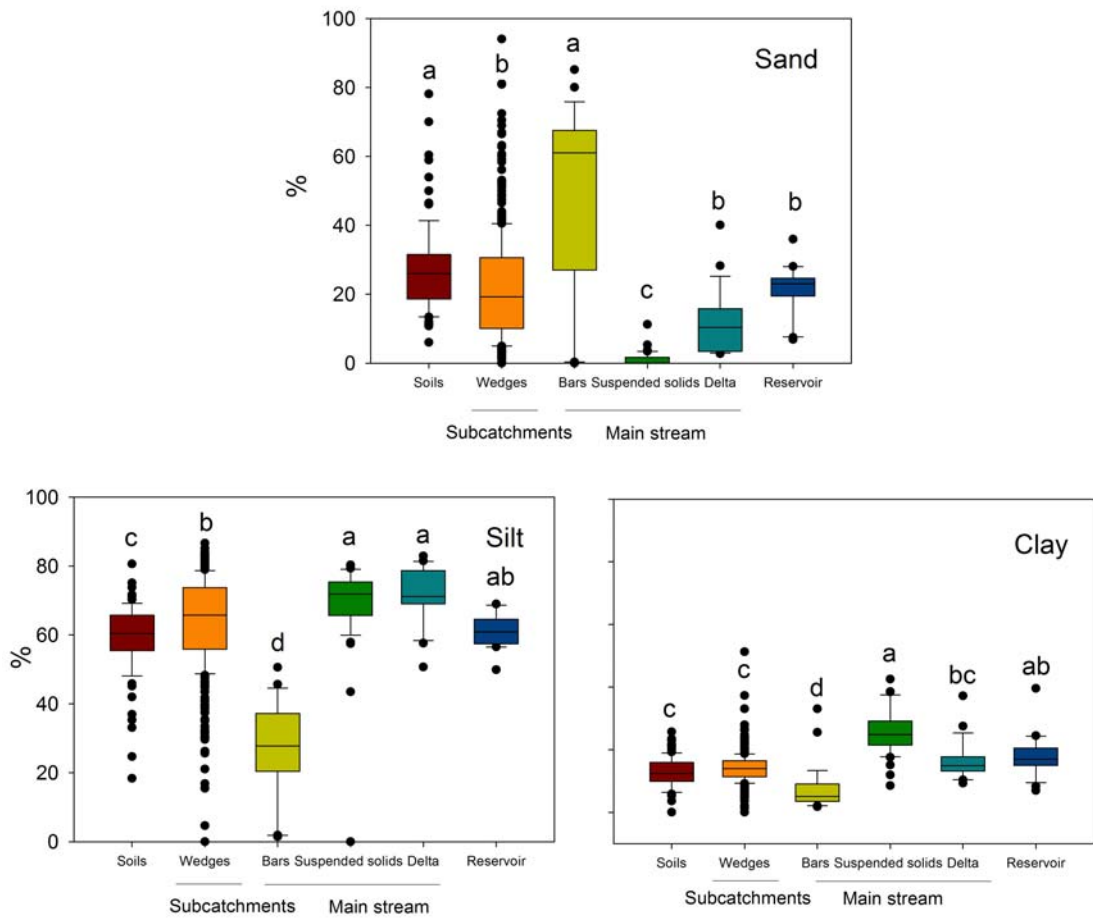


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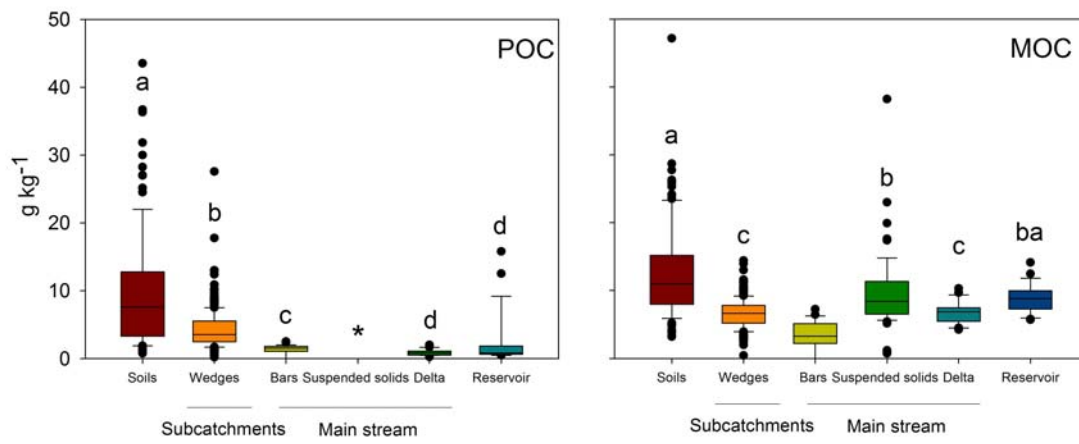
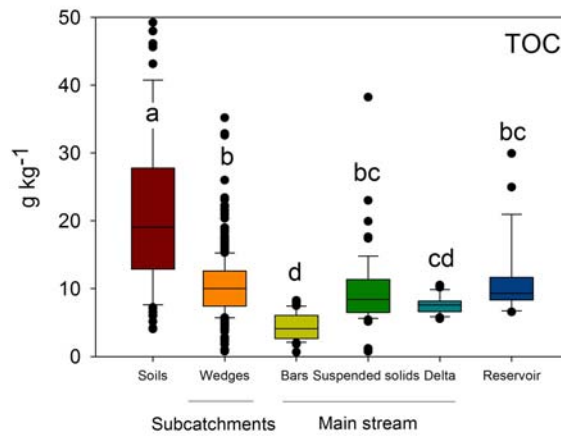
Figure 1



