

4 Murcia, Spain, 20<sup>th</sup> February 2019

5 Dear Dr. Ji-Hyung Park,  
6

7 First of all, we would like to thank once again the Handling Editor for the time dedicated to  
8 our manuscript and for their numerous and very constructive and helpful comments. We  
9 have addressed all the comments from the Handling Editor and we feel that the manuscript  
10 has substantially improved with the changes introduced. We have included a point-by-point  
11 response to the comments by the Handling Editor, and here we summarize the mayor  
12 changes in this revised version of this manuscript.

13 According to the Handling Editor comment regarding our previous responses:

14 *“1. Inadequate author response*

15 “...Nevertheless, higher basal respiration in sediments compared to the source soils only  
16 occurred at the surface layer of the deposits (e.g., alluvial wedges and reservoir; please see  
17 Table 4), where at the same time aggregate formation is occurring (as explained before). On  
18 the contrary, at the deep layers of the alluvial wedges and the reservoir, where the carbon is  
19 being stored and stabilized, much lower basal respiration rates than at the upper layers, and  
20 even than in the source soils are observed.”

21 AND

22 *“Another related review comment and response:*

23 P 13 L 17-21: Please rewrite (better split) this long and vague sentence. This and the following  
24 sentences are logically conflicting, because you are arguing that OC in the deposited  
25 sediments is more stabilized than the source soils, even though more labile materials, as  
26 evidenced by higher BR rates, exist in the same sediments. Please clarify this.

27 “Please note that our statements are not contradictory at all. On one hand, higher basal  
28 respiration is a surrogate of microbial activity and therefore can be used as an index of the  
29 lability of the material being degraded by the microorganisms (Paul et al., 2001).  
30 Microorganisms are activated by the presence of labile (easily decomposable) organic matter  
31 and then respire more. But this labile organic matter is further transformed in other  
32 bioproducts which can be more or less recalcitrant, can be chemically attached to other  
33 molecules and even physically protected by aggregates, as it has been pointed out by many  
34 previous studies (Six et al., 2000; 2002; Cotrufo et al 2013; 2015, Deneff et al 2014, etc). At  
35 the end organic matter can be protected (and preserved) by their inherent chemical  
36 composition, by physical protection mechanisms (intra-POM), or both. See Schmidt et al  
37 2011 for futher details.”

38 **Editor comment:** *“As you noted, the surface and deep layers of both alluvial wedges and*  
39 *reservoir sediments exhibited different degrees of “microbial degradation” and “protection”,*  
40 *which should be described and interpreted with caution. Please change ambiguous*  
41 *descriptions(e.g., L 33-35 on P 1: “Aggregate formation and OC accumulation, mainly*

42 *associated with macroaggregates and occluded microaggregates within macroaggregates,*  
43 *were predominant in depositional areas”.) throughout the manuscript and make it clear*  
44 *that your argument would be valid for the deeper layers in depositional areas. Of course*  
45 *you can cite some supporting papers in your discussion, but above all you need to base your*  
46 *argument on your own findings. For instance, protected OC and microbial degradation*  
47 *index values for surficial sediments (Table 4) don't support what you are saying here”*

48 **Response:** Following the Handling Editor suggestion, we have now clarified throughout the  
49 manuscript (hereafter, MS) whether we are referring to superficial or deep layers of the  
50 depositional sites. Moreover, the different role of both deposits in the OC mobilization by  
51 erosion processes throughout the MS has been highlighted.

52 According with our results (explained in page 14, lines 20-24 in the previous MS version)  
53 aggregate formation is occurring mainly at the surface layer of the depositional areas favored  
54 by the growth of roots and the stimulation of microorganisms by root exudates of established  
55 terrestrial and aquatic vegetation at these depositional areas. In addition, in the upper  
56 surface of the depositional areas an enrichment of the OC with respect to the most eroding  
57 areas is also observed, which means that in these surface layers OC is being accumulated. At  
58 the deep layers of the depositional areas, however, aggregate formation process unfolded very  
59 slowly compared to the upper layers, and a more passive (resistant to oxidation) OC, more  
60 preserved from mineralization (is also observed. This statement is also supported by the  
61 observed lower basal respiration rates in the deep layers than in the upper layers of the  
62 depositional areas.

63 We thus consider that the two indices proposed in the previous version of the MS support our  
64 interpretation. For example, the microbial degradation index is higher (OC less susceptible of  
65 being degraded by microorganisms) at the deep than at the surface layers, which is consistent  
66 with the higher content of the OC pool resistant to oxidation in the former than in the latter.  
67 Likewise, the fact that the microbial degradation index was strongly negatively correlated  
68 with basal respiration rates across the catchment support the reliability and validity of this  
69 index. On the other hand, the OC stabilized (protected) in macroaggregates is higher at the  
70 deep than at the surface layers, and even higher than that of the most eroded areas where the  
71 sediment are coming. Moreover, at the surface layer of both depositional areas the proportion  
72 of OC stabilized (protected) in macroaggregates is in the same range than that of the  
73 sediment source areas, but higher than that obtained in the solids in suspension, in which  
74 mineralization ratios are also higher and no aggregate formation is occurring. The fact that  
75 an accumulation of OC is occurring in the surface layer of the depositional areas whereas the  
76 degradation index is relatively low (*i.e.*, the OC is more susceptible to be degraded) is not  
77 contradictory at all because our results suggest that the labile OC inputs (more easily  
78 degradable, such as root exudates) are probably inductors of the formation of  
79 microaggregates within macroaggregates. This statement is supported by the observed  
80 positive correlations between the active OC pool (M<sub>pom</sub>) and the percentages of  
81 macroaggregates (M) and occluded microaggregates (M<sub>m</sub>) and its OC contents (total OC-M  
82 and OC-M<sub>m</sub>) in these areas.

83 Nevertheless, and in agreement with the Handling Editor, we acknowledge that the microbial  
84 degradation index might be used with caution since it would need to be combined with other  
85 analytical approaches (such as stable isotopes analysis), which unfortunately have not been  
86 conducted in this study, to enhance and reinforce its interpretation. Thus, and given that the  
87 suppression of this index does not modify the interpretation of the results nor the main  
88 conclusions drawn from this study, we have deleted it from the previous version of MS to  
89 avoid misunderstandings and conflicting interpretations by the readers.

90 “2. Inadequate author response

91 “...We agree with the comment made by the editor that soil and sediment respiration rates  
92 under field conditions may differ from those under laboratory controlled conditions.”

93 **Editor comment:** *Please provide this discussion in the main text and indicate clearly the*  
94 *new discussion if you had already done it.*”

95 **Response:**

96 Apologies beforehand, but we don't really understand the reason why the editor suggests to  
97 include this issue in the discussion section since it was a methodological issue that had been  
98 already addressed in the previous version of the manuscript. We acknowledge that we  
99 misunderstood the Handling Editor's comment and emphasize the reason why we used  
100 laboratory incubations rather than *in situ* field measurements to estimate organic matter  
101 mineralization rates when a different concern (*i.e.*, if soil and sediment aerobic incubations  
102 would be representative of anaerobic conditions of the deep layers at the depositional sites)  
103 had been raised. Regarding this issue, and in agreement with the Handling Editor comment,  
104 aerobic conditions can accelerate the degradation of organic matter in sediments otherwise  
105 limited by O<sub>2</sub>. But as previously explained, we needed to run laboratory incubations under  
106 standardized conditions (e.g., temperature, moisture, O<sub>2</sub> availability) in order to make  
107 mineralization rates comparable among sediment sources and deposits as well as to relate  
108 them to OC quality. Please note that although our approach (*i.e.*, incubations under aerobic  
109 conditions) would have slightly overestimated the mineralization rates of the sediments in  
110 the deep layers at the alluvial wedges and the reservoir, such mineralization rates were still  
111 about four-times lower than those from the surface sediments, which doesn't invalidate our  
112 rationale. Nevertheless, and in order to better justify the proposed methodology, we have  
113 now included a paragraph and provide references to justify our approach in section 2.4.

114 “3. Inadequate response to a specific comment:

115 P 2, L 1-2: Please be more specific in providing your major conclusion about the relative  
116 importance of “temporary and permanent deposits. Do you mean that both sources are  
117 equally important?”

118 “According to the results from our work we consider that both, temporary and permanent  
119 deposits are very important from the point of view of the physico-chemical mechanisms of  
120 OC protection and stabilization that are occurring across the catchment. In addition, it is not  
121 our purpose to determine if one of them is more important than the other. We consider that  
122 both temporary and permanent deposits should be preserved, although not beyond the  
123 natural fluvial dynamics specially for the natural transitory deposits (short-term residence  
124 times), due to their high potential as C sinks.”

125 **Editor comment:** *Again, please pay attention to differences in basal respiration and*  
126 *microbial degradation index among different sediment categories - i.e., the suspended/bed*  
127 *sediments, and the surface and deep layers of depositional sediments. At least you need to*  
128 *specify what role is important and how different it is among the sediment categories.*”

129 **Response:** In agreement with the Handling Editor comment, we have removed these  
130 general sentences from the abstract and the conclusion sections. Moreover, and as already  
131 stated, we have now clarified when we are referring to superficial or deep layers of the  
132 sediment deposits throughout the MS.

133 “4. Inadequate author response

134 P 7 L 10: Why don't you use simply “OC-M” as denominator? In addition, it is assumed here  
135 that OC in free microaggregates and mineral fractions is older than OC in macroaggregates.  
136 Do you have any data supporting this assumption? If not, you need to reformulate relevant  
137 sentences throughout the manuscript.

138 “We agree with the editor that the total OC in M could have been used in the denominator  
139 (the sum of each sub-fraction is equal to the total OC in macroaggregates). However, we feel  
140 that displaying the different sub-fractions contained within the macroaggregates is a clearer  
141 way to present this index.”

142 **Editor comment:** *Please clarify why you felt so, or the index makes sense. Based on*  
143 *what?”*

144 **Response:** As previously explained in the former response to the Handling Editor  
145 comments, this index is based on a recent study by De Clercq et al., (2015). Nevertheless, as  
146 already stated, we have dropped the microbial degradation index and all the interpretations  
147 regarding this index from the revised version of the MS, as it has been explained in point 1.

148 “5. Inadequate author response:

149 P 7 L 14: There must have been significant reductions in soil moisture given the high  
150 incubation temperature and 32 days of incubation. Please clarify this.

151 “As stated in the previous MS version (in page 8, lines 18-20) the moisture content of the  
152 samples was regularly checked for potential water losses by evaporation by weighting the  
153 bottles, but there was not water losses and therefore it was not necessary to add any water”.  
154 Please note that 32 days of incubation is not a long period for water losses to occur.”

155 **Editor comment:** *This would be very difficult to believe for any research who has ever*  
156 *done a similar soil incubation experiment. Where would evaporated water go during the*  
157 *'long' period of 32 days? Did you keep the bottles closed or open to the air?*

158 As mentioned in the previous version of the MS (page 7, line 20) soil and sediment samples  
159 were put “in hermetically-sealed flasks”, so flasks were closed during the incubation  
160 experiment, except after each measurement when flasks were opened for 30 minutes to avoid  
161 the accumulation of CO<sub>2</sub>. We have now clarified that flasks were opened for 30 minutes after  
162 each measurement in the revised version of the manuscript since we had omitted this  
163 important information. Apologizes for the missing information. Given the short time that  
164 flasks were opened over the whole incubation period no water losses by evaporation occurred  
165 (of course this was checked by monitoring gravimetric soil/sediment moisture periodically  
166 after each measurement by weighting the flasks and comparing the weights after each  
167 measurement with the initial weights).

168 6. Minor correction needs:

169 -“ Please check carefully editorial errors such as “100 cc volume” (P 5, L 13)”

170 We have changed “cc” to “cm<sup>3</sup>”.

171 -“ P3 L 5 - P4 L2: Please check and split paragraph if required”.

172 The paragraph has been split as requested.

173 - "P 8 L 1: Please describe why you opted for the nonparametric test. You might need to  
174 mention any prior test for normal distribution".

175 "We used a non-parametric test because our sampling design was not balanced. That is, we  
176 did not have the same amount of representative samples (n<sup>o</sup> of replicates) across eroding,  
177 transport and depositional areas (see Table 1). Please note that prior normality distribution  
178 tests are not required when non-parametric tests are performed."

179 *Editor comment: Please describe this in section 2.5.*

180 Following the editor suggestion, we have now clarified why we opted for the nonparametric  
181 test in the statistical analysis section from the revised version of the MS (page 8, lines 4-6)

182 - "P 11 L 14: Please rephrase "macroaggregates are the nucleus for microaggregate  
183 formation". How can larger macroaggregates function as the nucleus?"

184 This sentence is based on Oades (1984) theory who postulated that "the roots and hyphae  
185 holding together the macroaggregate form the nucleus for microaggregate formation in the  
186 center of the macroaggregates".

187 *Editor comment: Please provide the reference, whenever you cite a specific paper.*

188 Please note that the reference by Oades (1984) had been already included in the main text as  
189 well as in the reference list from the previous MS (page 12, line 12).

