Answers to comments of the referees

(C. Boix-Fayos, E. Nadeu, J.M. Quiñonero, M. Martínez-Mena, M. Almagro and J. de Vente).

We sincerely thank the editor Dr. Gooseff and Dr. Hoffmann and Referee 2 for their time reviewing this manuscript and for their constructive and positive comments. They have pointed out important aspects that we have fully taken into account and incorporated in a revised version. Besides, in addition to our previous reply to the reviewers' comments during the open discussion, in this letter we indicate how and where changes have been made. We attach following the answers, a manuscript where all changes are indicated in track changes. We appreciate the reviewers' effort going through the manuscript with detail. All their valuable suggestions have greatly contributed to improve it. The topics and the comments of the referees are presented in *italic* and our answers in standard fonts.

Main changes done to the manuscript:

1. More insight was provided in OC pools (POC:MOC ratios) and in the C:N ratios and their significance and interpretation in this context, adding emphasis that these indicators do not allow a final quantification of possible OC mineralization.
2. All figures have been changed following recommendations of the referees. A new Figure (Figure 5) was added illustrating POC:MOC ratios to explore further relations between OC pools as requested.
3. Data on microaggregated fractions have been reviewed and a new correlation matrix between OC pools and microaggregated particles was included in Table 3. The results on this part were also rewritten accordingly.
4. New references concerning the formation of organic carbon *in-situ* versus transport and deposition of allochthonous organic carbon in river systems were added and the discussion has been rewritten accordingly.
5. The results and discussion have been re-ordered following the suggestions of Dr. Hoffman, although part of the old structure was maintained as explained in the answers to referees comments.

Answers to Dr. Hoffmann (Reviewer 1)

1. Title: The title suggests that the authors are coupling the sediment flow-paths with organic carbon dynamics. I suggest to use an alternative title such as: 'Sediment associated organic carbon dynamics across a Med. catchment' or 'Organic carbon dynamics associated with soil erosion and sediment delivery in …'

Agree, title has been changed accordingly.

2. Abstract: The abstract summarizes the study very well. The final conclusion should be Strengthened.

OK. The last part of the abstract has been rewritten to strengthen the main conclusions.

3. Study site:
• If available you should give more details on rates and contributions of different processes (e.g. bank/gully erosion versus sheet/rill erosion).

We do not have quantitative data on rates of different erosion processes, however we have information and knowledge on the sedimentary dynamics of the channel in the last decades and its relation with the export of OC. More details are now added to the study site description on the relative contribution of different processes, based on some of our previous field and modelling studies and a qualitative geomorphological description of process dynamics (Boix-Fayos et al., 2007, Nadeu et al., 2011; 2012; 2014 and 2015; Quiñonero et al., 2014).

References:

Boix-Fayos, C., Barberá, G.G., López-Bermúdez, F., Castillo, V.M., 2007. 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.

Nadeu, E., de Vente, J., Martínez-Mena, M., Boix-Fayos, C., 2011. Exploring particle size distribution and organic carbon pools mobilized by different erosion processes at the catchment scale. J. Soil. Sediment. 11, 667-678.

Quiñonero-Rubio, J. M., Nadeu, E., Boix-Fayos, C., de Vente, J., 2014. Evaluation of the effectiveness of forest restoration and check-dams to reduce catchment sediment yield. Land Degradation & Development. Land Degradation and development DOI: 10.1002/ldr.2331

• Describe what you mean with non-selective erosion; and what is selective erosion?

Selective erosion refers to erosion of different grain sizes by different processes that may alter the composition of suspended sediments relative to the bulk source material. Through selective erosion finer grains are detached and transported over longer distances by overland and stream flows relative to coarser grain sizes (Kerr et al., 2011 and references therein). Selective erosion processes are mainly interrill and rill erosion affecting superficial soil layers. Non-selective erosion processes normally refer to massive erosion processes that also affect deeper soil layers and do not imply a selectivity of grain sizes, for instance bank erosion, channel erosion, gully erosion, and landslides.

A brief description of this definition is now included in the study area and illustrated with examples in the abstract.

References:

Kerr, J.G., Burford, M.A., Olley, J.M. Bunn, S.E., Udy,J., 2011. Examining the link between terrestrial and aquatic phosphorus speciation in a subtropical catchment: The role of selective erosion and transport of fine sediments during storm events. Water research 45, 3331-3340

• Improve Fig. 1, for a better representation of the study site, as suggested below.

Figure 1 has been split in two Figures, the location figure includes now the topography, rivers names and sampling areas, and Figure 2 includes the “cascade” conceptual approach with a new photo of the fluvial bars, as suggested

4. Methods:

• Motivate the fractionation; what do you expect for different fractions in terms of selective erosion and stability?

• Why do you use the IA and ASC? What is the expected impact of microaggregates on the lateral C flux?

A short paragraph justifying the study of the two fractions is now included in the methods section.

The motivation to introduce OC fractionation in our evaluation at the catchment scale was to define the preferential paths of movement of the different pools in comparison with the existing knowledge at the plot and hillslope scale. Given the differences in turnover times, it is of high interest to evaluate the behavior of POC and MOC separately, as this will give us insight in the long term stability of eroded carbon (more discussion on this can be seen in the first answer to the referees uploaded 12 September 2014).

The interest of measuring micro-aggregate size distribution in transported sediments arises from the fact that micro-aggregates provide physical protection to organic carbon (Jastrow and Miller, 1998; Polyakov and Lal, 2008), which is in agreement with our previous findings in eroded sediments measured in plots under different land uses (Martínez-Mena et al., 2012) and also in soils (García-Franco et al., 2014). So, in this study we aimed to evaluate what happens at a coarser scale in the fluvial system: do micro-aggregates break down or are they able to maintain the physical protection for OC over long transport distances? Our results (microaggregation indices) show that only a very small percentage of material is transported in aggregated form over long distances and in small aggregate sizes (Table 2) in suspended sediments. The percentages of aggregated material in the larger (250–63 µm) and medium micro-aggregate (63-20 µm) fractions of suspended sediments are ten and three times lower, respectively, to those in alluvial wedges (located in the upper part of the catchment). Thus, only a small percentage of micro-aggregated material resists over long transport distances (in suspended sediments). Furthermore, larger fractions and percentages of micro-aggregated material are found in sediments in the upper part of the catchment (alluvial wedges) probably due to short transport distances and re-aggregation of particles in deposited sediments.

Further research on the role of degree and sizes of micro-aggregation within suspended sediments associated to their organic carbon content will provide important insights on physical protection of OC in micro-aggregates during long transport distances. The IA correlates positively with TOC across all deposits, and it correlates also with the presence of POC across all deposits and in the alluvial wedges (Table 3).

References:

García-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. 2014

Jastrow, J.D., Miller, R.M., 1998. Soil aggregation stabilization ad carbon sequestration: feedback through organomineral associations. In: Lal, R., Kimble, J.M., Follet, R.F.(Eds.), Soil Processes and Carbon Cycle. CRC Press, Boca Raton, FL, pp. 207–223.

Martínez-Mena M., López, J., Almagro M., Boix-Fayos C. Albaladejo J. 2008. Effect of water erosion and cultivation on the soil carbon stock in a semiarid area of South-East Spain Soil & Tillage Research. 99, 119-129.

Martínez-Mena, M., López, J., Almagro, M., Albaladejo, J., Castillo, V., Ortiz, R., and Boix-25 Fayos, C. 2012. Organic carbon enrichment in sediments: effects of rainfall characteristics under different land uses in a Mediterranean area, Catena, 94, 36–42.

Polyakov, V.O. and Lal, R. 2008. Soil organic matter and CO2 emissions as affected by water erosion on field runoff plots. Geoderma 143, 216-222.

- ***State that C:N is used as a proxy of C-depletion. What happens to the N during transport?***

Description of the N contents in soils and sediments has been added to the results section (4.3), also further justification of the use of C:N ratios as a proxy for mineralization has been added in section 4.3 (more discussion on this in answer 7 to Referee 2). The discussion on the possible implications of a higher observed N content in the suspended load was further elaborated, emphasizing also the limitations in the interpretation of only C:N ratios without additional indicators for mineralization.

The use of the C:N ratio as an indicator for decomposition is based on the idea that decomposition of unprotected organic matter and its recycling by soil organisms leads to stabilization of organic matter residuals, while simultaneously to a decrease in their C:N ratios (Kramer et al., 2003). For this reason, C:N ratios have been extensively used alone or in combination with other proxies for mineralization. Some authors used C:N ratios as a mineralization proxy together with the $\delta^{15}N$, which increases with increasing stability during mineralization process because of N-rich organic compounds are increasingly utilized as a C source, while N excess is mineralized, resulting in ^{15}N enrichment of the remaining substrate (Conen et al., 2008 and references there in). Likewise, C:N ratios in combination with clay content and ^{13}C have been previously used as indicators of organic carbon mineralization in sediments along fluvial systems (Wang et al., 2010). We interpret high clay contents associated to low organic carbon (as occurs in our suspended sediment), as an additional indicator for losses of organic carbon, because organic carbon is, in general, positively associated to fine sediments (similar ratios of OC and clay between soils and sediments would indicate unimportant mineralization).