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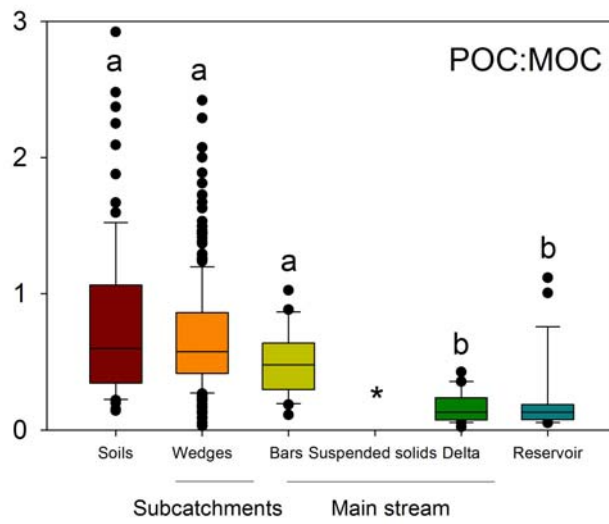
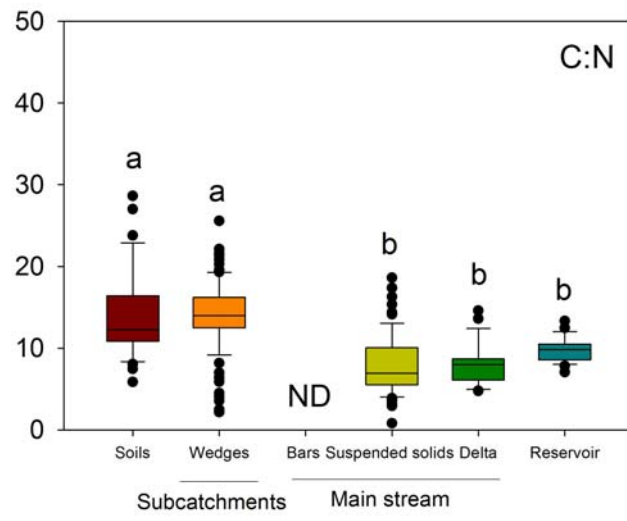


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Figure 4



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