190 - *Table 4: "Protected OC" is still in use in the table though the caption uses the new term.*

191 We have changed "protected OC" to "stabilized OC in macroaggregates" in table 4 and  
192 through the captions.

193 - *Figs. 3-4: Please double check different letters indicating significant differences. For  
194 example, I doubt whether free mineral is really not different between bedload and surface  
195 reservoir.*

196 Thank you very much for your deep review and apologizes for the mistake. We are so sorry.  
197 You are right. The free mineral fraction is significantly different between the bedload and the  
198 surface reservoir. We have now corrected ourselves and the letters indicating significant  
199 differences among deposits have been changed in Figures 3 and 4.

200 Also, we have detected other errors in Figure 4A: the letters for the occluded mineral fraction  
201 in suspended load and occluded microaggregates in the surface reservoir were missing; and  
202 Figure 4B: letters for the occluded mineral fraction in forest soil was missing. These letters  
203 have been now included.

204 A new reference from Paul E.A. 2001 (page 7, line 15) and Garcia-Franco 2015 has been now  
205 included (now, 2015a and 2015b: page 11, lines 14 and 16).

206

207 Yours sincerely,

208 Dr. María Martínez-Mena, on behalf of all the co-authors

209

# Fluvial sedimentary deposits as carbon sinks: organic carbon pools and stabilization mechanisms across a Mediterranean catchment

María Martínez-Mena<sup>1</sup>, María Almagro<sup>2</sup>, Noelia García-Franco<sup>3</sup>, Joris de Vente<sup>1</sup>, Eloisa García<sup>1</sup>, Carolina Boix Fayos<sup>1</sup>.

<sup>1</sup> Soil Erosion and Conservation Research Group, CEBAS-CSIC, Spanish Research Council, Campus de Espinardo 30100, P.O. Box 164, Murcia, Spain

<sup>2</sup> BC3-Basque Centre for Climate Change, Sede Building 1, 1st floor, Scientific Campus of the University of the Basque Country, 48940, Leioa, Basque Country, Spain

<sup>3</sup> Technische Universität München Lehrstuhl für Bodenkunde Wissenschaftszentrum Weihenstephan Emil-Ramann-Strasse 2 / I D 85354 Freising-Weihenstephan

Correspondence to: María Martínez-Mena ([mmena@cebas.csic.es](mailto:mmena@cebas.csic.es))

Código de campo cambiado

**Abstract.** The role of fluvial sedimentary areas as organic carbon sinks remains largely unquantified. Little is known about mechanisms of organic carbon (OC) stabilization in alluvial sediments in semiarid and sub-humid catchments where those mechanisms are quite complex because sediments are often redistributed and exposed to a range of environmental conditions in intermittent and perennial fluvial courses within the same catchment. The main goal of this study was to evaluate the contribution of transport and depositional areas as sources or sinks of CO<sub>2</sub> at the catchment scale. We used physical and chemical organic matter fractionation techniques and basal respiration rates in samples representative of the three phases of the erosion process within the catchment: i) detachment, representing the main sediment sources from forests and agricultural upland soils, as well as fluvial lateral banks; ii) transport, representing suspended load and bedload in the main channel ; and iii) depositional areas along the channel, downstream in alluvial wedges and in the reservoir at the outlet of the catchment, representative of medium and long-term residence deposits, respectively. Our results show that most of the sediments transported and deposited downstream come from agricultural upland soils and fluvial lateral bank sources, where the physico-chemical protection of OC is much lower than that of the forest soils, which are less sensitive to erosion. The protection of OC in forest soils and alluvial wedges (medium-term depositional areas-) was mainly driven by physical protection (OC within aggregates), while chemical protection of OC (OC adhesion to soil mineral particles) was observed in the fluvial lateral banks. However, in the remaining sediment sources, in sediments during transport, and after deposition in the reservoir (long-term deposits-), both mechanisms are equally relevant. Mineralization of the most labile OC (~~the~~ intra-aggregate particulate organic matter (Mpom), was predominant during transport. Aggregate formation and OC accumulation, mainly associated with macroaggregates and occluded microaggregates within macroaggregates, were predominant in the upper layer of depositional areas. However, OC was highly protected and stabilized at the deeper layers, -mainly in the long-term deposits (reservoir). being even more protected than the OC from the most eroding sources (agricultural soils and

fluvial lateral banks). Altogether our results show that both temporary-medium and long-term depositional areas can play an important role in erosive areas within catchments, compensating OC losses from the eroded sources and functioning as C sinks.

## 1 Introduction

Soil erosion, a complex process that causes transport and deposition of sediments with accompanying soil organic carbon (SOC) (Gregorich et al., 1998; Wang et al., 2014), affects the dynamics of the terrestrial carbon (C) cycle and have important implications for the rate of C inputs into the soil (i.e. plant net primary productivity), as well as for the accumulation and stability of organic matter in soil (Berhe and Kleber, 2013). According to some authors (Van Oost et al., 2007), lateral C fluxes are key to determine the fate of soil organic carbon (SOC) at the landscape scale. However, current estimations of those fluxes (based on soil erosion and the associated SOC content), show high variation among studies (Quinton et al., 2010; Doetterl et al., 2012). The fate of the redistributed organic carbon (OC) depends on multiple factors: i) the nature of the soil organic matter being detached from different “sources” within a catchment (Nadeu et al., 2011; 2012; Kirkels et al., 2014); ii) its turnover rates during transport; iii) the type of erosion processes (selective or non-selective); iv) the connectivity and distance of travel between eroding sources and the streambed (Boix-Fayós et al., 2015; Wang et al., 2010); and v) the micro-environmental conditions under which the OC is stored in sedimentary settings (Van Hemelryck et al., 2011; Berhe and Kleber, 2013). All these factors, which affect the protection of OC against decomposition through physical and chemical mechanisms, remain considerably uncertain. Despite a combination of different techniques (isotopic, spectroscopic, and traditional wet chemistry) has been used (Wang et al., 2014, Kirkels et al., 2014, Liu et al., 2018) to determine if the eroded OC is lost after erosional redistribution, a full understanding of the dynamics of and interactions between OC sources and sinks, in relation to soil erosion and redistribution, is still absent (Doetterl et al., 2016; Hoffman et al., 2013).

Determination of how the different OC pools are transported by erosion from different sources, as well as the effect of new soil structure formation by aggregation and OC stabilization after deposition, can partially contribute to answering this question (Doetterl et al., 2016). In fact, it is already known that soil aggregates physically protect OC from rapid decomposition by microorganisms (Razafimbelo et al., 2008; Six et al., 2000), and aggregate formation appears to be closely linked to soil C storage and stability (Barreto et al., 2009; Golchin et al., 1995, Salomé et al., 2010). Related to this, Wang et al. (2014) found that soil erosion and transport result in disaggregation and, consequently, SOC mineralization (Lal, 2005; Boix Fayos et al., 2015), while depositional and burial processes promote the formation of macroaggregates that contribute to the physical stabilization or protection of soil organic matter from decomposition, as well as enhance sorptive interactions of soil minerals with organic matter protecting OC from microbial decomposition (Berhe et al., 2013) The protection provided by macroaggregates as an important mechanism for SOC stability in soils affected by soil erosion processes has been also reported (Berhe et al., 2018, Nie et al., 2018). However, to the best of our knowledge, this is the first study to

Con formato: Normal

Con formato: Fuente: Negrita, Español (alfab. internacional)

include the assessment of microaggregates contained within macroaggregates and their associated OC, widely acknowledged as an indicator of the physical protection of SOC (Six, et al 2004, Deneff et al., 2007, Six and Paustian, 2014), during the different phases of soil erosion at the catchment scale.

Several authors have suggested that alluvial settings are important, non-quantified OC sinks (Wisser et al., 2013; Hoffmann et al., 2013; Ran et al., 2014) with mechanisms of OC stabilization that can be quite complex in semiarid and sub-humid areas. In these areas sediments are often redistributed and exposed to a wide range of environmental conditions (e.g., rewetting-drying cycles, high temperatures, and solar radiation) and subcatchments with ephemeral, intermittent, and perennial fluvial courses, sometimes within the same catchment (Boix-Fayos et al., 2015). Ephemeral and intermittent fluvial courses represent more than 50% of the global river network, being the dominant type of water course in dry climates (Datry et al., 2014). In addition, it is expected that this percentage will increase worldwide in response to climate change and increased water extraction for human use (Shewe et al., 2014), because intermittent and ephemeral rivers will suffer more severe, sustained, and more frequent droughts (De Girolamo et al., 2017; Vadher et al., 2018). Thus, understanding the OC stabilization mechanisms within these fluvial systems might have important implications for the stability of soil OC stocks as affected by soil erosion and in response to climate change. Moreover, the quantification of the mineralization rates and the assessment of the stabilization mechanisms of OC induced by soil erosion, together with the identification of source materials contributing to the sediment OC dynamics within a catchment, are key to determine C budgets, to feed and develop prediction models, and to handle soil conservation strategies at catchment scales (Liu et al., 2018). From previous works, where the importance of soil disturbance in determining the way that OC associated with sediments is transported and deposited within the catchment were highlighted (Nadeu et al., 2011; Boix-Fayos et al, 2015), new research questions arose regarding the main stabilization mechanisms of this OC during transport and in several sedimentary deposits within the catchment. To answer these questions, here we take a step further, studying in depth how OC is mobilized across the catchment using a combination of physical and chemical organic matter fractionation techniques and comparing the different sedimentary deposits with three main sediment sources (forest and agricultural upland soils, and fluvial lateral banks) to determine their relevance in OC-sediment transport and deposition. Furthermore, we estimate basal respiration rates of upland soils, fluvial lateral banks, sediments in transit, and the different deposits. Combining information on the OC associated with soil aggregates and mineral particles, as an indicator of OC physico-chemical protection, and basal respiration rates can shed light on whether OC is being accumulated or lost by erosion at the catchment scale. The main objective of this work was to evaluate the potential of the transport and depositional areas as sources or sinks of atmospheric CO<sub>2</sub> in a subhumid Mediterranean catchment. The specific objectives were to: i) assess the changes in the aggregate size distribution and associated OC in the sediments mobilized by soil erosion processes; ii) identify the main stabilization mechanisms of OC in eroded soils and different sedimentary deposits; and iii) determine the potential sources of eroded OC in the sedimentary deposits through the catchment. We performed the study in a sub-catchment (111 km<sup>2</sup>) in the headwaters of the Segura catchment in South East Spain.



## 2 Methods

### 2.1 Study area

The study area is located in the headwaters of the Segura catchment (Murcia, South East Spain), which drains to the Taibilla reservoir (Turrilla catchment) and is formed by three adjacent sub-catchments (Rogativa, Arroyo Blanco, and Arroyo Tercero) covering a total area of 111 km<sup>2</sup> (Fig. 1a). The dominant lithology of the catchment consists of marls, limestones, marly limestones, and sandstones of the Cretaceous, Oligocene, and Miocene. The dominant soils in the area are Lithosols, Regosols, and Cambisols (IUSS Working Group WRB, 2015). The catchment is representative of the environmental conditions in Mediterranean mountainous areas of medium altitude. This catchment receives 530 mm of precipitation per year and has an average temperature of 13.5°C. It has experienced important land use changes since the 1970s involving reforestations, including the construction of a dense network of check-dams for hydrological and sediment control. Socio-economic changes in the region resulted in the abandonment of agricultural activities and the recovery of the shrubland and forest. Nowadays, forests and shrublands represent approximately 80% of its area while agricultural land represents 20%. The hydrological and geomorphological effects of these catchment changes were studied in Boix-Fayos et al., (2008), Quiñero-Rubio et al., (2016), and Pérez-Cutillas et al., (2018). The main fluvial course studied is ephemeral with water only flowing a few times per year, during intense rain events. A deeper description of the study area is given in Boix-Fayos et al., (2015).

### 2.2 Field experimental design and sampling

The experimental design combines a fluvial geomorphological perspective and soil science analysis. Previous geomorphological analyses of the channel and adjacent areas and of the dynamics of the fluvial morphology in the last 60 years (Boix-Fayos et al., 2007; Nadeu et al., 2011) were used to identify the main sources and sinks of sediments in the Turrilla basin. This previous analysis was the background to the combination of the sediment cascade (Hoffman et al., 2013; Boix-Fayos et al., 2015) and the erosion cycle as the experimental approach (Fig. 1b). To accomplish it a sampling design was established (Fig. 1a, Fig. 1b, Table 1) that represented: (i) the eroding areas (source of sediments) and detachment phase; (ii) the transport areas (main channel) and the main transport processes (suspended sediments and bedload); and (iii) the depositional areas (along the channel and downstream), representing the sedimentary phase with medium-term depositional areas (alluvial wedges) and long-term depositional areas (reservoir sediments).