In our deposits, we observed a decrease in C:N ratios along the fluvial path, except for the alluvial wedges. When further exploring the patterns of C:N ratios, it can be observed that their decrease across sediment deposits is accompanied by a simultaneous decrease in total OC (Figure 2 in the paper) and N contents (graph below, data not shown in the paper), with the exception of suspended load, in which a higher N content with respect to those in the other deposits was observed. On the other hand (as also highlighted by reviewer 2), nitrogen dynamics along the fluvial path have been suggested to be very complex, and several of the involved process are still largely unknown (Robertson and Groffman, 2007). For instance, a previous study has suggested that N increases in suspended load can be due to the presence of ammonium (NH4+) in those sediments with high clay contents (as it is the case of suspended load) or to the contribution of fresh-water algae in suspended sediments (Sánchez- Vidal et al., 2013). The N cycle in sediments of intermittent rivers (as in our case) is even more complex because nitrification and denitrification processes are also dependent on the drying and rewetting cycles of sediments (Gómez et al., 2012; Arce et al., 2013). So, although, the C:N ratio is often used as indicator for C decomposition, there is still important uncertainty over its interpretation. Therefore in our study we used several supporting indicators regarding C decomposition as is now further explained in the text (for more details on this see response to reviewer 2).

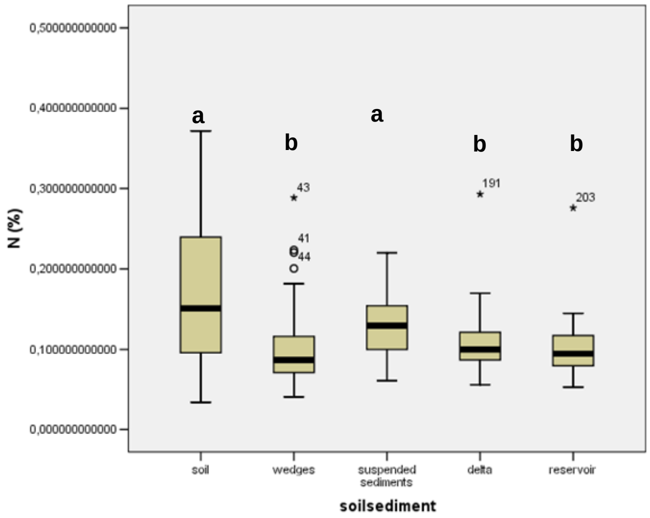


Figure 1. Box-whisker plots of the N concentration in soils and different sediment deposits Differences according to Kruskal-Wallis test ($p<0.05$).

References:

Arce, M. I., Gómez, R., Suárez, M. L., and Vidal-Abarca, M. R.: Denitrification rates and controlling factors in two agriculturally influenced temporary Mediterranean saline streams, Hydrobiologia, 700, 169-185, 2013.

Conen, F., Zimmermann, M., Leifeld, J., Seth, B., and Alewell, C.: Relative stability of soil carbon revealed by shifts in δ15N and C:N ratio, Biogeosciences, 5, 123-128, 2008.

Gómez, R., Arce, I. M., Sánchez, J. J., and Sánchez-Montoya, M. del Mar: The effects of drying on sediment nitrogen content in a Mediterranean intermittent stream: A microcosms study, Hydrobiologia, 679, 43-59, 2012.

Kramer, M. G., Sollins, P., and Sletten, R. S.: Soil carbon dynamics across a windthrow disturbance sequence in southeast Alaska, Ecology, 85, 2230-2244, 2004.

Robertson GP, Groffman PM (2007) Nitrogen transformations. In Paul EA (ed). Soil Microbiology, Biogeochemistry, and Ecology. Springer, New York, NY, USA, pp 341-364

Sanchez-Vidal, A., Higueras, M., Martí, E., Liquete, C., Calafat, A., Kerhervé, P., Canals, M., 2013. Riverine transport of terrestrial organic matter to the North Catalan margin, NW Mediterranean Sea. Progress in Oceanography, 118: 71-80.

Wang, Z., Govers, G., Steegen, A., Clymans, W., Van den Putte, A., Langhans, C., Merckx, R., and Van Oost, K.: Catchment-scale carbon redistribution and delivery by water erosion in an intensively cultivated area, Geomorphology, 124, 65-74, 2010.

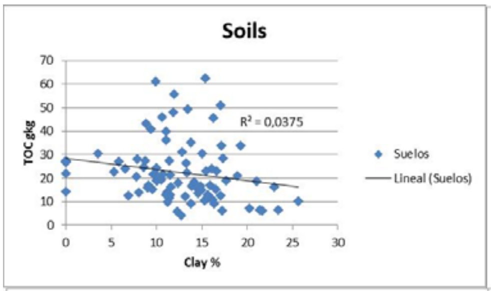
5. Results:

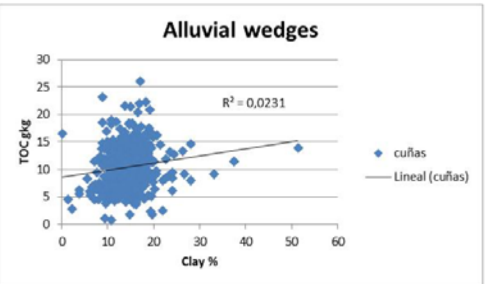
- ***I suggest to restructure the results presenting first the data on grain size and texture in different pools, secondly the OC and C:N and finally the link between the texture and OC and C:N.***

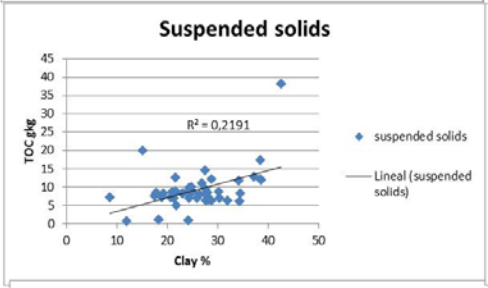
The presentation of the results are structured now following this suggestion.

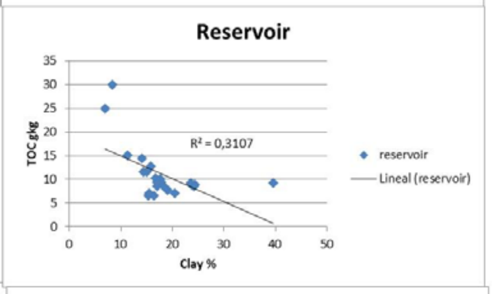
- ***In addition to Tab. 3, I would like to see scatter-plots of clay content and IA versus TOC, C:N etc. with different symbols for different pools***.

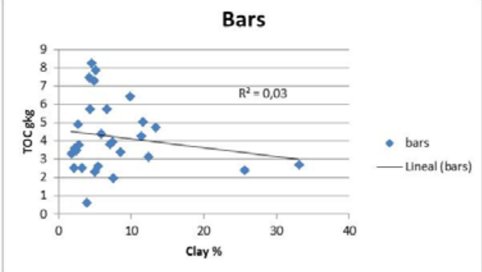
Below we have included several scatter plots for the exploration of the results as suggested. We have also reviewed and refined all the microaggregation data, and we produced a new correlation matrix. We have added the new correlation matrix to Table 3 (only the part referring microaggregation particles and indicators is new). Description of results on microaggregation correlations have been changed accordingly, although the main message was maintained. We have finally not included the scatter plots in the manuscript because we have added also two other figures to the reviewed version and we consider that the new correlation matrix gives also sufficient insight in the data.

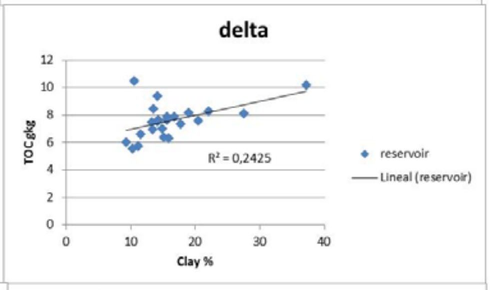

Soils
TOC g/kg
Clay %
0
10
20
30
40
50
60
70
5
15
25
R² = 0,0375
Suelos
Lineal (Suelos)

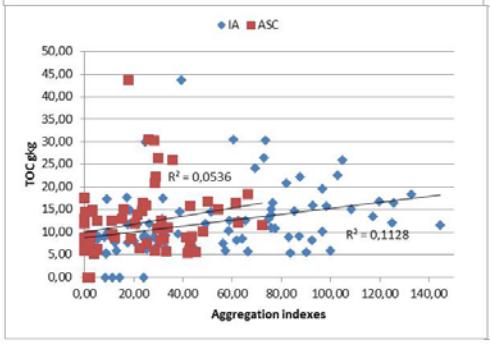

Alluvial wedges
TOC g/kg
Clay %
0
5
10
15
20
25
30
40
50
60
R² = 0,0231
cuñas
Lineal (cuñas)

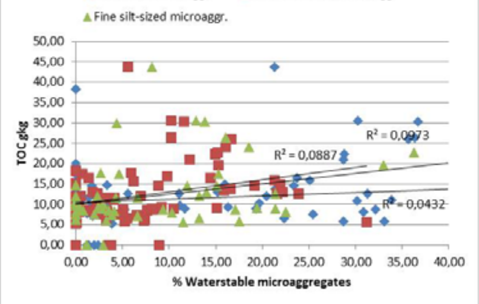

Suspended solids
TOC g/kg
Clay %
0
5
10
15
20
25
30
35
40
45
50
R² = 0,2191
suspended solids
Lineal (suspended solids)

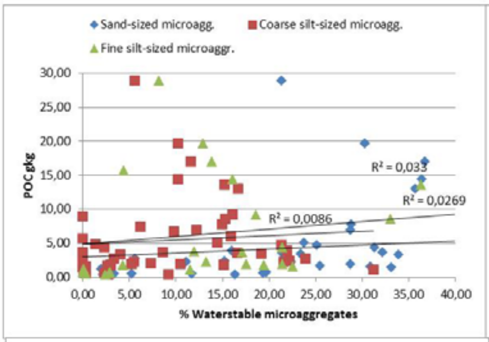

Reservoir
TOC g/kg
Clay %
0
5
10
15
20
25
30
35
40
50
R² = 0,3107
reservoir
Lineal (reservoir)

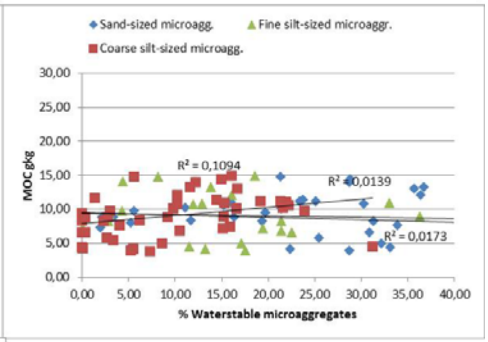

Bars
TOC g/kg
Clay %
0
1
2
3
4
5
6
7
8
9
10
20
30
40
R² = 0,03
bars
Lineal (bars)

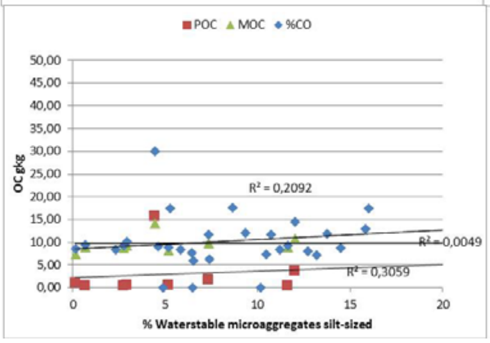

delta
TOC g/kg
Clay %
0
2
4
6
8
10
12
20
30
40
R² = 0,2425
reservoir
Lineal (reservoir)


IA
ASC
TOC g/kg
Aggregation indexes
0,00
5,00
10,00
15,00
20,00
25,00
30,00
35,00
40,00
45,00
50,00
20,00
40,00
60,00
80,00
100,00
120,00
140,00
R² = 0,0536
R² = 0,1128


Sand-sized microagg.
Coarse silt-sized microagg.
Fine silt-sized microaggr.
TOC g/kg
% Waterstable microaggregates
0,00
5,00
10,00
15,00
20,00
25,00
30,00
35,00
40,00
45,00
50,00
R² = 0,0973
R² = 0,0887
R² = 0,0432


Sand-sized microagg.
Coarse silt-sized microagg.
Fine silt-sized microaggr.
POC g/kg
% Waterstable microaggregates
0,00
5,00
10,00
15,00
20,00
25,00
30,00
35,00
40,00
R² = 0,033
R² = 0,0269
R² = 0,0086


Sand-sized microagg.
Fine silt-sized microaggr.
Coarse silt-sized microagg.
MOC g/kg
% Waterstable microaggregates
0,00
5,00
10,00
15,00
20,00
25,00
30,00
35,00
40,00
R² = 0,1094
R² = 0,0139
R² = 0,0173


POC
MOC
%CO
OC g/kg
% Waterstable microaggregates silt-sized
0,00
5,00
10,00
15,00
20,00
25,00
30,00
35,00
40,00
45,00
50,00
0
5
10
15
20
R² = 0,2092
R² = 0,0049
R² = 0,3059

• You compare the OC in soils (sampled in the upper 10cm) with the OC in wedges sampled down to 125cm. This is very problematic, since OC is typically much higher in the upper 20cm than below!

Yes, we are aware of that. Our assumption is that if all sediments are mobilized by superficial erosion processes (rill-interril) and no OC is lost during transport, than OC characteristics of sediment should be similar to those of source soils over the full depth of the sediment profile. If this is not the case, this is an indication that other erosion processes (gully, bank erosion) are dominant, or that other processes affect OC (selectivity o particles, mineralization etc.) during transport and deposition phases.

The sediments in the wedges until 125 cm have been progressively deposited in successive events, so to have a whole characterization of the sediment profile accounting for the variability among events, we sampled the whole sedimentary profile, assuming a minimum soil development in those wedges.

• You should give more information on the depth distribution of OC on slopes and in different deposits. Additionally, this should give information about the stability of C in the deposits (see e.g. Van Oost et al 2013, PNAS).

For soils, we currently have C data available for the topsoil (0-10 cm) throughout the catchment. However, for two small subcatchments within la Rogativa we have in-depth information for a selected number of soil profiles (Nadeu et al. 2012). For sediments, we have added in-depth information for various alluvial wedges (Boix-Fayos et al., 2009; Nadeu et al., 2012). This information has been added to the study area description in the paper (section 2). For the sediment data used in this paper, depth information is available for C in profiles on the sediment wedges and the reservoir downstream. In the case of the bars, all observable depositional layers were sampled, in the case of the suspended sediment, we sampled various heights, and in the case of the delta sediments two superficial layers were sampled. Details on this can be seen in the methods section (3)

When analyzing our results, we have merged information from all depth layers available for a more straightforward comparison, given also the complexity of comparing different sediment deposits. Our goal is to show and understand the complexity of C dynamics in a heterogeneous catchment while focusing on lateral processes. Nevertheless, for better understanding, we have added some more information and reference to our earlier work regarding the in-depth distribution of OC to support our interpretations of stability in the study area (section 2, study area).

References:

Boix-Fayos, C., de Vente, J., Albaladejo, J., Martínez-Mena, M., 2009. Soil carbon erosion and stock as affected by land use changes at the catchment scale in Mediterranean ecosystems. Agric. Ecosyst. Environ. 133, 75–85.

Nadeu, E., de Vente, J., Martínez-Mena, M., Boix-Fayos, C., 2011. Exploring particle size distribution and organic carbon pools mobilized by different erosion processes at the catchment scale. J. Soil. Sediment. 11, 667-678.

Nadeu, E., Berhe, A. A., de Vente, J., and Boix-Fayos, C.: Erosion, deposition and replacement of soil organic carbon in Mediterranean catchments: a geomorphological, isotopic and landuse change approach, Biogeosciences, 9, 1099–1111.

6. Discussion:

• Currently, the chapter 5.1 and 5.2 are partially redundant and there is not clear distinction between the topics.
• I suggest to structure the discussion in terms of changes along the sediment cascade as depicted in Fig. 1; starting at the soil and C sources and then discuss the lower pools with greater transport distances. This information is then used to highlight the mechanisms of C loss and gain in different depositional settings and with greater transport distance.
• In line with my suggestion to restructure the results, I suggest to discuss the selectivity of the grain sizes first and then to stress the effects of grain size selectivity on the mechanisms of the C cycle.