In each area and erosion phase, a representative number of samples were taken to cover the spatial variability of soils and sediments within the catchment. At each sampling point paired undisturbed (core of 100 ~~cc~~ <sup>cm<sup>3</sup></sup> volume) and disturbed (~~sampling auger of 100 cc~~) soil and sediment samples were collected for bulk density estimations and chemical analyses, respectively. All soil and sediment samples were stored in a freezer (4 °C) until they were processed for chemical analyses. A total of 89 samples were analyzed:

Con formato: Superíndice

1. In eroding areas (Fig. 1a, 1b), representing the detachment phase of the erosion cycle, two sources of sediments were sampled: (i) surface soils under the two main land uses of the catchment area: forest and agricultural land; and ii) fluvial lateral banks well connected to the channel (Fig. 1b, Table 1). Surface soils (0-10 cm) samples were taken from different locations distributed from upstream to downstream in the catchment, representing the main land uses and covers (high- and low- density forests, pastures, shrublands and croplands), but were finally pooled in two main groups: forest (less eroded) and agricultural soils. Fluvial banks (considered as subsoils) were sampled at an average depth of 80 cm.

2. In transport areas, suspended sediments and bedload samples were collected in two fluvial reaches with different flow regimes, representing an ephemeral and a permanent stream. Suspended sediment samples were taken using a siphon sampler device designed to collect suspended sediment samples at different depths immediately after each flooding event (Dielh 20018). Six samplers (1-L bottles) were spaced vertically (7.5 cm apart) and connected to an intake tube and an air vent. A limited number of samples, for which sufficient material was available to carry out all the analyses, were selected. The suspended sediment samples were taken over a period of 2 years. Moreover, the bedload in the fluvial bars and the channel of both ephemeral and permanent reaches were sampled at 0-10 cm depth as representative of the bed load sediments, which are considered to be ephemeral at these dynamic positions. Both bare and vegetated fluvial bars were sampled in two periods of the year, representing dry and wet conditions.

3. At depositional sites, samples were taken at 0-40 cm and at 40-80 cm depths in four alluvial wedges behind check-dams that were installed in the catchment in the 1970s. These alluvial wedges had a depth of between 1 and 3 meters and many of them were covered by vegetation. They were considered sedimentary areas of medium-term residence times. At the outlet of the catchment the reservoir sediments were sampled down to a depth of 3 meters, at several points. The different depths were pooled into surface layer (0-40 cm depth) and deep layer (40-300 cm). These sediments were considered to have long-term residence times. The samples used in the analysis, and their depths, are shown in Table 1.

## **2.3 Soil and sediment analyses**

### **2.3.1 Water-stable soil aggregate size distribution**

Water-stable soil aggregate-size separation was carried out using a modified wet sieving method adapted from Elliott (1986). Briefly, a 100-g sample of air-dried soil, disaggregated by hand, was placed on top of a 2000- $\mu\text{m}$  sieve and submerged for 5 minutes in deionized water at room temperature. The sieving was performed manually by moving the sieve up and down 3 cm, 50 times in 2 minutes, to achieve aggregate separation. Two sieves (250 and 63  $\mu\text{m}$ ) were used to obtain three aggregate classes: (i)  $>250 \mu\text{m}$  (macroaggregates; M), (ii) 63–250  $\mu\text{m}$  (microaggregates; m), and (iii)  $<63 \mu\text{m}$  (silt plus clay-sized particles; min). The aggregate-size classes were oven dried (50°C), weighed, and stored in glass jars at room temperature (21° C) (Fig. 2). From this, the mean weight diameter (MWD) was obtained as an indicator of aggregate stability.

Secondly, and in order to quantify the protected microaggregates contained within macroaggregates, the procedure described by Six et al. (2000) and Deneff et al. (2004) was carried out (Fig. 2, square in green). A subsample (10 g) of the macroaggregates was immersed in deionized water on top of a 250- $\mu\text{m}$  mesh screen, inside a cylinder. The macroaggregates were shaken together with 50 glass beads (4-mm diameter) until complete macroaggregates disruption was observed. Once the macroaggregates had been broken up, microaggregates and other material  $<250\ \mu\text{m}$  passed through the mesh screen, with the help of a continuous water flow to the sieve. The material retained on the 63- $\mu\text{m}$  sieve (silt+clay; min) was wet sieved, to ensure that the isolated occluded microaggregates were water-stable (Six et al., 2000). These microaggregates obtained from macroaggregates (Mm) were oven-dried, at 50°C (24 h) in aluminum trays, and weighed. The material retained on top of the 250- $\mu\text{m}$  mesh was considered the intra-aggregate particulate organic matter (Mpom), representing the most labile fraction (active). It was separated and weighed after drying in an oven at 50°C (Fig. 2).

### 2.3.2 Oxidation of mineral fractions

The free and occluded mineral fractions ( $< 63\mu\text{m}$ ) obtained in steps 1 and 2 (Fig. 2) were oxidized by NaOCl to obtain a chemically-resistant C fraction (rOC) (Zimmermann et al., 2007) representing the passive pool. One gram of every mineral fraction was oxidized for 18 h at 25 °C, with 50 mL of 6% NaOCl adjusted to pH 8 with concentrated HCl. The oxidation residue was centrifuged at 1000 g for 15 min, decanted, washed with deionized water, and centrifuged again. This oxidation step was repeated twice. The residue was dried at 50 °C and weighed.

### 2.3.3 Organic carbon and nitrogen analysis and pools ratios

The organic carbon (OC) and total nitrogen (N) concentrations were determined separately for each water-stable aggregate-size class and for the occluded microaggregates and occluded mineral fractions using an Elemental Analyzer (LECO TRUSPEC CN, Michigan, USA), after carbonates removal using 2 M HCl. All the samples were analyzed in triplicate. When necessary, the OC concentration of each water-stable aggregate-size class, as well as that of the intra-aggregate POM and of the microaggregates occluded in the macroaggregates, was expressed on a sand-free aggregate basis (Elliott et al., 1991). The OC content was also expressed on a soil basis, by multiplying the C concentration in each fraction by the weight proportion of that fraction:

$$\text{OC content (g OC kg}^{-1}\text{soil)} = (\text{OC})_{\text{fraction}} * (\text{proportion of the fraction})_{\text{soil}}$$

Where (OC) fraction is the OC concentration in each fraction and (proportion of the fraction) soil is the percentage of the OC that each fraction represents in the bulk soil. The same procedure was followed to analyze total N in each of the separated fractions.

The total OC and total N in the bulk soil were considered as the sum of the OC or total N in each separated water-stable aggregate-size fraction:

$$\text{Total OC (or total N)} = \text{macroaggregate (M)} + \text{microaggregate (m)} + \text{mineral fraction (min)}$$

In addition, we used the macroaggregate stabilization ratio, as an indicator of the physico-chemical stabilization of OC in macroaggregates (OC occluded in microaggregates and occluded in mineral particles, compared to that associated to the intra-aggregate particulate organic matter fraction), based in the macroaggregate turnover index by Six et al., (2000):

$$\text{Stabilized OC in macroaggregates} = \frac{(\text{OC-Mm} + \text{OC-Mmin})}{(\text{OC-Mpom})}$$

Where OC-Mm and OC-Mmin refer to the OC associated with the occluded microaggregates and the mineral fraction, respectively, and OC-Mpom refers to the OC associated with the intra-aggregate particulate organic matter fraction. The higher the ratio, the more stabilized the OC is within macroaggregates (Fig. 2).

~~ii) The degree of microbial degradation index, as an indicator of the OC stability/decomposability, based on De Clercq et al. (2015). According to this, the OC associated with the free microaggregate and mineral fractions (OC m and OC min, respectively) was more degraded (representing the oldest OC), while the OC occluded in macroaggregates (in the intra-aggregate particulate organic matter fraction (OC Mpom) and OC in the occluded microaggregate and mineral fractions within macroaggregates (OC Mm and OC Mmin, respectively) was less degraded (representing the youngest and intermediate OC). The degree of microbial degradation decreases in the following order: Mpom > occluded microaggregates > occluded mineral > free microaggregates > free mineral.~~

~~$$\text{Degree of microbial degradation index} = \frac{(\text{OC}_m + \text{OC}_{\text{min}})}{(\text{OC}_{\text{Mm}} + \text{OC}_{\text{Mmin}} + \text{OC}_{\text{Mpom}})}$$~~

~~Where OC m and OC min refer to the OC associated with the free microaggregates and mineral fraction, respectively, OC Mm and OC Mmin refer to the OC associated with the occluded microaggregates and occluded mineral fraction, respectively, and OC Mpom refers to the OC associated with the intra-aggregate particulate organic matter fraction (Fig. 2). The higher the ratio, the the lower the OC decomposability is.~~

#### 2.4 Soil and sediment incubations

Soils and sediments were incubated under controlled aerobic conditions (28 °C, 60% of the water holding capacity) for 32 days while keeping moisture (at 60% of the water holding capacity) and temperature (28 °C) constant to estimate their potential OC mineralization rates (mg CO<sub>2</sub> kg<sup>-1</sup> soil or sediment). Thus optimal conditions for microbial activity were

Con formato: Inglés (Reino Unido)

Con formato: Inglés (Reino Unido)

Con formato: Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos), Sin Resaltar

Con formato: Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos), Subíndice

Con formato: Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos), Superíndice

Con formato: Inglés (Estados Unidos)

mimicked in order to induced the (natural) potential maximum heterotrophic CO<sub>2</sub> soil respiration, following the approach adopted by other authors (Paul et al., 2001; Doetterl et al 2012). We incubated three replicate samples of 30 g of fresh soil and sediment material that was prepared for the fractionation from the different areas throughout the catchment- and at different depth intervals (see Table 1). Previously, the maximum water holding capacity for each sample was estimated in triplicate, following the procedure of Howard and Howard (1993). Each sample was put in a hermetically-sealed flask (125 ml) with no further additives. The CO<sub>2</sub> released was measured periodically (every day for the first four days, every three days during the second week, and then weekly) using an infrared gas analyzer (CheckmateII, PBI Dansensor, Denmark) and the flasks were opened for 30 minutes after each measurement to avoid the accumulation of CO<sub>2</sub>. The moisture content of the samples was also checked periodically, but replacement of the evaporated water was not necessary during the experiment. We used linear interpolations between sampling dates and then summed them across all dates to estimate the cumulative amount of CO<sub>2</sub> released (mineralized) after 32 days of incubation; basal soil or sediment respiration was expressed as mg CO<sub>2</sub>-C kg<sup>-1</sup> soil per day.

Con formato: Inglés (Estados Unidos), Subíndice

Con formato: Inglés (Estados Unidos)

## 2.5 Statistical analysis

Statistical tests to detect differences between the means of the sediment sources and sinks -representative of the eroding, transport, and deposition phases of the erosion process - were performed separately for each erosion phase and depth (when applicable) using the non-parametric Kruskal-Wallis test for independent measurements. This non-parametric test was used because the sampling design was not balanced due to the different number of samples (n) in the eroding, transport and depositional areas (Table 1). Significant differences were identified at the 0.05 probability level of significance. Spearman correlations were performed to explore the relationships between most of the studied variables within each erosion phase. All statistical analyses were carried out using SPSS 24.0 (SPSS Inc., Chicago, IL, USA).

Con formato: Inglés (Estados Unidos)

Con formato: Fuente: Sin Negrita

Con formato: Inglés (Estados Unidos)

## 3. Results

### 3.1 Water-aggregate size distribution and associated OC: M, m, and min

Eroding areas: On average, the forest soils had the highest percentage of total macroaggregates (M) and MWD values (Table 2) when compared to the agricultural soils and fluvial lateral banks. The agricultural soils and fluvial lateral banks showed the same distribution trend - a decrease in the percentage of aggregates with increasing aggregate size - while in the forest soils a predominance of macroaggregates existed (Fig. 3a).

The forest soils also showed the highest content of OC associated with the macroaggregates (OC-M), followed by the agricultural soils. The agricultural soils showed higher OC in macroaggregates than the fluvial lateral banks, despite their lower percentage of macroaggregates (Fig. 3a, Fig. 3b). A decrease in OC with decreasing aggregate size was found in the

forest and agricultural soils, while no differences in the OC associated with different aggregate sizes were observed in the fluvial lateral banks (Fig. 3b).

Transport areas: A higher percentage of total macroaggregates and higher MWD, but a lower percentage of free mineral particles, were observed in the bedload, in comparison with the suspended sediments (Table 2). However, the suspended  
5 sediments had a higher OC-M content while the OC content in the free mineral fraction (OC-min) was similar between the two types of transport sediment (Fig. 3a, Fig. 3b). In addition, for the suspended sediments the distribution of aggregates (min>m>M) and the OC associated with the largest aggregates (OC-M) were similar to that of the agricultural soils,

Depositional areas: In the alluvial wedges, the free mineral fraction predominated over the macroaggregates and microaggregates, regardless of depth. The opposite was observed when considering the OC content associated with these  
10 fractions, the order being OC-M > OC-m > OC-min (Fig. 3a, Fig. 3b), similar to the trend in the forest and agricultural soils. In addition, a decrease in the percentage of macroaggregates from the upper to the deepest layer of the alluvial wedges was found. For the reservoir sediments, the percentage of macroaggregates and the OC-M content were highest in the upper layer, while the free mineral fraction represented the highest percentages among the distinct fractions in the deep sediment layers, and had the highest percentages across the catchment (Fig. 3a, Fig.3b).