We thank the reviewer for this suggestion. However, in our opinion the basic structure of the discussion as it is now does not differ much from what is suggested by Dr. Hoffmann. To increase clarity and prevent redundancy, we have separated aspects related to the selectivity of material sizes from those aspects related to its influence on the mechanisms of the OC cycle more strictly. To avoid redundancy we have removed some text of section 5.1 and we have changed the former 5.1 to a section analyzing the effect of spatial scale on OC measured in sediments. Section 5.2. discusses particle sizes and OC more separately. All this following our "sediment cascade structure" (as pointed out by Dr. Hoffman) for interpretation of the results from the beginning of the paragraph. In the last paragraph of the section 5.2.2 a summary of processes affecting OC along the cascade of sediments is given.

• No information of the age of the sediments is given. Do you have any ideas of the age of the dated samples and how this relates to depletion and enrichment of OC in the different pools.

The sediments deposited in the alluvial wedges have been stored there since the 1970s, when the check-dams were built. Although, we don't have information on the age of the sediments through dating, in a previous study (Nadeu et al., 2012) we used radiocarbon dating to date the buried OC in two profiles from alluvial wedges behind check-dams. From these results, we observed that the age of the buried OC was related to the sediment and geomorphological dynamics that had taken place in the catchments during the period over which sediment accumulated. We expect the OC concentration and division in different pools to be strongly related to the sediment dynamics and to the particle-size distribution and state of aggregation of mineral particles, as explained above. Based on our previous work (Nadeu et al. 2011; 2012), we are inclined to think that the OC concentration in sediments in our study catchment is related on the one hand to the erosion process delivering these sediments to the streams, and consequently to the characteristics of the sedimentary sources. On the other hand, OC concentration in sediments is also influenced by post-depositional carbon dynamics (stabilization upon burial, mineralization or new OC formation). As we already referred to these hypothesis in various parts of the paper, as we do not know the age of sediments and consider further discussion on this beyond the objectives of the manuscript, we did not add additional discussion on the issue.

References:

Nadeu, E., de Vente, J., Martínez-Mena, M., Boix-Fayos, C., 2011. Exploring particle size distribution and organic carbon pools mobilized by different erosion processes at the catchment scale. J. Soil. Sediment. 11, 667-678.

Nadeu, E., Berhe, A. A., de Vente, J., and Boix-Fayos, C., 2012: Erosion, deposition and replacement of soil organic carbon in Mediterranean catchments: a geomorphological, isotopic and landuse change approach, Biogeosciences, 9, 1099–1111,

- ***Differences between TOC, POC and MOC are only marginally discussed. Are there major differences in the processes of erosion, transport, deposition and depletion between these fractions?***

Please refer to answer on question 4 on OC pools.Ratios between POC and MOC pools have been calculated, a graph on this is included in Figure 5, a description of the results is included in section 4.3 and some sentences were added to the discussion (section 5.1).

- ***In contrast to other cited studies, the authors highlight that their results show a decrease/depletion of OC after soil erosion and transport. However, the reasons for this difference are insufficiently discussed. Is this basically due to different erosion and transport processes, or different spatial and temporal scales of OC transport, or both, or something else? Please extend the discussion on the controlling factors of the depletion/enrichment of OC during transport.***

Discussion on this subject has been added in the text in the section 5.1. Basically we suggest that at this coarse scale the sediments have less organic carbon than the surface layer of the soils of the catchment (taking them as a reference), contrary to the common results at finer spatial scales, where normally the eroded sediments are found to be enriched in organic carbon compared to the original soils (Martínez,-Mena et al., 2008 and references there in). Chaplot and Poesen (2011) already suggested a decrease in the SOC delivery from the microplot to 60 ha and upto the 1000 ha scale. We suggest that at these larger scales the decrease of OC in sediments compared to soils is due to a combination of factors: (1) the sediments come from a mixture of sediment sources mobilized by different erosion processes affecting deeper soil layers with low OC content; (2) when measuring deposited sediments in a specific sediment sink and OC therein, we only measure part of the sediment, while another part that is relatively rich in OC (transport selectivity) is travelling further distances (OC is related to the finest particles with longest transport distances); and (3) at this scale we measure organic carbon across the different phases of the erosion process and so our observation window is wider than when results are reported at the plot or hillslope scale. Furthermore, when sediments travel longer distances, lower physical protection of OC within aggregates might lead to an increase in the mineralization rates. When data are collected at plot and hillslope scales, the sediments are very close to their sources during the detachment and transport phases and organic carbon has less opportunity for mineralization. Note also that this is not a predominantly agricultural catchment where interill and rill erosion dominate, but a catchment with a heterogeneous land use in which a variety of soil erosion processes, affecting different soil profile depths, deliver sediment intro the streams.

References:

Chaplot, V., and Poesen, J.: Sediment, soil organic carbon and runoff delivery at various spatial scales, Catena, 88, 46-56, 2012.

Martínez-Mena M., López, J., Almagro M., Boix-Fayos C. Albaladejo J. 2008. Effect of water erosion and cultivation on the soil carbon stock in a semiarid area of South-East Spain Soil & Tillage Research. 99, 119-129.

Referee 2

7. ***The most significant comment I have on this work is that the manuscript heavily uses C:N ratios of soils and sediments as indicators of SOM processing and decomposition. I think caution should be exercised in the use of C:N ratio for this purpose because the only way that C:N ratios can be used as reliable indicators of SOM mineralization is if the C:N ratio of the inputs across the different sites was identical. It is very likely that even similar vegetation groups have grown in different parts of a catchment can have slightly different C:N ratios. In the absence of such info on above- and below-ground vegetation inputs, one can only say changes in C:N ratios are likely due to differences in mineralization rates or selectivity of transport . . . it is hard to justify any definitive statements on C:N rations indicating differences in mineralization rates without furtherdata.***

We fully agree with the limitations of the C:N ratio as indicator for mineralization as stated by the referee. Therefore, we tried to be cautious with the interpretations of C:N for this purpose and we tried to better reflect that in the manuscript. The following two arguments help to better interpret our results on C:N ratios and possible implications for mineralization of OC:

1. The interpretation of C:N ratios along with the clay content of soils and sediments can possibly give an indication of mineralization. A low C:N ratio with a high clay content has been interpreted as an indicator of potential mineralization suffered by OC in sediments when compared to high C:N and high clay content of original soil sources (Wang et al., 2010; Nadeu et al., 2012).
2. In agreement with the comment of the referee, sediment sources with different C:N inputs would determine different C:N evolution along the fluvial path. In our study catchment soils with different land cover (agricultural soils, forest, shrubland and pasture) act as potentially different sediment sources. Particularly POC in soils can be a good indicator of vascular plants and potentially different C:N ratios. Yet, the soils under different land covers in our study catchment do not show significant differences in the C:N ratios of the POC fraction (Kruskal-Wallis test, p-level>0.05) neither in the MOC fraction. However, we observe differences in C:N ratios between soils and most of sediment deposits (except alluvial wedges) (Figure 5 in the manuscript). This indicates that carbon inputs from different soil sources should not show large differences with respect to C:N and this can give (together with clay content) some indication on mineralization.

Apart from this, lower C:N ratios can be expected if the sediment source is from deeper soil layers (e.g. bank and channel erosion), but this variable is taken into account in the interpretation of results in the discussion.

The data indicating similar C:N ratios of the POC and MOC fractions between soils from different land uses have now been incorporated in the text (section 4.3 of the results) to help clarifying that the “soil references” had similar C:N ratios and were significantly higher than those of most of the sedimentary deposits. Although all the arguments above give a strong indication that indeed mineralization must have been responsible for at least part of the OC loss, we do agree that no definitive statements can be made based on these multiple indicators, which was further emphasized in the text.

References:

Nadeu, E., Berhe, A. A., de Vente, J., and Boix-Fayos, C., 2012: Erosion, deposition and replacement of soil organic carbon in Mediterranean catchments: a geomorphological, isotopic and landuse change approach, Biogeosciences, 9, 1099–1111,

Wang, Z., Govers, G., Steegen, A., Clymans, W., Van den Putte, A., Langhans, C., Merckx, R., and Van Oost, K.: Catchment scale carbon redistribution and delivery by water erosion in an intensively cultivated area, Geomorphology, 124, 65–74, 2010.

8. ***Furthermore, when inferring input of autochthonous C from aquatic systems, I think the authors should further discuss literature that compares C:N ratios and other differences between autochthonous and allochtonous sources of OM in aquatic (or at least periodically inundated) systems.***

This subject is further discussed in the text (section 5.2.2) as suggested by the referee, including recent references supporting the idea of a potential significant contribution of autochthonous organic matter in deposited fluvial sediments.

9. ***Abstract, Line 6 - add 'after it is' before 'stored'***

Added

10. Page 5009, Lines 8-11, the reasons for how and why erosion constitutes a sink for atmospheric carbon dioxide stated here is not complete, please revise this section and make sure to also include partial replacement of eroded C as part of why erosion constitutes a sink (along with burial of eroded C).

Paragraph has been reviewed and rewritten accordingly.

11. The explanation for the patterns in clay and OC concentrations in soils vs. sediments in page 5021 doesn't seem correct. The high clay content and low OC in sediments could simply be a reflection of the selective transport of light organic particles. Consider revising this section to account for that possibility (Referee 2).

Although we acknowledge the possible explanation as proposed by the reviewer, based on our data we do not have evidences that show relations between high clay content, low OC and selective transport of light organic particles. On the contrary, in the studied sedimentary deposits lighter organic particles indicated by POC (Particulate Organic Carbon) content were not related to high clay contents. Clay content is positively associated with TOC in sediments of alluvial wedges and suspended sediments and negatively associated to the labile organic fraction (POC) in all data and in the reservoir sediments (Table 3). Also along the fluvial path the selectivity during transport shows finer particles and lower OC with a dominance of the MOC fraction (’heavier’) in the suspended sediment.
Sediment with low OC contents found in the catchment in previous research (Nadeu et al., 2012) hasa dominance of MOC fraction (66%) versus the lighter POC fraction (34%).
Those reasons led us to think that low OC, high clay content and low C:N could likely indicate mineralization more than selectivity of light organic particles associated to clay. Similar interpretations are supported by other authors comparing soils and sediments (Wang et al., 2010).

References:

Nadeu, E., Berhe, A. A., de Vente, J., and Boix-Fayos, C., 2012: Erosion, deposition and replacement of soil organic carbon in Mediterranean catchments: a geomorphological, isotopic and landuse change approach, Biogeosciences, 9, 1099–1111,

Wang, Z., Govers, G., Steegen, A., Clymans, W., Van den Putte, A., Langhans, C., Merckx, R., and Van Oost, K.: Catchment scale carbon redistribution and delivery by water erosion in an intensively cultivated area, Geomorphology, 124, 65–74, 2010.

~~Coupling~~ S~~s~~ediment flow-paths and associated ~~with~~ organic carbon dynamics across a Mediterranean catchment

Carolina Boix-Fayos[1*], Elisabet Nadeu[1], Juan M. Quiñonero[2], María Martínez-Mena[1], María Almagro[1], Joris de Vente[1]

[1]Soil Erosion and Conservation Research Group. CEBAS-CSIC (Spanish Research Council), Campus de Espinardo 30100, PO BOX 164, Murcia, Spain.
[2] Department of Geography. University of Murcia. Campus de la Merced, 30001, Murcia, Spain
*Corresponding autor: cboix@cebas.csic.es

Abstract

Terrestrial sedimentation buries large amounts of organic carbon (OC) annually, contributing to the terrestrial carbon sink. The temporal significance of this sink will strongly depend on the attributes of the depositional environment, but also on the characteristics of the OC reaching these sites and its stability upon deposition. The goal of this study was to characterise the OC during transport and stored in the depositional settings of a medium sized catchment (111 km^2) in SE Spain, to better understand how soil erosion and sediment transport processes determine catchment scale OC redistribution. Total Organic Carbon (TOC), Mineral-Associated Organic Carbon (MOC), Particulate Organic Carbon (POC), Total Nitrogen (N) and particle size distributions were determined for soils (i), suspended sediments (ii) and sediments stored in a variety of sinks such as sediment wedges behind check-dams (iii), channel bars (iv), a small delta in the conjunction of the channel and a reservoir downstream (v) and the reservoir at the outlet of the catchment (vi). The data show that the OC content of sediments was approximately half of that in soils (9.42±9.01 g kg^{-1} versus 20.45±7.71 g kg^{-1}, respectively) with important variation between sediment deposits. Selectivity of mineral and organic material during transport and deposition increased in a downstream direction. The ~~OC~~ mineralisation, burial or *in situ* ~~formation~~ incorporation of OC ~~occurred~~ in deposited sediments depend~~ing~~ed on their transport processes and on their post-sedimentary conditions. Upstream sediments (alluvial wedges) showed low OC contents because they were partially mobilised by non-selective erosion processes affecting deeper soil layers and with low selectivity of grain sizes (e.g. gully and bank erosion). We hypothesise that the relatively short transport distances, the effective preservation of OC in micro-aggregates and the burial of sediments in the alluvial wedges ga~~i~~ve rise to low OC mineralisation, as is arguably indicated by ~~with~~ C:N ratios similar to those in soils. Deposits in middle stream areas (fluvial bars) were enriched in sand, selected upon deposition and had low OC concentrations. Downstream, sediment transported over longer distances was more selected, poorly microaggregated, with a prevalence of~~dominated by~~ silt and clay fractions and MOC pool~~associated with OC~~. Overall, the study shows that OC redistribution in the studied catchment is highly complex, and that the results obtained at finer scales cannot be extrapolated at catchment scale. ~~that~~Selectivity of particles during detachment and transport, and protection of OC during transport and deposition are key~~processes have a strong effect on the~~ for the concentration and quality of OC found at ~~the~~ different depositional settings. Hence, eco-geomorphological processes during the different phases of the erosion cycle have~~, with~~ important consequences for the temporal stability and preservation of the buried OC and in turn for the OC budget.

1. Introduction

Con formato: Numeración: Continua

Con formato: Sangría: Izquierda: 0 cm, Sangría francesa: 0,63 cm

Terrestrial ecosystems have captured up to 28% (2.6±0.8 Pg y^{-1}) of the CO_2 emitted annually over the last decade (Le Quéré et al. 2013). Among the processes involved in this terrestrial carbon (C) sink, terrestrial sedimentation of eroded soil and replacement of soil organic carbon at eroding sites have~~s~~ been regarded as ~~an ~~active components (Stallard, 1998) (Harden et al., 1999; Van Oost et al., 2007)~~.~~ The magnitude of its contribution to the sink has been estimated by some to be between 0.6 and 1.5 Pg of C annually (Stallard, 1998; Aufdenkampe et al., 2011) through the burial of large quantities of laterally transported organic carbon (OC). The significance of this contribution, however, will depend on the long-term preservation of the buried OC, an issue that remains under debate (Van Oost et al., 2012 and references therein). The fate of the redistributed OC will ultimately depend on the mechanisms of its physical and chemical protection against decomposition, its turnover rates and the conditions under which the OC is stored in sedimentary settings (Van Hemelryck et al., 2011; Berhe and Kleber, 2013).

Con formato: Fuente: Garamond

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

Understanding how OC moves along with sediments through the different phases and types of erosion and transport processes is crucial to explain partially~~understanding~~ the large variation in C contents found in depositional sites. Along these lines, ~~Pr~~pro~~o~~gressively~~;~~ the geomorphic factors that control the redistribution of OC within watersheds are being defined (Berhe and Kleber, 2013; Evans et al., 2013; Hoffmann et al., 2013; Nadeu et al., 2011; Nadeu et al., 2015~~2;~~). The study of OC transport from erosion sites to depositional settings implies the consideration of a large number of factors and processes taking place.~~ along the whole way.~~ Several studies have described the impact of variation of~~the differing~~ transport and deposition of different OC size-fractions and the role of OC mineralisation, as well as the breakdown of soil aggregates or re-aggregation at depositional sites~~ettings~~ (Wang et al., 2010; Van Hemelryck et al., 2011; Martínez-Mena et al., 2012) and the contribution of new OC formation from vegetation at depositional settings. Altogether, these factors are considered responsible for the transformations undergone by OC from source to sink. Yet, comprehensive studies of source to sink processes are, to the best of our knowledge, lacking.

The objective of this study is to characterise the OC in transit and at a range of depositional settings in a medium size catchment and to associate our observations with the catchment sediment dynamics. We aimed to: (i) characterise the OC concentrations in the main sedimentary deposits along the catchment's drainage system; (ii) assess the main processes involved in sediment redistribution; (iii) establish links between these processes and the OC concentration and quality.