### 15 **3.2 Intra-aggregate particulate organic matter, occluded microaggregates and occluded mineral fraction within macroaggregates and associated OC: Mpom, Mm and Mmin**

Eroding areas: The Mpom fraction represented about 14% of the total in the forest soil, followed by the fluvial lateral banks and agricultural soils (lower than 5%). The associated OC content in the Mpom (OC-Mpom) oscillated between 3% and 0.61%, decreasing in the following order: forest soils > agricultural soils > fluvial lateral banks (Fig. 4a, Fig. 4b). Although  
20 no clear differences were observed in the percentages of Mm and Mmin among the sediment sources (due to high spatial variability), the contents of OC-Mm and OC-Mmin were highest in the forest soils (Fig. 4b).

Transport areas: The percentage of the Mpom fraction in the sediments in transit (suspended and bedload sediments) was about 10%. The OC-Mpom content in the suspended sediments and in the bedload sediments was similar and lower, respectively, when compared to those of the soils and fluvial lateral banks (Fig. 4b). The sediments in transit displayed a  
25 significant decrease (about 50%) in the Mm and Mmin percentages as well as in the OC associated with these fractions (OC-Mm and OC-Mmin) with respect to the forest soils. However, sediments in transit showed lower percentage of Mm but similar OC-Mm and OC-Mmin contents compared to the agricultural soils and fluvial lateral banks (Fig. 4a, Fig. 4b).

Depositional areas: In the upper sediment layer of the reservoir the percentage of the Mpom fraction was higher and the OC-Mpom content was lower than those of the alluvial wedges (Fig. 4b). In the upper sediment layer of the alluvial wedges the  
30 OC-Mpom content was higher, compared to the forest and agricultural soils (Fig. 4b), while the opposite happened at the

reservoir. In addition, the OC-Mpom content decreased with depth in both depositional areas, although this decrease was more pronounced in the case of the alluvial wedges sediments (Fig. 4b). The percentages of occluded microaggregates (Mm) at the reservoir, and of the occluded mineral fraction (Mmin) in the alluvial wedges, were higher than in the eroding areas (forest and agricultural soils and fluvial lateral banks) (Fig. 4a). However, both types of sedimentary deposit had slightly lower OC contents associated with these occluded fractions (OC-Mm and OC-Mmin) than the forest soils, but the values were similar to those obtained in the agricultural soils and fluvial lateral banks (Fig. 4a, 4b).

The percentage of the total Mm and Mmin decreased significantly with depth in both depositional areas, being more pronounced in the case of the reservoir, where the Mm percentage in the deep sediment layer was reduced up to 75% when compared to that in the upper sediment layer (Fig. 4a). The alluvial wedges had a higher OC-Mm content in the upper sediment layer, and higher OC-Mm and OC-Mmin contents in the deep sediment layer, compared to the reservoir (Fig. 4b).

### 3.3 OC quality

The contents of free and occluded mineral fraction resistant to NaClO oxidation: rOC-min and rOC-Mmin, respectively, were higher in the forest and agricultural soils than in the fluvial lateral banks, with values ranging from 0.55% to 0.13%. Interestingly, the rOC-min in the suspended sediments was comparable to that in the forest and agricultural soils, and was lower than that in the bedload sediments, which displayed contents similar to those of the fluvial lateral banks (Table 3). It is noteworthy that the rOC was higher in the occluded fraction than in the free mineral fraction in the lateral fluvial bank, bedload, and deep layer of the reservoir (Table 3).

The degree of physico-chemical stabilization of the OC in macroaggregates -  $(OC-Mm + OC-Mmin)/OC-Mpom$  - could be divided into three groups: i) the reservoir, showing the highest ratios (more stabilized); ii) the soils and fluvial lateral banks, bedload sediments, and alluvial wedges, with medium ratios; and iii) the suspended sediments, with the lowest value (less stabilized) (Table 4).

~~The microbial degradation index  $(OC_{m} + OC_{min})/(OC_{Mm} + OC_{Mmin} + OC_{Mpom})$  also displayed three groups: i) the fluvial lateral banks, bedload sediments, and deep layer of the reservoir, with higher indices (representing a lower degree of OC decomposability); ii) the agricultural soils, suspended sediments, and deep layer of the alluvial wedges; and iii) the upper layers of the forest soils and depositional sites, with the lowest indices (representing higher degree of OC decomposability) (Table 4).~~

The basal respiration (BR) rates ranged between 0.81 and 6.04 mg CO<sub>2</sub> kg<sup>-1</sup> day<sup>-1</sup> in the eroding sources, the lowest values occurring in the fluvial lateral banks (Table 4). In the sedimentary deposits, BR ranged from 0.70 to 13.8 mg CO<sub>2</sub> kg<sup>-1</sup> day<sup>-1</sup>, the values being highest in the upper sediment layers of the alluvial wedges and lowest in the deep layer of the reservoir. In

Con formato: Fuente: Sin Negrita, Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos)

the transport areas, the suspended sediments had higher respiration rates than the bedload sediments being the second-highest rate observed through the catchment (Table 4).

Higher C:N ratios were found in the upper sediment layers of the alluvial wedges, forest soils, and suspended sediments. A decrease in the C:N ratio with increasing depth occurred in soils and sediments of the alluvial wedges, but no changes with depth were observed at the reservoir (Table 4).

### 3.4 Correlations between the different physical, chemical, and biological variables

Positive correlations between the labile fraction, OC-Mpom, and the total OC associated with macroaggregates (OC-M) were observed in all areas. However, correlations between OC-Mpom and the macroaggregates (M), micro within macroaggregates (Mm) percentage, and OC-Mm content were found in the eroding and depositional areas but not in transport areas. Moreover, in the eroding and depositional areas (the reservoir), positive correlations were obtained between OC-Mpom and the oxidable occluded OC (Table 5). The BR was highly and positively correlated with the OC-Mpom content in eroding and depositional areas but not in transport areas. On the other hand, negative correlations between BR and the ~~protected-OC~~ stabilized in macroaggregates ( $r = -0.40$ ;  $p=0.01$ ) and the ~~degree of microbial degradation index~~ ( $r = -0.60$ ;  $p=0.00$ ) across the study areas were obtained.

## 4. Discussion

### 4.1 Dynamics of OC in the eroding areas

The differences among the sources of the sediments, in terms of aggregation and OC distribution within aggregates, determined the way in which the sediment and associated OC moved across the catchment, which is in line with results given by some authors reporting that aggregation considerably reduces the potential transport distance of eroded OC and hence potentially skews its re-distribution in watersheds towards terrestrial deposition (Hu et al., 2016).

The distribution of OC within aggregates (the OC content increased with increasing aggregate size) observed in the forest soils, together with the high percentage of occluded microaggregates within macroaggregates (Mm), rich in OC, indicates a hierarchical order of aggregation in which macroaggregates are the nucleus for microaggregate formation (Oades, 1984). Other authors (Sodhi et al. 2009; Wang et al., 2011) also reported a higher OC content in macroaggregates than in microaggregates in soils, which means that organic matter could be the major binding agent in such soils (Oades and Waters, 1991). Here, the agricultural soils displayed a lower aggregate stability and total OC content than the forest soils, but the same pattern of OC distribution within aggregates. Moreover, in the agricultural soils, the OC content in the free mineral fraction (OC-min) was higher than that in the occluded fractions (OC-Mm and OC-Mmin) (Fig. 4b). Altogether, this indicates the perturbation of these agricultural soils by land use change, tillage, and water erosion which is also supported by

Con formato: Inglés (Estados Unidos)



the higher proportion of OC resistant to oxidation (Table 3) compared to forest soils, and indicates the high capacity of Mediterranean calcareous soils for OC stabilization in organo-mineral complexes, in which the OC is less susceptible to mineralization (Courtier-Murias et al., 2013; Trigalet et al., 2014; Garcia-Franco et al., 2015a). These stabilization mechanisms are common in Mediterranean areas and have been found in other sites, close to the study area (Garcia-Franco et al., 2014, 2015b). From a geomorphological perspective, the agricultural soils showed a more rigorous selection of the detached OC produced by the perturbations cited above; as a consequence, the most stable, passive pool remained in these soils.

Con formato: Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos)

In the fluvial lateral banks, a lack of hierarchical order of aggregation was found, together with a lower OC content in all the aggregate size classes, compared to the forest and agricultural soils, despite the fact that the three sources had similar percentages of M and Mm aggregates were observed. This indicates a decoupling of aggregates and OC, as found for other soils and land uses (Del Galdo et al., 2003; Deneff et al., 2007), and contrasts with Elliott (1986), who reported that the distribution of OC associated with the aggregate fractions is primarily controlled by the amount of soil present in the fraction. However, in our study, such decoupling is explained by the fact that the fluvial lateral banks were sampled on average at 80 cm depth, which is equivalent to a C-horizon with a lack of soil formation and slow OC accumulation being consistent with the significantly lower BR rates observed in this source of sediments, compared to those in the forest and agricultural soils, indicating very low microbial activity at this depth (Table 4). At the fluvial reach scale, the lateral banks were well connected to the channel, providing sediments to the main fluvial channels.

#### 4.2 Sediments and associated OC dynamics during transport

During transport, a strong selection of the texture and OC pools indicates that the most resistant OC was bound strongly to the mineral fraction (free and occluded), travelling the longest distances within the fluvial network. Compared to the bedload, the suspended sediments displayed a lower percentage of total macroaggregates (M) (30% and 10% for the bedload and suspended sediments, respectively), but a greater amount of OC associated with this fraction (decoupling), mainly as intra-aggregate particulate organic matter (Mpom) (Fig. 3a, Fig. 3b), suggesting mobilization of the most labile OC once the macroaggregates had been broken and resulting in less protection by physical and chemical processes. Other authors have reported how erosion enhances the release of easily mineralizable C encapsulated within aggregates in the mineralization process (Six et al., 2004; Polyakov and Lal, 2008; Van Hemelryck et al., 2010; Wang et al., 2014; Nie et al., 2018) and how that labile OC is also more easily transported in suspension (Starr et al., 2000). On the other hand, the high labile OC (OC-Mpom) content in suspended sediments is consistent with the overall relatively higher BR rate and C:N ratio, and lower aggregation and macroaggregate stabilization index, compared to the eroding and depositional areas (Tables 2 and 4), indicating that mineralization might be predominant during sediment transport.

Depletion of the total OC in suspended and bedload sediments of about 66% and 80%, respectively, was observed when compared to the richest sediment source (forest soils). However, enrichments of OC in the suspended sediments and depletion in the bedload were observed compared to the agricultural soils and fluvial lateral banks. This, together with the positive correlation between total OC and clay ( $r=0.48$ ;  $p<0.05$ ) and the higher clay content and lower aggregation observed in sediments during transport, demonstrates the selectivity of erosion during detachment and transport, as other authors have also reported (Starr et al., 2000).

Textural comparison of the sediment sources and sinks indicates that coarse material with low aggregation and a low OC content is selectively transported as bedload, while the finest and most labile material with a high OC content continues in transport as suspended load. Part of this OC might be i) mineralized before deposition, ii) mineralized once it has been deposited and before it is buried by following flooding events (Stallard et al., 1998), or iii) transported longer distances. The similar presence of stable OC in macroaggregates (total occluded OC: OC-Mm + OC-Mmin), ~~microbial degradation index (an indicator of OC stability/decomposability)~~ and OC resistant to oxidation (the most passive pool) in the transported sediments and the most erodible sources (agricultural soils and fluvial lateral banks; Table 3, Figure 4B see Table 4) indicates that the sediment came mainly from these eroding areas and that the more stable and resistant OC was not mineralized during transport. These results highlight the important role of both physical and chemical mechanisms in the protection of OC in transported sediments in semiarid and sub-humid climates, where erosion can be a significant contributor to the regulation of catchment C budgets.

#### 4.3 Sediments and associated OC dynamics in depositional areas

Compared to the sediments coming from the poorest sources of OC within the catchment (i.e., agricultural soils and fluvial lateral banks) and to the transported sediments, a significant increase in the total OC, mainly associated with macroaggregates (OC-M;  $> 250 \mu\text{m}$ ) and microaggregates within macroaggregates (OC-Mm), was observed at the surface layer of the depositional areas. This increase, ~~much~~ greater in the alluvial wedges than at the reservoir, supports the idea of new macroaggregate formation following the breakdown of aggregates during transport. Moreover, in the alluvial wedges, the OC concentration in these newly formed macroaggregates was higher than that in the microaggregates within macroaggregates in the agricultural soil and fluvial lateral bank sources, suggesting that aggregate formation and OC sequestration processes are occurring at the surface layer of these depositional areas. In fact, in these depositional areas, the distribution of OC within aggregates was similar to that observed in the forest soils showing a decrease in the OC content with decreasing aggregate size, reinforcing the idea that OC accumulation occurs in the sediments of depositional sites. The formation of new aggregates, providing physical stabilization for eroded soil organic carbon in depositional positions, has been reported elsewhere (Berhe et al., 2012).