2. Study area

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

The average annual rainfall for the period 1933-2004 was 583 mm and the average annual temperature 13.3° C, at a station located in the centre of the basin at 1200 m above sea level. Snow in the mountains, especially above 1700 m, is not abundant but is frequent in winter. The dominant soils in the area are Lithosols, ~~Calcaric~~ Regosols and ~~Calcaric~~ Cambisols (IUSS-WRB~~FAO~~, 2006). They have an average OC concentration in the first 10 cm between 3.2 and 1 % depending on the land use, being the lowest for agricultural use mainly on marl lithology (Nadeu et al., 201~~3~~4). ~~Variations of~~ A previous study at the site showed that OC concentration in soil profiles down to 1 meter located in forest and shrub areas in a subcatchment was 1.5±1.4% and ~~of~~ 2.2±1% in profiles located in forest areas in another Rogativa subcatchment ~~decreased from X%±X in the tospoil to X%±X at 0.9 m depth under shrubland and from X%±X to X%±X under forest cover for the same depth for two soil profiles~~. In both subcatchments average OC concentration in channel sediments down to 80 cm were 1.1±0% and 1.4±0.1% respectively~~-~~ (Nadeu et al., 2012). Boix-Fayos et al. (2009) attributed ~~shows also~~ variation of OC concentration in depth ~~variations~~ in sediment~~s~~ profiles down to 120 cm located in the main channel of~~ in~~ 7 subcatchments of ~~the~~ Rogativa to changes in the land use pattern of the drainage area over the last decades. ~~Furthermore lateral redistribution of OC by erosion processes causes an accumulation of 26 % of the mobilized C to be deposited within the catchment hillslopes, occupying 6 % of the hillslope area in a non-homogenous way (Nadeu et al., 2014a).~~

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

~~The TurrillaRogativa catchment shows a dendritic channel pattern. The main channel has an average slope of 7.7 ° and a total longitude of 22 km. The stream is discontinuous upstream and continuous downstream.~~

~~The landscape is a mix of dryland farming (mainly barley), plantations of walnuts (*Junglans regia* L.), forests and shrublands.~~ The catchment has been affected by important land use

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

The Turrilla catchment shows a dendritic channel pattern. The main channel has an average slope of 7.7 ° and a total longitude of 22 km. The stream is discontinuous upstream and continuous downstream. Geomorphological characterisations of channel reaches along the main and tributary streams of the Rogativa catchment indicated dominance of non-selective erosion processes affecting deeper soil layers and with no selectivity of grain sizes, such as gully, bank and river bed erosion (Nadeu et al., 2012), often activated as a consequence of the decrease in sediment input from the adjacent slopes caused by a generalised recovery of the vegetation, following agricultural land abandonment and reforestations (Nadeu et al., 2014a). Furthermore land use and morphological characteristics of the drainage area were identified to be important driving factors determining the concentration and organic carbon yield exported by lateral fluxes in smaller subcatchments of the Rogativa watershed (Nadeu et al., 2015). In general terms, the main channel of Rogativa moved from an aggradation period with large sediment volumes coming from a well-connected agricultural catchment (1950's-1980's), to an incision and degradation phase after afforestation, land abandonment and hydrological control-works (Boix-Fayos et al., 2007). Nowadays the Rogativa catchment is under a transition phase with an armoured main channel and sediments being incorporated in the channel through gullies and bank erosion (Boix-Fayos et al., 2007; Nadeu et al., 2011). ~~Sediments reaching the streams originate from upper soil layers, through interril erosion processes (Nadeu et al., 2012), and from deeper soil layers below the channel, due to massive erosion processes (bank collapses, deep gullies and the river bed) (Boix-Fayos et al., 2007; Nadeu et al., 2011).~~

3. Methods

The field experimental design was based on the main pathways of sediment and soil OC during their transport through the catchment. Therefore, soils and different sedimentary deposits within the catchment were sampled as shown in Figure 2~~1~~.

3.1. Soil data

Topsoil (0-10 cm) samples were taken from 109 locations distributed from upstream to downstream in the catchment, representing all land uses and their spatial extent. Of these samples, 20 % were from high density forest soils, 30 % from low density forest soils, 20 % from shrubland soils, 13 % from pasture soils and 20 % from agricultural soils. Disturbed samples were taken for laboratory analyses and undisturbed samples (rings of 100 cm^3) for estimating soil bulk density.

Figure 1 and 2

3.2. Sediment data

a) Alluvial wedges (behind check-dams)

The sediment deposited behind the network of check-dams installed in the 1970s was used as representative of material mobilised by erosion processes and fluvial transport from the upper catchment areas (Figure 1 and 2). Nineteen (sub) catchments, evenly distributed, were sampled. The sediment wedges deposited behind each check-dam were sampled at the front (close to the check-dam) to a maximum depth of 1.25 m. In addition, 14 of the sediment wedges were sampled also upstream, at the back of the sediment wedge, to a maximum depth of 96 cm. At all points, bulk samples of 100 cm^3 were taken at intervals of 7 cm depth until the maximum depth was reached. Moreover, for 14 wedges, replicate samples of the first layers were taken at 7-cm intervals, to 35 cm depth. A total of 537 undisturbed samples were collected.

b) Fluvial bars

Sedimentary fluvial bars located at two different reaches in the Rogativa main stream, 2 km apart in the middle section of the catchment, were sampled. One of the reaches is a permanent-flow reach where four different bars were sampled during three seasons (autumn 2009, winter and spring 2010), and the other is an intermittent reach where four bars were sampled in autumn 2009 and winter 2010 (the same dates as the bars of the permanent-flow reach). A disturbed sample of the first 5 cm of the bar was taken at each sampling period, as no different layers corresponding to different events could be distinguished in these bars.

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

c) Suspended sediment

Two devices for the sampling of suspended sediments were installed in the main stream of the Rogativa channel in October 2010: one was installed in the downstream area, draining a catchment area of 54.4 km^2, and the other was installed ~~just~~ below the confluence with the perennial stream of Arroyo Blanco, draining a total catchment area of 78.1 km^2.

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

d) Delta sediments

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

e) Reservoir sediments

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

3.3. Laboratory analysis

All soil and sediment samples (from erosion deposits and deposition bars) were air-dried or dried in an oven at a low temperature (<60º) and then sieved at 2 mm. Primary particle size distribution was measured using a combination of wet sieving (particles >63 µm) and laser diffractometry (particles <63 µm) using a Coulter LS, for the sand fraction and the silt and clay fractions, respectively. The organic matter in these samples was oxidised with hydrogen peroxide and chemically dispersed with a mixture of sodium hexametaphosphate and sodium carbonate (anhydrous) for 18–24 h. The fractions obtained were classified as: coarse sand (2000–250 µm), fine sand (250–63 µm), coarse silt (63–20 µm), fine silt (20–2 µm) and clay (<2 µm). The effective particle size distribution was measured by introducing air-dried samples into a Coulter LS, without previous physical or chemical dispersion. From these two measurements, micro-aggregation indices were derived.

The percentage of micro-aggregated particles of fine sand, coarse silt and fine silt sizes and two aggregation indices that give an indication of the total percentage of micro-aggregated particles were used: IA (Index of Aggregation), as the sum of the differences of the dispersed and non-dispersed material in each size group (Wang et al., 2010), and ASC (Aggregation Silt and Clay) as the difference between dispersed clay and silt and non-dispersed clay and silt (Igwe, 2000).

Given that~~e~~ soil carbon pools might have differences in turnover times, it is of high interest to evaluate the behaviour of different carbon pools separately, as this will give us insight in the long term stability of the mobilized~~eroded~~ carbon by lateral fluxes. For this purpose t~~T~~he OC was divided into physical fractions by wet sieving: particulate organic carbon (>53µm) (POC) was separated from mineral associated organic carbon (<53µm) (MOC) after shaking 10 g of air-dried soil sieved at 2 mm with 50 ml of sodium hexametaphosphate for 18 h (Cambardella and Elliot, 1992). Fractions were oven-dried at 60ºC for water evaporation and the dry material was weighed prior to OC determination. The OC and nitrogen contents were determined by dry combustion in an elemental analyser (FLASH EA 1112 Series Thermo). The total organic carbon (TOC) was assumed to be the sum of the POC and MOC. The MOC accounted for micro-aggregate and intra-aggregate OC in the silt and clay size fractions. Duplicate or triplicate soil samples were used for laboratory analysis.

Table 1

3.4. Statistical analysis

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

4. Results

~~4.1. Organic carbon and nitrogen in soils and sediments~~

~~The TOC concentrations of sediments were, on average, around half (9.42±9.01 g kg~~$^{-1}$~~) of the average TOC concentrations of surface soils (0-10 cm) (20.45±7.71 g kg~~$^{-1}$~~), being higher in alluvial wedges behind dams, in the suspended solids, in the delta and in the reservoir and lower in active alluvial bars in the main channel (4.44±1.98 g kg~~$^{-1}$~~) (Figure 2). The concentrations of OC present as POC were highest in sediment samples taken from the alluvial wedges, while suspended solids and reservoir profiles contained the highest levels present as MOC fractions. The lowest MOC fraction was found in the bars. The C:N ratios were higher in soils and alluvial wedges and lower for the rest of the deposits (Figure 2).~~

~~Figure 2~~

~~4.2.~~4.1. Primary particle size distribution of soils and sediments

The particle size distribution of soils differed from that of most of the sediments (Figure 3). The alluvial bars showed, on average, higher sand contents and lower contents in the fine fractions than the soils (Figure 3). This enrichment in coarse fractions in the alluvial bars was accompanied by the lowest TOC (Figure~~s 2 and~~ 4~~3~~). In contrast, the suspended solids and the delta and reservoir sediments had higher percentages of silt and clay than the soils. The high sand contents in the reservoir and delta sediments were similar to the sand content (around 20%) of the wedges. The particle size distributions of the alluvial wedges were more similar to those of the soils of the catchment than to those of the rest of the deposits (Figure 3).