Con formato: Fuente: Sin Negrita, Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos)

The similar (at the reservoir) or even higher (in the alluvial wedges) BR rates and enrichment of OC, compared to their main eroding sources (agricultural soils and fluvial lateral banks), indicate that new aggregates are being formed. Likewise, the positive correlations of the active pool (M<sub>pom</sub>) with the percentages of M and M<sub>m</sub> at the reservoir, and with total OC-M and OC-M<sub>m</sub> in both types of deposits, stress the role of the labile material in the activation of aggregate formation, as was observed also in the eroding areas. The formation of new aggregates might be favored by the growth of roots and the stimulation of microorganisms stimulated by the root exudates of established terrestrial and aquatic vegetation of terrestrial and aquatic origin at these depositional areas (Boix-Fayos et al., 2015). The formation of microaggregates within larger aggregates driven by microorganisms protects the OC associated with these microaggregates by increasing its turnover time, and leads to long-term C sequestration (García-Franco et al., 2015). Strong positive correlations between the OC-M<sub>pom</sub> and the occluded oxidable mineral fraction (rOC-M<sub>min</sub> minus OC-M<sub>min</sub>) at the reservoir, similar to the correlations in the eroding areas (Table 5), suggest that, at this site, fresh OC inputs are more rapidly transformed by microorganisms in an oxidable pool, promoting aggregate formation and supporting the idea that the absorption of OC into the mineral surfaces (silt and clay) by chemical processes could enhance physical protection and thus long-term OC preservation (Kennedy et al. 2002, Kirkles et al 2014, García Franco et al., 2015). Von Lützow et al. (2007) found that organic molecules stabilized by strong molecular interactions with mineral surfaces decomposed more slowly than OC stabilized by physical mechanisms (e.g. occlusion in soil aggregates). A lack of difference between the OC-M<sub>m</sub> and OC-M<sub>min</sub> contents (Fig. 4b) indicates that both physical and chemical processes might be important with regard to the reservoir sediments acting as a long-term residence deposits.

The higher percentages of M<sub>pom</sub> and M<sub>m</sub>, but lower OC content associated with them, at the reservoir, compared to the alluvial wedges at the surface layers (Fig. 4a, Fig. 4b), suggest differences in the temporal dynamics of aggregate formation between these two types of sediment deposit. The aggregate formation is slower in the alluvial wedges than at the reservoir, where the OC is also much more protected from decomposition by microorganisms than in the alluvial wedges (higher values of the protected-stabilized OC in macroaggregates; Table 4). In fact, at the reservoir, the OC occluded in aggregates (M<sub>m</sub>) represented about 20% more of the total OC, while BR rates were 66% lower (Table 4), than in the alluvial wedges. Altogether, these results indicate greater physico-chemical protection of the OC in the long-term residence deposits, compared to the medium-term ones.

In the deep layers of the depositional areas, the relatively lower OC, mineralization rates, and M<sub>pom</sub> were less favorable for aggregate formation and the process unfolded very slowly compared to the upper layers, which confirms the results reported by other authors (Xie et al., 2017). The reduction in BR rates with depth suggests that the OC has less chance of being released, which reinforces the C sink potential of these deposits at the deep layers where higher concentration of OC resistant to oxidation in the occluded fraction, relative to the free mineral fraction, and a high degree of microbial degradation index, indicate a higher OC stabilization (lower OC decomposability) compared to the upper layers (Table 3). Moreover, the

Con formato: Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos)

Con formato: Fuente: Sin Negrita, Inglés (Estados Unidos)

Con formato: Fuente: Sin Negrita, Inglés (Estados Unidos), Sin Resaltar

Con formato: Inglés (Estados Unidos)

Con formato: Fuente: Sin Negrita, Inglés (Estados Unidos)

~~similarity of the microbial degradation indices for the deep sediments and the sediment sources (agricultural soils and fluvial lateral banks) and for the suspended and bedload sediments indicates once again that the sediments come from the identified sources distributed across the catchment and that the buried topsoil might not experience any substantial change before burial.~~

Con formato: Inglés (Estados Unidos)

5 In summary, the medium and long-term depositional areas identified seem to be not only the main sinks for OC coming from different sources, but also areas of aggregate formation due to the microorganism activity stimulated by fresh OC inputs of established terrestrial and aquatic vegetation~~different origins~~. Our results are in agreement with those of Doetterl et al. (2016), who indicated that depositional landform positions are not only able to store large OC stocks but also preserve OC more effectively when compared to eroding landscape positions.

Con formato: Inglés (Reino Unido)

## 10 Conclusions

15 Physico-chemical mechanisms favoring OC stabilization in the sources or eroding areas and the connectivity between the latter and channel areas determine both the redistribution of OC within catchments and the C-dynamics. Good physico-chemical protection of OC in the original sediment sources results in better protection of OC in sediments during fluvial transport and deposition downstream. In ephemeral or intermittent Mediterranean streams similar to our study site, sediments often originate from agricultural soils and fluvial lateral banks, in which the physico-chemical protection of OC is much lower than that of forest soils less sensitive to erosion.

20 Different stabilization mechanisms were detected in the different sediment sources, transported sediments, and sedimentary deposits: (i) the predominance of physical mechanisms of OC stabilization in forest soils and alluvial wedges; and (ii) the predominance of chemical protection of OC in fluvial lateral banks. In the other positions, both processes are equally important in the stabilization of OC.

New processes of soil formation in the upper layers of the depositional areas and highly protected and stabilized OC stored in the deeper layers (mainly in the long-term deposits) strength the important role of depositional areas in erosive catchments, compensating OC losses from eroded sources and functioning as C sinks.

25 ~~The OC stored in depositional areas is even more protected and stabilized than the OC of the most active sediment sources (agricultural soils and fluvial lateral banks). New processes of soil formation in these deposits strengthen the role of these areas as C sinks. These results imply that both temporary and permanent sediment deposits within the catchment have an important role in erosive areas, compensating OC losses from the eroded sources and functioning as C sinks.~~

These results underline the importance of studying soil erosion, soil formation, and geomorphological processes together in semiarid and sub-humid catchments, where intermittent fluvial courses are predominant. Good management of these environments will be a powerful tool for climate change mitigation, given the high potential of alluvial settings as C sinks.

#### **Data availability**

5 All relevant data are presented within the manuscript.

#### **Authors contribution**

CBF, MA, MMM and JDV performed the field experimental design and field sampling. MA designed and performed the laboratory incubations, including the data-processing and analysis; NGF and MMM developed the organic matter  
10 fractionation techniques, EG performed them at the laboratory, and MA performed data-processing and analysis together with MMM; MMM wrote the manuscript with contributions from all authors.

#### **Competing interests**

The authors declare that they have no conflicts of interest.

15

#### **Acknowledgements**

This work was financially supported by the project DISECO (CGL2014-55-405-R) from the Spanish Government, National Plan of Science. We thank the members of the Soil and Water Conservation Group (CEBAS-CSIC) who helped us in the field and laboratory work. María Almagro was supported by the Juan de la Cierva Program (Grant IJCI-2015-23500).  
20 Carolina Boix-Fayos was also supported by a project from the program “Salvador de Madariaga” (PRXI7/00045) (Ministry of Education, Culture and Sport of Spain) and a project (20186/EE/17) of the Fundación Séneca (Regional Agency of Science of the Murcia Region), in the program “Jiménez de la Espada”.

#### **References**

25 Barreto, R.C., Madari, B.E., Maddock, J.E.L., Franchini, J., Costa, A.R.: The impact of soil management on aggregation, carbon stabilization and carbon loss as CO<sub>2</sub> in the surface layer of a Rhodic Ferralsol in Southern Brazil, *Agriculture, Ecosystems and Environment*, 132, 243-251. doi:10.1016/j.agee.2009.04.008, 2009.

Berhe, A.A., Harden, J.W., Torn, M.S., (...), Burton, S.D., Harte, J.: Persistence of soil organic matter in eroding versus depositional landform positions, *Journal of Geophysical Research: Biogeosciences*, 117, G02019,  
30 doi:10.1029/2011JG001790, 2012.

Berhe, A.A., Kleber, M.: Erosion, deposition, and the persistence of soil organic matter: Mechanistic considerations and problems with terminology, *Earth Surface Processes and Landforms*, 38, 908-912, doi: 10.1002/esp.3408, 2013.

- Berhe, A.A., Barnes, R.T., Six, J., Marín-Spiotta, E.: Role of Soil Erosion in Biogeochemical Cycling of Essential Elements: Carbon, Nitrogen, and Phosphorus, *Annual Review of Earth and Planetary Sciences*, 46, 521-548, doi: 10.1146/annurev-earth-082517-010018, 2018.
- 5 Boix-Fayos, C., Barbera, G. G., López-Bermúdez, F., Castillo, V. M.: Effects of check dams, reforestation and land use changes on river channel morphology: Case study of the Rogativa catchment (Murcia, Spain), *Geomorphology*, 91, 103–123, doi: 10.1002/hyp.7115, 2007.
- Boix-Fayos, C., Nadeu, E., Quiñero, J.M., Martínez-Mena, M., Almagro, M., de Vente, J.: Sediment flow paths and associated organic carbon dynamics across a Mediterranean catchment, *Hydrological Earth Systems Science*, 19, 1209-1223, doi:10.5194/hess-19-1209, 2015.
- 10 Courtier-Murias, D., Simpson, A.J., Marzadori, C., Baldoni, G., Ciavatta, C., Fernández, J.M., López-de-Sá, E.G., Plaza, C.: Unraveling the long-term stabilization mechanisms of organic materials in soils by physical fractionation and NMR spectroscopy, *Agriculture, Ecosystems and Environment*, 171, 9– 18, doi: 10.1016/j.agee, 2013.
- Datry T, Larned, S.T., Tockner, K.: Intermittent Rivers: A Challenge for Freshwater Ecology, *BioScience Advance Access* 64, 229-235, doi: 10.1093/biosci/bit027, 2014.
- 15 De Clercq, T., Heiling, M., Dercon, G., Resch, C., Aigner, M., Mayer, L. Mao, Y., Elsen, A., Steier, P., Leifeld, J., Merck, R.: Predicting soil organic matter stability in agricultural fields through carbon and nitrogen stable isotopes. *Soil Biology and Biochemistry*, 88, 29-38, doi: 10.1016/j.soilbio.2015.05.011, 2015.
- De Girolamo, A.M., Bouraoui, F., Buffagni, A., Pappagallo, G., Lo Porto, A.: Hydrology under climate change in a temporary river system: Potential impact on water balance and flow regime. *River Research and Applications*, 33, 1219-1232, doi:10.1002/rra.3165, 2017.
- 20 Del Galdo, I. Six, J., Peressotti, A., Cotrufo, M.F.: Assessing the impact of land-use change on soil C sequestration in agricultural soils by means of organic matter fractionation and stable C isotopes, *Global Change Biology*, 9, 1204–1213, doi:10.1046/j.1365-2486.2003.00657, 2003.
- Denef, K.J., Six, J., Merckx, R., Paustian, K.: Carbon sequestration in microaggregates of no-tillage soils with different clay mineralogy, *Soil Science Society of America Journal*, 68, 1935-1944, ISSN: 0361-5995, 1435-0661, 2004.
- 25 Denef, K., Zotarelli, L., Boddey, R.M., Six, J.: Microaggregate-associated carbon as a diagnostic fraction for management-induced changes in soil organic carbon in two Oxisols, *Soil Biology & Biochemistry*, 39, 1165–1172, doi:10.1016/j.soilbio.2006.12.024, 2007.
- Diehl, T.H.: A modified siphon sampler for shallow water: U.S. Geological Survey Scientific Investigations Report 2007–5282, 11 p, 2008.
- 30 Doetterl, S., Six, J., Van Wesemael B., Van Oost, K.: Carbon cycling in eroding landscapes: geomorphic controls on soil organic C pool composition and C stabilization, *Global Change Biology*, 18, 2218–2232, doi: 10.1111/j.1365-2486.2012.02680.x, 2012.

- Doetterl, S., Berhe, A., Nadeu, E., Wang, Z., Sommer, M., Fiener, P.: Erosion, deposition and soil carbon: A review of process-level controls, experimental tools and models to address C cycling in dynamic landscapes, *Earth-Science Reviews* 154, 102–122, doi:10.1016/j.earscirev.2015.12.005, 2016.
- 5 Elliott, E.T.: Aggregate structure and carbon, nitrogen, and phosphorus in native and cultivated soils, *Soil Science Society of America Journal*, 50, 627-633, doi: 10.2136/sssaj1986.03615995005000030017x, 1986.
- Elliott, E.T., Palm, C.A., Reuss, D.E., Monz, C.A: Organic matter contained in soil aggregates from a tropical chronosequence: correction for sand and light fraction, *Agriculture, Ecosystems & Environment*, 34, 443-451, DOI: 10.1016/0167-8809(91)90127-J, 1991.
- 10 Garcia-Franco, N.; Wiesmeier, M., Goberna, M., Martínez-Mena, M., Albaladejo, J.: Carbon dynamics after afforestation of semiarid shrublands: implications of site preparation techniques, *Forest Ecology and Management*, 319, 107-115, doi: 10.1016/j.foreco.2014.01.043, 2014.
- Garcia-Franco, N., Martínez-Mena, M., Goberna, M., Albaladejo, J.: Changes in soil aggregation and microbial community structure control carbon sequestration after afforestation of semiarid shrublands, *Soil Biology & Biogeochemistry*, 87, 110-121, doi:10.1016/j.soilbio.2015.04.012, 2015a.
- 15 Garcia-Franco, N., Albaladejo J., Almagro, M., Martínez-Mena, M.; Beneficial effects of reduced tillage and green manure on soil aggregation and stabilization of organic carbon in a Mediterranean agroecosystem. *Soil & Tillage Research*, 153, 66-75. doi: 10.1016/j.still.2015.05.010, 2015b.
- Golchin, A, Clarke, P., Oades, JM., Skjemstad JO: The effects of cultivation on the composition of organic-matter and structural stability of soils, *Australian Journal of Soil Research*, 33, 975 – 993, doi:10.1071/SR9950975, 1995.
- 20 Gregorich, E.G., Greer, K.J., Anderson, D.W., Liang, B.C.: Carbon distribution and losses: Erosion and deposition effects, *Soil & Tillage Research*, 47, 291-302, doi: 10.1016/S0167-1987(98)00117-2, 1998.
- Hoffmann, T., Mudd, S. M., Van Oost, K., Verstraeten, G., Erkens, G., Lang, A., Middelkoop, H., Boyle, J., Kaplan, J. O., Willenbring, J., and Aalto, R.: Short Communication: Humans and the missing C-sink: erosion and burial of soil carbon through time, *Earth Surface Dynamics Discussions*, 1, 93–112, doi:10.5194/esurf-1-45-2013, 2013.
- 25 Howard, D. M., Howard, P. J. A.: Relationships between CO<sub>2</sub> evolution, moisture content and temperature for a range of soil types, *Soil Biology and Biochemistry*, 25, 1537-1537, doi:10.1016/0038-0717(93)90008-Y, 1993.
- Hu, Y., Kuhn, NJ.: Erosion-induced exposure of SOC to mineralization in aggregated sediment, *Catena*, 137, 517–525, doi: 10.1007/s10533-016-0211-y, 2016.
- 30 IUSS Working Group WRB. World Reference Base for Soil Resources 2014, Update 2015. International Soil Classification System for Naming Soils and Creating Legends for Soil Maps. World Soil Resources Reports No. 106, Rome: FAO. 2015.
- Kendall C, Silva SR, Kelly VJ. Carbon and nitrogen isotopic compositions of particulate organic matter in four large river systems across the United States, *Hydrological Processes*, 15:1301–1346, doi:10.1002/hyp.216, 2001.
- Kennedy MJ, Pevear DR, Hill RJ.: Mineral surface control of organic carbon in black shale, *Science*, 295:657–660. doi: 10.1126/science.1066611, 2002.

Con formato: Inglés (Estados Unidos)

Con formato: Inglés (Estados Unidos)

- Kirkels, F.M.S.A., Cammeraat L.H, Kuhn N.J.: The fate of soil organic carbon upon erosion, transport and deposition in agricultural landscapes — A review of different concepts, *Geomorphology*, 226, 94–105, doi:10.1016/j.geomorph.2014.07.023, 2014.
- Lal, R.: Soil erosion and the global carbon budget, *Environment International*, 29, 437–450, doi:10.1016/S0160-4120(02)00192-7, 2003.
- Lal, R.: Soil erosion and carbon dynamics. *Soil and Tillage Research*, 81, 137-142, doi: 10.1016/j.still.2004.09.002, 2005
- Liu, C., Li, Z., Chang, X, He, J., Nie, X., Liu, L., Xiao, H., Wang, D., Peng, H., Zeng, G.: Soil carbon and nitrogen sources and redistribution as affected by erosion and deposition processes: A case study in a loess hilly-gully catchment, China, *Agriculture, Ecosystems and Environment*, 253, 11–22, doi:10.1016/j.agee.2017.10.028, 2018.
- 10 Nadeu, E., de Vente, J. Martínez-Mena, M., Boix-Fayos, C. Exploring particle size distribution and organic carbon pools mobilized by different erosion processes at the catchment scale. *Journal of Soils and Sediments* 11, 667-678, doi: 10.1007/s11368-011-0348-1, 2011.
- Nadeu, E., Berhe, A.A., De Vente, J., Boix-Fayos, C.: Erosion, deposition and replacement of soil organic carbon in Mediterranean catchments: a geomorphological, isotopic and land use change approach, *Biogeosciences*, 9, 1099–1111, doi:10.5194/bg-9-1099-2012, 2012.
- 15 Nie, X., Li, Z., Huang, J., Liu, L., Xiao, H., Liu, C., Zeng, G.: Thermal stability of organic carbon in soil aggregates as affected by soil erosion and deposition, *Soil & Tillage Research*, 175, 82–90, doi: 10.1016/j.still.2017.08.010, 2018.
- Oades, J.M. Soil organic matter and structural stability: mechanisms and implications for management, *Plant and Soil*, 76, 319-337, 1984.
- 20 Oades, J.M., Waters, A.G.: Aggregate hierarchy in soils, *Australian Journal of Soil Research*, 29, 815-825, doi:10.1071/SR9910815, 1991.
- Paul, E.A, Morris, S.J., Boehm, S.: The determination of soil C pool sizes and turnover rates: biophysical fractionation and tracers. In: Assessment Methods for Soil Carbon (Advances in Soil Science) (eds Lal R, Kimble JM, Follett RF, Stewart BA), pp. 193–206. CRC Press LLC, Boca Raton, FL, USA. 2001.
- 25 Pérez-Cutillas, P., Cataldo, M.F., Zema, D., de Vente, J., Boix-Fayos, C.: Efectos de la revegetación a escala de cuenca sobre el caudal y la evapotranspiración en ambiente mediterráneo. *Cuenca del Taibilla (SE de España)*. Bosque 39, 119-129, doi: 10.4067/S0717-92002018000100011, 2018.
- Polyakov, V.O., Lal, R.: Soil organic matter and CO<sub>2</sub> emission as affected by water erosion on field runoff plots, *Geoderma* 143, 216–222, doi: 10.1016/j.geoderma.2007.11.005, 2008.
- 30 Quinton, J., Govers, G., Van Oost., Bardgett, R.: The impact of agricultural soil erosion on biogeochemical cycling, *Nature Geoscience*, 3, 311-314, doi: 10.1038/ngeo838, 2010.
- Quiñonero-Rubio, J.M., Nadeu, E., Boix-Fayos, C., de Vente, J: Evaluation of the Effectiveness of Forest Restoration and Check-Dams to Reduce Catchment Sediment Yield, *Land Degradation and Development*, 27, 1018-1031, doi:10.1002/ldr.2331, 2016.

Con formato: Alemán (Alemania)

Con formato: Español (alfab. internacional)



- Ran, L., Lu, X. X., Xin, Z.: Erosion-induced massive organic carbon burial and carbon emission in the Yellow River basin, China, *Biogeosciences*, 11, 945–959, doi:10.5194/bg-11-945-2014, 2014.
- Razafimbelo, T.M., Albrecht, A., Oliver, R., Chapuis-Lardy, L., Feller, C. Aggregate associated-C and physical protection in a tropical clayey soil under Malagasy conventional and no-tillage systems, *Soil & Tillage Research*, 98, 140-149, doi: 10.1016/j.still.2007.10.012, 2008.
- 5
- Salomé, C., Nunan, N., Pouteau, V., Lerch, T.Z., Chenu, C.: Carbon dynamics in topsoil and in subsoil may be controlled by different regulatory mechanisms, *Global Change Biology*, 16, 416-426, doi:10.1111/j.1365-2486.2009.01884.x, 2010.
- Schewe, J., Heinke, J., Gerten, D., Haddeland, I., Arnell, N.W., Clark, D.B., Dankers, R., Eisner, S., Fekete, B.M., Colón-González, F.J., Gosling, S.N., Kim, H., Liu, X., Masaki, Y., Portmann, F.T., Satoh, Y., Stacke, T., Tang, Q., Wada, Y., Wissler, D., Albrecht, T., Frieler, K., Piontek, F., Warszawski, L. and Kabat, P.: Multimodel assessment of water scarcity under climate change, *Proceedings of the National Academy of Sciences*, 111, 3245-3250, doi: 10.1073/pnas.1222460110, 2014.
- 10
- Six, J., Elliott, E.T., Paustian, K.: Soil structure and organic matter: I. Distribution of aggregate-size classes and aggregate-associated carbon, *Soil Science Society of American Journal*, 64, 681–689, doi: 10.2136/sssaj2000.642681x, 2000.
- 15
- Six, J., Bossuyt, H., Degryze, S., Denef, K.A.: history of research on the link between (micro)aggregates, soil biota, and soil organic matter dynamics, *Soil Tillage Research*, 79, 7–31, doi:10.1016/j.still.2004.03.008, 2004.
- Six, J., Paustian, K.: Aggregate-associated soil organic matter as an ecosystem property and a measurement tool, *Soil biology and Biochemistry*, 68, A4-A9, doi:10.1016/j.soilbio.2013.06.014, 2014.
- Sodhi, G.P.S., Beri, V., Benbi, D.K.: Soil aggregation and distribution of carbon and nitrogen in different fractions under long-term application of compost in rice–wheat system, *Soil & tillage Research*, 103, 412-418, doi:10.1016/j.still.2008.12.005, 2009.
- 20
- Stallard, R.F.: Terrestrial sedimentation and the carbon cycle: coupling weathering and erosion to carbon burial, *Global Biogeochemical Cycles*, 12, 231–257, doi:10.1029/98GB00741, 1998.
- Starr, G.C., et al.: Modeling soil carbon transported by water erosion processes, *Land Degradation and Development*, 11, 83–91, doi: 10.1002/(SICI)1099-145X(200001/02)11:1<83::AID-LDR370>3.0.C, 2000.
- 25
- Trigalet S., Van Oost, K. Roisin, C., van Wesemael, B.: Carbon associated with clay and fine silt as an indicator for SOC decadal evolution under different residue management practices, *Agriculture, Ecosystems and Environment*, 196, 1–9, doi: 10.1016/j.agee.2014.06.011, 2014.
- Van Hemelryck, H., Fiener, P., VanOost, K., Govers, G.: The effect of soil redistribution on soil organic carbon: an experimental study, *Biogeosciences*, 7, 3971–3986, doi.org/10.5194/bg-7-3971-2010, 2010.
- 30
- Van Hemelryck, H., Govers, G., Van Oost, K., Merckx, R.: Evaluating the impact of soil redistribution on the in situ mineralization of soil organic carbon, *Earth Surface Processes and Landforms*, 36, 427–438, doi:10.1002/esp.2055, 2011.

- Van Oost, K., Govers, G., Quine, T.A., Heckrath, G., Olesen, J.E., De Gryze, S., Merckx, R.: Landscape-scale modeling of carbon cycling under the impact of soil redistribution: the role of tillage erosion, *Global Biogeochemical Cycles*, 19, 1-13, doi:10.1029/2005GB002471, 2005.
- 5 Van Oost, K., Quine, T. A., Govers, G., De Gryze, S., Six, J., Harden, W.J., Ritchie, J. C., McCarty, G. W., Heckrath, G., Kosmas, C., Giraldez, J. V., Marques da Silva, J. R., Merckx, R.: The Impact of Agricultural Soil Erosion on the Global Carbon Cycle, *Science New Series*, 318, 626-629, doi: 10.1126/science.1145724, 2007.
- Vadher, A.N., Millett, J., Stubbington, R., Wood, P.J.: Drying duration and stream characteristics influence macroinvertebrate survivorship within the sediments of a temporary channel and exposed gravel bars of a connected perennial stream, *Hydrobiologia*, 814, 121-132, doi:10.1007/s10750-018-3544-9, 2018.
- 10 von Lützow, M., Kögel-Knabner, I., Ekschmitt, K., Flessa, H., Guggenberger, G., Matzner, E., Marschner, B.: SOM fractionation methods: relevance to functional pools and to stabilization mechanisms. *Soil Biology and Biochemistry* 39, 2183–2207, doi:10.1016/j.soilbio.2007.03.007, 2007.
- 15 Wang, Z., Govers, G., Steegen, A., Clymans, W., Van den Putte, A., Langhans, C., Merckx, R., Van Oost, K.: Catchment-scale carbon redistribution and delivery by water erosion in an intensively cultivated area, *Geomorphology*, 124, 65–74, doi:10.1016/j.geomorph.2010.08.010, 2010.
- Wang, X., Liu, G., Liu, S.: Effects of gravel on grassland soil carbon and nitrogen in the arid regions of the Tibetan Plateau, *Geoderma*, 166, 181–188, doi:10.1016/j.geoderma.2011.07.028, 2011.
- Wang, X., Cammeraat, E., Cerli, C., Kalbitz, K.: Soil aggregation and the stabilization of organic carbon as affected by erosion and deposition, *Soil Biology and Biochemistry*, 72, 55-65, doi:10.1016/j.soilbio.2014.01.018, 2014.
- 20 Wissler, D., Frohling, S., Hagen, S., Bierken M.F.P: Beyond peak reservoir storage? A global estimate of declining water storage capacity in large reservoirs, *Water Resources Research*, 49, 5732–5739, doi: 10.1002/wrcr.20452, 2013.
- Xie, J., Hou, M., Zhou, Y., Wang, R., Zhang, S., Yang, X., Sun, B.: Carbon sequestration and mineralization of aggregate-associated carbon in an intensively cultivated Anthrosol in north China as affected by long term fertilization, *Geoderma*, 296, 1–9, doi: 10.1016/j.geoderma.2017.02.023. 2017.
- 25 Zimmermann, M., Leifeld, J., Schmidt, N.W.I., Smith, P., Fuhrer, J.: Measured soil organic matter fractions can be related to pools in the RothC model. *European Journal of Soil Science*, 58, 658-667, doi:10.1111/j.1365-2389.2006.00855.x, 2007.