Figure 3

~~4.3.~~4.2. Micro-aggregated particles in soils and sediments

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

Table 2

4.3. Organic carbon and nitrogen in soils and sediments

The TOC concentrations of sediments were, on average, around half (9.42±9.01 g kg^{-1}) of the average TOC concentrations of surface soils (0-10 cm) (20.45±7.71 g kg^{-1}), being higher in alluvial wedges behind dams, in the suspended solids, in the delta and in the reservoir and lower in alluvial bars in the main channel (4.44±1.98 g kg^{-1}) (Figure 4). The highest particulate organic carbon (POC) content ~~concentrations of POC were highest~~was observed in sediment samples taken from the alluvial wedges, while suspended solids and reservoir profiles contained the highest contents~~centrations~~ of mineral associated organic carbon (MOC)~~ fractions~~. The lowest MOC content~~fraction~~ was found in the bars (Figure 4). ~~Soils, wedges and bars showed~~S~~ s~~imilar POC:MOC ratios were found on soils, wedges and bars (ranging from 0.5 ~~on average~~ in bars to~~and~~ 0.8 ~~on average~~ in soils), whereas m~~. M~~uch lower POC:MOC ratios were found for the delta and reservoir (~ 0.2), reflecting the prevalence of the MOC pool transported downstream (Figure 4 and 5). Furthermore, while C:N ratios of the MOC and POC fractions of soils sampled under different land uses in the catchment did not show significant differences (data not shown, K-W test, p>~~<~~0.05), most of the sediment deposits (except for alluvial wedges) showed significantly lower C:N ratios than soils (Figure 5). In general a ~~The~~ decrease of C:N ratios of sediment deposits along the fluvial path was accompanied by a simultaneous decrease in N, with the exception of suspended load, in which a higher N content was found than in the other deposits (data not shown, K-W test, p<0.05).

Figure 4 and Figure 5

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

The Spearman correlation coefficients showed different patterns in the relationships between the percentages of primary and micro-aggregated particles and the OC pools and C:N ratios across the deposit types (Table 3). The Index of Aggregation (total amount of micro-aggregated particles)~~ (Index of Aggregation, IA)~~ was positively correlated with~~ the~~ TOC (for all data) and for the reservoir (p<0.06), POC (for all data and alluvial wedges) and C:N ratio (in soils). Furthermore, the percentages of micro-aggregates of~~in the~~ 250-63 µm ~~fraction~~ were positively correlated with ~~the~~ POC for all data and alluvial wedges~~ (in the reservoir deposits)~~ and negatively with C:N ratio (for ~~all data, as well as in soils,~~ wedges and suspended sediments~~ separately~~). Percentages of microaggregates~~d particles~~ of 63-20 µm were positively correlated to TOC and MOC of all deposits data and ~~particularly~~ to the TOC and MOC of alluvial wedges. Furthermore this~~63-20 µm~~ microaggregate size~~fraction~~ was positively correlated to C:N of all data, soils and wedges. The smallest microaggregated particles (20-2 µm) were positively correlated to ~~the~~ TOC and MOC of the reservoir and to ~~the~~ POC of all data. In contrast, the ASC index (micro-aggregated material of silt and clay size fractions~~), consistently,~~ was negatively correlated with the OC pools, only in the cases that not correlation with fine-sized microaggregates (20-2 µm) was found. ASC correlated also negatively with~~ and~~ C:N ratios for all data, soils and alluvial wedges.~~in soils and different deposits.~~ In general, it seemed~~s~~ that the TOC content and its fractions correlated positively with micro-aggregated material (IA). Some microaggregated sizes showed significant positive correlations with OC of some deposits. ~~;~~ Larger microaggregated sizes correlated with TOC and POC of alluvial wedges and the finest microaggegated size with ~~the~~ TOC an MOC in the reservoir downstream. ~~while this relationship became negative when smaller aggregate sizes were considered for the aggregation index (ASC).~~

No~~There were no~~ consistent correlation patterns were found~~in the relationships~~ between the percentages of primary particles and the OC pools and C:N ratio across deposit types. OC (total and/or different pools) ~~The TOC~~ was~~as~~ associated positively with the sand fraction and negatively with the clay fraction, for all the data considered together, soils and the reservoir. By contrast, the clay fraction was associated positively with TOC in wedges and suspended sediment, and with MOC in alluvial wedges. ~~The POC was correlated positively with the sand fraction in the reservoir sediments.~~

Overall, as may be expected, the OC content was positively correlated with the clay fraction in some deposits (e.g., alluvial wedges and suspended sediments), indicating selectivity during detachment and transport. However, OC was also positively correlated with the content of sand particles in the soils and reservoir, probably indicating the entrance of organic material from other processes (e.g. in situ formation of C in lakes (Tranvik et al., 2009)) in the reservoir and OC formation in the upper layers of sediment. Positive correlations of the sand and OC in soils are probably due to the presence of several samples of sandy soils covered by dense forest (25% of dense forest soil samples had 45-70 % sand).

Table 3

5. Discussion

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

5.1. OC redistribution: effect of scale of observation~~from source to sink~~

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

~~We suggest that at these larger scales the decrease of OC in sediments compared to soils is due to a combination of factors related to the spatial scale of observation, namely; (i) variety of sediment sources, that dilute the source effect; (ii) interaction of multiple erosion processes, both selective and unselective; (iii) long transport distances that favour continuous remobilisation of sediment facilitating aggregate breakdown and a reduced physical protection of OC; and (iv) the ample time lapse (from hours to several years) that occurred between sediment detachment and its sampling. This contrasts with sediments collected at the plot and hillslope scales, which are collected close to their sources during the detachment and transport phases, and often shortly after the erosion event, with little opportunity for OC mineralization.~~

Previous studies have also found a depletion of OC in sediments measured at a catchment scale (Avnimelech and McHenry, 1984; Haregeweyn et al., 2008; Chaplot and Poesen, 2012). For a small catchment (22 ha), Fiener et al. (2005) observed that sediment deposited in ponds was depleted in OC and clay relative to the source soils, while the sediment collected at the outlet of the pond was enriched in OC and clay compared to soils. This demonstrates the important changes in OC content that may occur during different transport and deposition phases due to preferential deposition. Similar results were reported by Wang et al. (2010) and Rhoton et al. (2006), who found OC impoverishment in deposited sediments within their study catchments, while the sediment transported in suspension and exported out of the catchment was enriched in OC (1.2-3 times) compared to the OC concentration in the source soils. Ran et al. (2014) attributes also lower values of OC than soils for different sedimentary settings, based on several methods, to estimate the OC carbon budget of the Yellow River. In our case, the suspended sediments as well as the reservoir sediments showed significantly higher contents~~centrations~~ of MOC compared to the other sediment deposits, while the MOC contents~~centration~~ in soils was similar to that in reservoir sediments and slightly higher than in suspended sediments~~;~~. ~~t~~This was a~~s~~lso found by Amegashi et al. (2011) and can be probably attributed partially to new, ~~autochthonous~~in-situ formation of OC in the reservoir (Einsele et al., 2001). ~~, this is discussed in the following sections.~~.

We suggest that at these larger scales the decrease of OC in sediments compared to soils is due to a combination of factors related to the spatial scale of observation, namely; (i) variety of sediment sources, that dilute the source effect; (ii) interaction of multiple erosion processes, both selective and unselective; (iii) long transport distances that favour continuous remobilisation of sediment facilitating aggregate breakdown and a reduced physical protection of OC; and (iv) the ample time lapse (from hours to several years) that occurred between sediment detachment and its sampling. This contrasts with sediments collected at the plot and hillslope scales, which are collected close to their sources during the detachment and transport phases, and often shortly after the erosion event, with little opportunity for OC mineralization.