Table 1. Characteristics and representativeness of soil and sediment samples at the different morphological positions representing the different phases of the erosion process across the catchment.

<b>Morphological Position</b>	<b>Erosion phase</b>	<b>Sampling sites or processes</b>	<b>Depth (cm)</b>	<b>Sample (n)</b>
Eroding areas (sediment sources)	Detachment	Forest soils*	0-10	9
		Agricultural soils	0-10	4
		Fluvial lateral bank	80	5
Transport areas (main channel)	Transport	Suspended load	Not applicable	9 (5 events)
		Bedload**	0-10	24
Depositional areas: Along the channel and downstream	Deposition	Alluvial wedges	0-40	12
			40-80	4
			0-40	8
		Reservoir	40-300	14

\*Include a variety of soils representing the main land covers in the catchment: forests, shrublands and grasslands (3, 4 and 2 samples, respectively). \*\*include alluvial bars and channel (18 and 6 samples, respectively).

Table 2. Soil texture, MWD and total organic carbon (OC) in different morphological positions across the catchment: eroding, transport and depositional areas.

Morphological positions	Texture (%)			MWD (mm)	Total OC (g kg <sup>-1</sup> )
	Clay	Silt	Sand		
Forest soil	17.1±1.9 <sup>b</sup>	51.9±3.8 <sup>a,b,c</sup>	30.1±5.72 <sup>b</sup>	0.96±0.26 <sup>c</sup>	22.1±8.9 <sup>b</sup>
Agricultural soil	18.9±3.2 <sup>b</sup>	58.6±8.5 <sup>b,c</sup>	22.4±11.1 <sup>b</sup>	0.26±0.05 <sup>b</sup>	6.8±0.9 <sup>b</sup>
Fluvial lateral bank	15.9±3.5 <sup>b</sup>	43.2±7.3 <sup>b</sup>	38.2±10.0 <sup>b</sup>	0.39±0.08 <sup>b</sup>	3.4±1.07 <sup>a</sup>
<b>Eroding areas</b>	<b>17.2±1.5<sup>B</sup></b>	<b>50.9±3.4<sup>A</sup></b>	<b>30.6±4.5<sup>A,B</sup></b>	<b>0.63±0.2<sup>B</sup></b>	<b>13.4±4.8<sup>B</sup></b>
Suspended load	34.1±5.55 <sup>c</sup>	61.7±4.8 <sup>c</sup>	4.0±1.47 <sup>a</sup>	0.22±0.04 <sup>b,a</sup>	7.5±0.5 <sup>b</sup>
Bedload	9.9±0.6 <sup>a</sup>	39.1±2.3 <sup>a</sup>	50.4±2.9 <sup>c</sup>	0.41±0.05 <sup>c,b</sup>	4.3±0.3 <sup>a</sup>
<b>Transport areas</b>	<b>16.5±2.4<sup>A</sup></b>	<b>45.2±2.7<sup>A</sup></b>	<b>37.7±4.2<sup>B</sup></b>	<b>0.39±0.04<sup>A</sup></b>	<b>5.2±0.3<sup>A</sup></b>
Alluvial wedge surface	10.9±0.4 <sup>a</sup>	69.2±1.9 <sup>c</sup>	19.8±2.17 <sup>b</sup>	0.73±0.04 <sup>c</sup>	12.8±1.8 <sup>b</sup>
Alluvial wedge deep	14.9±1.9 <sup>a,b</sup>	76.0±3.2 <sup>c</sup>	8.9±1.49 <sup>a</sup>	0.23±0.02 <sup>b</sup>	7.6±0.5 <sup>a</sup>
<b>Alluvial wedges</b>	<b>11.9±0.7<sup>A</sup></b>	<b>70.9±1.7<sup>B</sup></b>	<b>17.1±2.04<sup>A</sup></b>	<b>0.60±0.06<sup>B</sup></b>	<b>11.5±1.4<sup>B</sup></b>
Reservoir surface	14.7±0.8 <sup>b</sup>	60.6±0.9 <sup>b,c</sup>	24.6±1.2 <sup>b</sup>	0.76±0.45 <sup>c</sup>	9.26±1.16 <sup>b</sup>
Reservoir deep	18.0±0.9 <sup>b</sup>	61.9±0.9 <sup>c</sup>	20.0±1.8 <sup>b</sup>	0.14±0.01 <sup>a</sup>	5.9±0.4 <sup>a,b</sup>
<b>Reservoir</b>	<b>16.8±0.7<sup>B</sup></b>	<b>61.5±0.7<sup>A</sup></b>	<b>21.7±1.3<sup>A,B</sup></b>	<b>0.37±0.06<sup>A</sup></b>	<b>7.1±0.6<sup>A,B</sup></b>

Numeral values are means ± standard errors. Columns with different lowercase letters indicate significant differences among sites or processes. Columns with different uppercase letters means significant differences between big pooled groups (p < 0.05), according to Kruskal Wallis test.

Table 3. OC resistant to NaOCl oxidation in the free mineral fraction (rOC-min) and occluded mineral fraction (rOC-Mmin); and contribution of rOC-min and rOC-Mmin to the total mineral fraction in the different morphological positions across the catchment: eroding, transport and depositional areas.

	OC resistant to NaOCl oxidation			
	rOC-min	rOC-Mmin	rOC-min	rOC-Mmin
	g 100 g <sup>-1</sup> aggregate		Contribution to the total mineral fraction (%)	
<b>Eroding</b>				
Forest soil	0.55±0.25 <sup>b</sup>	0.35±0.11 <sup>b</sup>	15.33±4.5	3.58±1.25
Agricultural soil	0.29±0.01 <sup>b</sup>	0.29±0.01 <sup>b</sup>	28.87±2.6	1.24±0.53
Fluvial lateral bank	0.13±0.02 <sup>aA</sup>	0.29±0.03 <sup>bB</sup>	40.04±21.22	12.17±8.03
<b>Transport</b>				
Suspended load	0.21±0.1 <sup>b</sup>	0.31±0.02 <sup>b</sup>	24.24±11.47	1.44±1.35
Bedload	0.13±0.02 <sup>aA</sup>	0.22±0.02 <sup>bB</sup>	18.71±3.01	0.72±0.15
<b>Deposition</b>				
Alluvial wedge surface	0.23±0.04 <sup>b</sup>	0.29±0.02 <sup>b</sup>	27.55±6.49	1.56±0.41
Alluvial wedge deep	0.17±0.04 <sup>b</sup>	0.29±0.01 <sup>b</sup>	20.76±7.33	1.22±0.05
Reservoir surface	0.19±0.04 <sup>b</sup>	0.26±0.02 <sup>b</sup>	16.92±1.39	2.01±0.41
Reservoir deep	0.12±0.01 <sup>aA</sup>	0.16±0.001 <sup>aB</sup>	11.93±0.94	0.62±0.09

5 Numerical values are means ± standard errors. Columns with different lower case letters indicate significant differences among sites or processes. ( $p < 0.05$ ) according to Kruskal Wallis test. Rows with different capital letters means significant differences between OC fractions within each site or process.

10

15

Table 4. C:N ratios, basal respiration rate, and indicators of OC ~~macroaggregates stabilization~~ OC stabilized in macroaggregates (OC-Mm+OC-Mmin/OC-Mpom) ~~and microbial degradation rate (OC-m+OC-min/OC-Mm+OC-Mmin+OC-Mpom-M)~~ in the different morphological positions across the catchment: eroding, transport and depositional areas.

	C:N Ratio	Basal Respiration (mg CO <sub>2</sub> -kg <sup>-1</sup> soil/sed day-1)	<del>Protected-OC</del> <u>OC stabilized in macroaggregates*</u>
<b>Eroding areas</b>			
Forest soil	12.1±1.2 <sup>b</sup>	6.04±1.74 <sup>bc</sup>	0.88±0.3 <sup>b</sup>
Agricultural soil	8.8±0.7 <sup>a,b</sup>	2.59±0.29 <sup>bc</sup>	1.22±0.7 <sup>b</sup>
Fluvial lateral bank	9.5±2.8 <sup>a</sup> ,	0.81±0.18 <sup>a</sup>	1.09±0.4 <sup>b</sup>
<b>Transport areas</b>			
Suspended load	10.6±1.3 <sup>b</sup>	6.94±0.77 <sup>c</sup>	0.44±0.2 <sup>a</sup>
Bedload	8.2±0.5 <sup>a</sup>	3.25±0.53 <sup>b</sup>	0.98±0.2 <sup>b</sup>
<b>Depositional areas</b>			
Alluvial wedge surface	13.4±1.6 <sup>b</sup>	13.80±1.04 <sup>c</sup>	0.82±0.1 <sup>b</sup>
Alluvial wedge deep	8.4±2.1 <sup>a</sup>	2.83±1.04 <sup>a,b</sup>	1.40±0.3 <sup>b</sup>
Reservoir surface	8.5±0.6 <sup>a</sup>	4.58±1.47 <sup>b</sup>	5.53±2.8 <sup>c</sup>
Reservoir deep	8.9±2.3 <sup>a</sup>	0.70±0.02 <sup>a</sup>	11.47±1.8 <sup>c</sup>

5 Numeral values are means ± standard errors. Columns with different lower case letters indicate significant differences among sites or processes ( $p < 0.05$ ) according to Kruskal Wallis test. \* The higher the value the more stabilized OC in macroaggregates is. ;\*\* The higher the value the more stable (lower decomposability) OC is.

10

15

Table 5. Spearman correlation coefficients between the OC associated to the intra-aggregate POM (OC-Mpom) or basal respiration (BR) and aggregates percentage and associated total OC, MWD, occluded oxidable OC, CN ratios, ~~and protected OC stabilized in macroaggregates and degree of microbial degradation index~~ in the different geomorphological positions within the catchment: eroding, transport and depositional areas.

OC-Mpom (g kg <sup>-1</sup> soil/sed)	<b>Eroding</b>	<b>Transport</b>	<b>Depositional areas</b>	
	<b>areas</b>	<b>areas</b>	<b>Alluvial wedges</b>	<b>Reservoir</b>
Macroaggregates (%)	.82**	.04	.23	.77**
Micro within macro (%)	<b>.52</b>	<b>.39</b>	.04	.27
OC macroaggregates (g kg <sup>-1</sup> soil/sed)	.85**	.57**	.96**	.71**
OC micro within macro (g kg <sup>-1</sup> soil/sed)	.72**	.22	<b>.58</b>	.58*
MWD	.78**	.13	.30	.81**
Occluded Oxidable OC (g Kg <sup>-1</sup> soil/sed)	.76**	.28	.09	.61**

Basal respiration (mg C-CO <sub>2</sub> kg <sup>-1</sup> soil/sed day <sup>-1</sup> )	<b>Eroding</b>	<b>Transport</b>	<b>Depositional areas</b>	
	<b>areas</b>	<b>areas</b>	<b>Alluvial wedges</b>	<b>Reservoir</b>
Micro within macro (%)	.27	.11	.07	.61**
OC macroaggregates (g kg <sup>-1</sup> soil/sed)	.57*	.16	.95**	.55**
OC micro within macro (g kg <sup>-1</sup> soil/sed)	.46	.27	.13	.52*
CN	.43	.12	.65*	-.02
Occluded Oxidable OC (g Kg <sup>-1</sup> soil/sed)	<b>.51</b>	.10	.16	<b>.41</b>
OC-Mpom	.73**	.10	.68**	.75**
<del>Protected OC stabilized in macroaggregates</del>	<del>-.36</del>	<del>.13</del>	<del>-.61*</del>	<del>-.38</del>
<del>Degree of microbial degradation index</del>	<del><b>-70**</b></del>	<del><b>-.25</b></del>	<del><b>-.67</b></del>	<del><b>-.45</b></del>

5 \* . \*\*. bold: significant at p<0.05; p<0.001 and p< 0.10, respectively.

### Figure captions

Figure 1A). Location of the Turrilla catchment and main sampling areas selected for sampling of soils and sediments: 1) The Taibilla reservoir at the outlet of the catchment (as representative of long term depositional areas) where reservoir sediments were sampled; 2) Permanent stream (in blue; Turrilla subcatchment) and intermittent stream (in orange; Rogativa subcatchment). In this area alluvial bars (as bedload), suspended load, soils and lateral fluvial banks were sampled; (3) Middle and upstream areas where forest soils and alluvial wedges (as representative of medium term depositional areas behind check-dams) were sampled.

Figure 1B). Sediment cascade of the Turrilla catchment, with representation of sampling areas according to Table 1. Please notice that some depositional areas (mainly alluvial wedges behind check-dams) were also sampled in the middle and upstream areas. Some soils were also sampled downstream.

Figure 2. Description of the main steps and physico-chemical analysis (wet-sieving and oxidation) used to obtain the different aggregate sized fractions (square in red), the intra-aggregate organic matter, the occluded microaggregates and the occluded mineral fractions within macroaggregates (square in green), and fractions resistant to oxidation (square in blue).

Figure 3A. Water-stable aggregate size distribution (g aggregate 100 g<sup>-1</sup> soil): > 250 μm (macroaggregates: M), 63-250 μm (free microaggregates, m) and < 63 μm (free mineral fraction: min) in different geomorphological positions within the catchment: eroding, transport and depositional areas. Numeral values are means ± standard errors. Bars with different lowercase indicate significant differences in the percentage of each water-stable aggregate size among the different soils and sediments (p < 0.05) according to Kruskal Wallis test.

Figure 3B. Organic carbon content (g 100g<sup>-1</sup> aggregate): > 250 μm (macroaggregates: M), 63-250 μm (free microaggregates, m) and < 63 μm (free mineral fraction: min) in different geomorphological positions within the catchment: eroding, transport and depositional areas. Numeral values are means ± standard errors. Bars with different lowercase indicate significant differences in the percentage of each water-stable aggregate size among the different soils and sediments (p < 0.05) according to Kruskal Wallis test.

Figure 4A. Weight percentage (g 100 g<sup>-1</sup> aggregate) of the fractions occluded within macroaggregates (M): intra-aggregate particulate organic matter (Mpom), occluded microaggregates (Mm) and occluded mineral fraction (Mmin) in different geomorphological positions within the catchment: eroding, transport and depositional areas. Numeral values are means ± standard errors. Bars with different lowercase indicate significant differences in the percentage of each water-stable aggregate size among the different soils and sediments (p < 0.05) according to Kruskal Wallis test.



Figure 4B. Organic carbon content ( $\text{g } 100 \text{ g}^{-1}$  aggregate) associated to the occluded fractions within total macroaggregates (%): intra-aggregate particulate organic matter (Mpom), occluded microaggregates (Mm) and occluded mineral fraction (Mmin) in different geomorphological positions within the catchment: eroding, transport and depositional areas. Numeral values are means  $\pm$  standard errors. Bars with different lowercase indicate significant differences in the percentage of each

5 water-stable aggregate size among the different soils and sediments ( $p < 0.05$ ) according to Kruskal Wallis test.

10

15

20

25

30

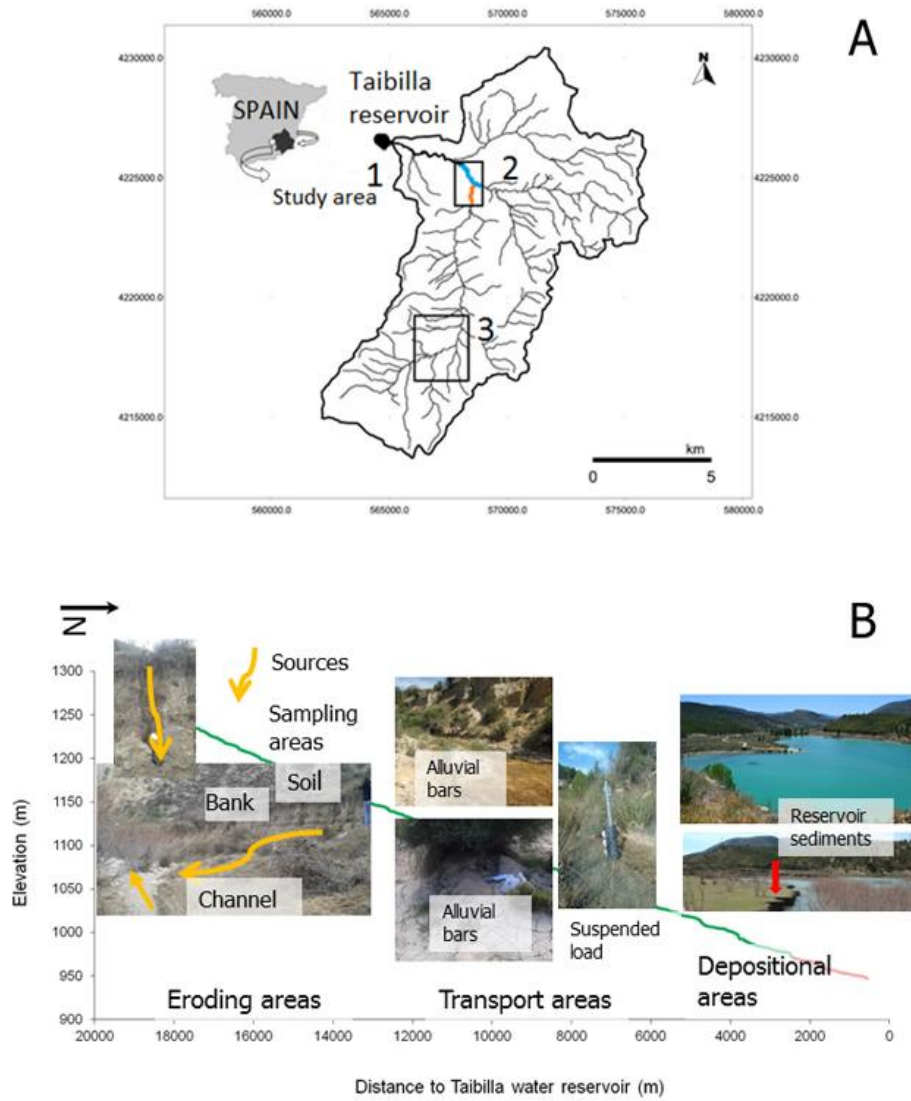
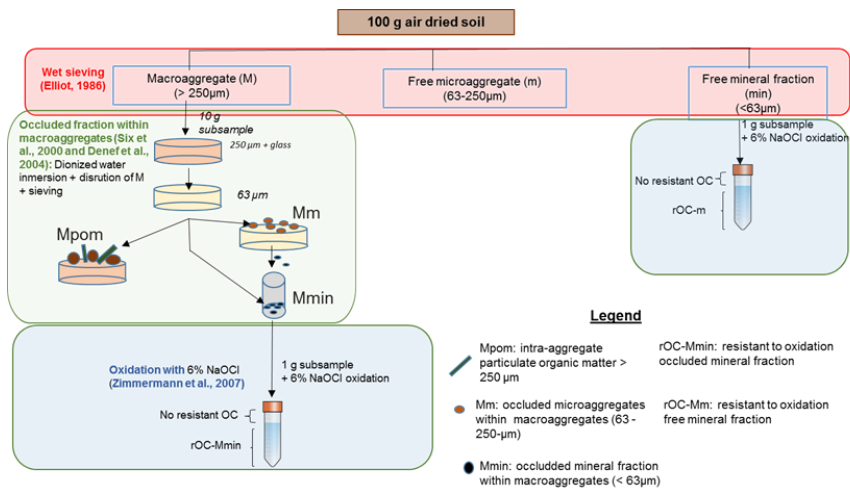


Figure 1

Figure 2



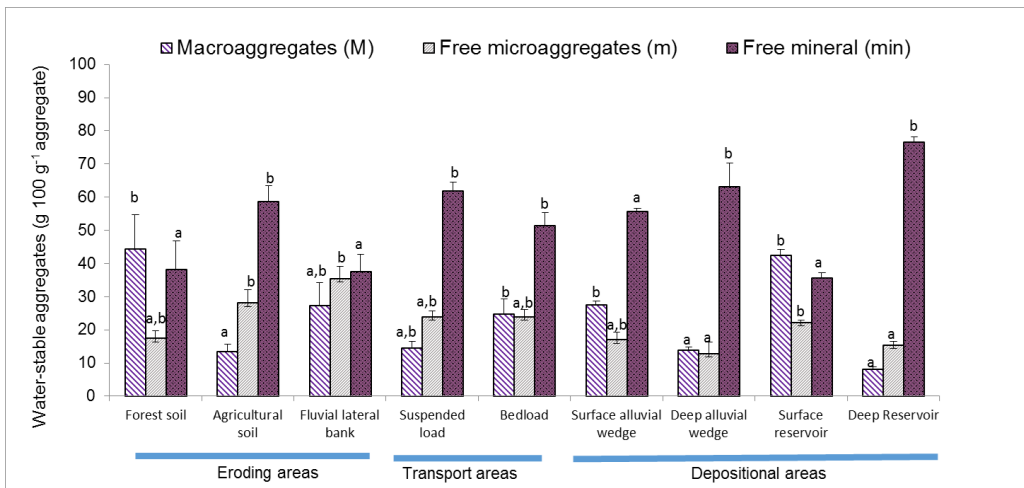
5

10

15

20

Figure 3A



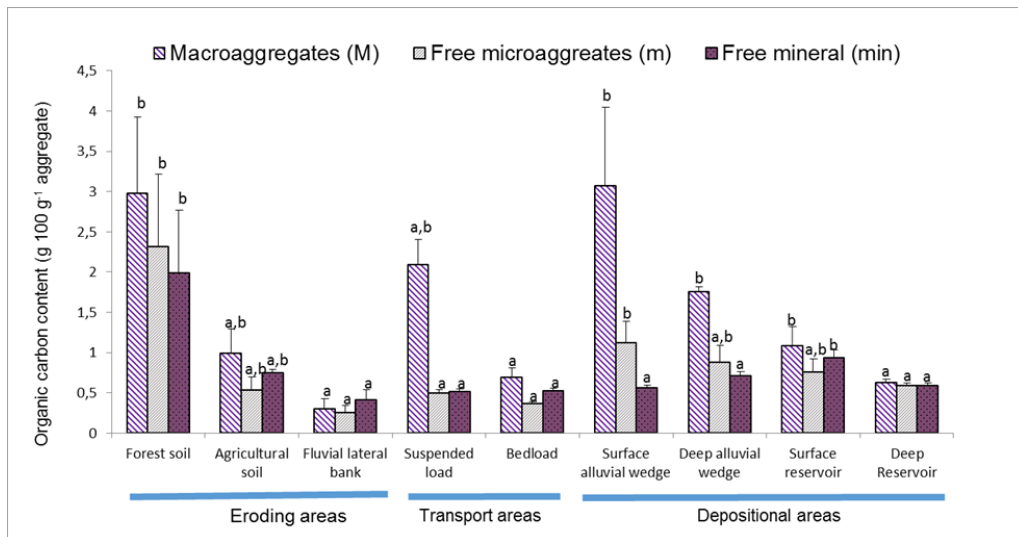
5

10

15

20

Figure 3B



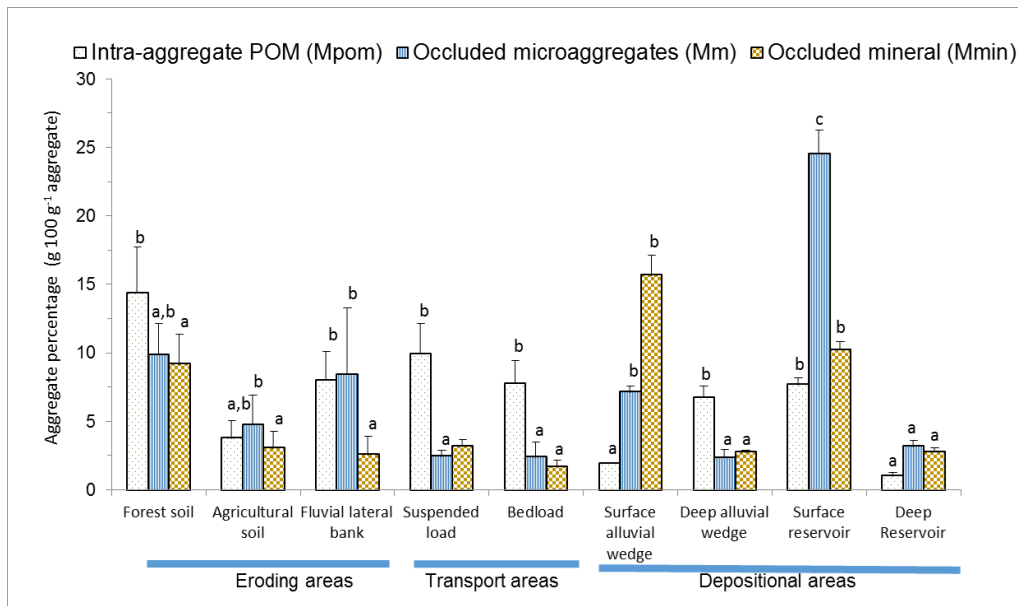
5

10

15

20

Figure 4A



5

10

15

Figure 4B