Con formato: Justificado

~~Some authors have found sediments deposited in ponds with high trapping efficiencies and anoxic conditions to be enriched in OC, relative to the source soils (Amegashie et al., 2011). Except for the MOC concentration in reservoir sediments, in our case all sediments were depleted in different OC fractions and in TOC, while significant differences were also found between deposits. The fact that reservoir sediments showed higher TOC concentrations than sediments deposited in wedges behind check-dams, as also found by Amegashie et al. (2011), can probably be attributed partially to new, autochthonous formation of OC in the reservoir (Einsele et al., 2001).~~

5.2. Mechanisms of OC loss or gain in sediments

Among all the factors and processes abovementioned ~~above~~ that condition~~s~~ a general decrease of OC in sediments in our studied catchment compared to soils, three of them closely interact: (i) sources of sediment linked to specific erosion processes; ~~,~~(ii) size selectivity; ~~(ii)~~ and (iii) processes affecting ~~The following sub-paragraphs discuss three complementary explanations for the different OC concentrations observed in sediments, compared with the original soils: (i) size selectivity of material upon detachment, transport and deposition; (ii) variation in the sources of sediments and OC and (iii) processes that affect~~ the organic components of ~~the~~ sediments during the erosion and fluvial transport pathway such as burial, mineralisation or new OC formation.~~ (iii)~~

5.2.1. Size selectivity and sources of sediment

Wang et al. (2010) explained how selectivity of soil material determines the OC concentration during transport. They concluded that there was very little mineralisation of TOC during the erosion process, since they found~~based partially on the~~ higher C:N ratios ~~being higher~~ in sediments than in source soils and similar enrichment ratios of clay and C in deposited sediments. Based on multiple indicators o~~O~~ur data show evidence for~~to~~ the contrary in the middle and lower catchment deposits (suspended sediments, delta and reservoir): higher clay contents~~ than soils~~, lower OC and low C:N ratios in sediments than in soils. These findings could indicate C losses by mineralisation during transport over longer distances. However, the ~~I~~interpretation of narrowed C:N ratios as indicator of mineralization must be done cautiously and in combination with other indicators (Wang et al., 2010), given the complexity of N behaviour along the fluvial path (Robertson and Groffman, 2007). This is even more complex in sediments of intermittent rivers (as in our case) because nitrification and denitrification processes are also dependent on the drying and rewetting cycles of sediments (Gómez et al., 2012; Arce et al., 2013). In our study, total TOC and N were lower in all sediment deposits compared to soils, with the exception of suspended load. Increased of N in sediments rich in clay (as suspended load) has been attributed to the presence of ammonium or to the contribution of fresh-water algae (Sánchez-Vidal et al., 2013).~~In our study area apart from the mentioned combined indicators of mineralisation, wSo, although no definitive conclusions can be drawn, our findings seem to indicate C losses by mineralisation during transport over longer distances, since apart from the decrease in C:N ratiossource-., higher clay contents and lower OC contents in middle and lower catchment deposits. These findings could indicate C losses by mineralisation during transport over longer distances.~~

~~However, t~~tThe low OC concentration~~s~~ of sediments ~~transported and~~ deposited in the upper catchment ~~areas seem to~~~~could~~may have a different~~t~~ explanation~~s~~. The sediments in the alluvial wedges showed textural classes and C:N ratios similar to those of the soils, but lower OC concentrations. The low OC concentration ~~in the sediment wedges~~ may be explained by the fact that the sediments originate from deeper soil layers, mobilised by non-selective erosion processes such as bank, gully and channel erosion, with lower OC concentrations than the surface soil. Geomorphological field assessments ~~carried out~~ in the area indicated that -these deeper operating erosion processes are indeed important sources of sediments in the catchment ~~that were activated by land use change and installation of check-dams~~ (Boix-Fayos et al., 2007). Yet, sediments derived from deep soil layers not only have lower OC concentration but also different OC pool composition (Nadeu et al. 2011) and different turnover rates (Nadeu et al. 2012) than those derived from topsoil. ~~Moreover, Nadeu et al. (2012) related the different dominant erosion processes to different OC pools transported (Nadeu et al., 2011a) and different ages of OC in mobilised sediments (Nadeu~~

~~et al., 2012), in different subcatchments of the Rogativa~~ watershed~~catchment.~~ Differences in OC pools between sediment deposits can also be the result of transport conditions due to higher or lower ~~Other studies related differences in the pools of OC exported by erosion to differences in~~ rainfall intensity (Martínez-Mena et al., 2011; Zhang et al., 2013) or to the discharge~~. (ref?)~~. Smith et al. (2013) related high discharge rates~~flows~~ with transport of modern C, associated with erosion and the release of organic matter (Sanchez-Vidal et al., 2013), and C from vascular sources (Goñi et al., 2013). Low flows were associated with export of fossil OC (Smith et al., 2013), from biogenic sources dominated by non-vascular plants (Goñi et al., 2013) or from fresh water primary producers (Sanchez-Vidal et al., 2013).

A comparison of the particle sizes of the studied sediments and soils of the catchment points to a selection of transported material in a downstream direction. Finer micro-aggregated and single particles were present in the suspended load and in the reservoir. In the soils and in the alluvial wedges behind check-dams, the material was much more heterogeneous~~ierarchical~~, having similar particle size distribution and larger ~~being~~ micro~~-~~aggregates~~d at a large size (sand fraction)~~, indicating again its transport by non-selective erosion processes and over short distances.

~~In general, the micro-aggregation of particles (<250 µm, e.g. IA index) and clay content were positively correlated with TOC, as found by other authors (Martinez-Mena et al., 2008; Jin et al., 2009; Martínez-Mena, 2011), and with MOC in some deposits.~~

Comentario [E1]: Can this be further discussed? Otherwise we can maybe leave it in the results section…

5.2.2. Processes affecting organic carbon dynamics during transport and deposition

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

The C:N ratios showed clearly two groups (Figure 5~~3~~): lower values in suspended sediments,~~ and~~ delta and reservoir sediments, and higher values in soils and alluvial wedges. This suggests a relatively low mineralisation of OC in the sediments of alluvial wedges due to efficient burial or a short transport time~~ because of the proximity to sediment sources in the upper catchment areas~~, as reported also by Smith et al. (2013) and ~~as~~ suggested by Ran et al. (2014). Further downstream in the suspended sediments,~~ and~~ delta and reservoir sediments, the lower C:N ratios could indicate mineralisation of OC in sediment transported over longer distances (Figure ~~3~~5) (Bouchez et al., 2010; Hovius et al., 2011; Raymond and Bauer, 2001). Ran et al. (2014) recently reported a mineralization of 27 % during erosion and transport of sediment and associated OC through the Yellow River catchment. Although, as previously stated~~said before~~, interpretation of C:N ratios for mineralization must be done with caution. Nevertheless, and interestingly, the observed trend in the C:N ratios in soil and among different sediment deposits was consistent with that found in the POC:MOC ratios (~~-~~Figure 5) which support our statement that the degree of OC mineralisation is increased during transport along the flow-path.

~~The N cycle in sediments of intermittent rivers (as in our case) is even more complex because nitrification and denitrification processes are also dependent on the drying and rewetting cycles of sediments (Gómez et al., 2012; Arce et al., 2013). Several studies have suggested that N increases in suspended load due to the presence of ammonium (NH4+) in sediments with high clay contents (as it is the case of suspended load) or to the contribution of fresh-water algae in suspended sediments (Sánchez- Vidal et al., 2013). However, in our studyIn our case TOC and N are lower in all sediment deposits compared to soils with the exception of suspended load that shows N contents similar to soils. In~~

~~previous studies was suggested that N increases in suspended load can be due to the presence of ammonium (NH4+) in those sediments with high clay contents (as it is the case of suspended load) or to the contribution of fresh-water algae in suspended sediments (Sánchez- Vidal et al., 2013). The N cycle in sediments of intermittent rivers (as in our case) is even more complex because nitrification and denitrification processes are also dependent on the drying and rewetting cycles of sediments (Gómez et al., 2012; Arce et al., 2013).~~

Apart from the indications of OC mineralisation, the MOC ~~values~~ concentration ~~(?)~~ of suspended sediments ~~were~~ was low and very variable (3.8~~2.08~~-0.1~~29~~ %) which can be attributed to the diverse characteristics of the events that mobilise material from different sources (Smith et al., 2013; Goñi et al., 2013; Sanchez-Vidal et al., 2013) or to sediments mobilised by different erosion processes (Nadeu et al.,, 2011;~~,~~ 2012~~-~~).

Furthermore, the relatively high C content of the reservoir sediments could indicate *in situ* organic matter formation from ecological lake processes stimulating primary production (Einsele, 2001) or allochthonous OC input from the establishment of vegetation and soil formation in the frequently exposed upper sediment layers~~ (as observed in the field)~~.

Moreover, while other deposits (wedges and suspended sediment) showed the well-known relationship between clay and OC (Rodriguez-Rodriguez, 2004; Rhoton et al., 2006; Martínez-Mena et al., 2008), ~~the~~ reservoir sediments presented ~~showed~~ a correlation between the sand fraction and concentration of OC in the two pools, that could be indicative of *in situ* C formation. Autochthonous ~~sources~~input of organic matter in river and lakes~~ ecosystems~~ can account around 50 % of the total organic matter in aquatic ecosystems of~~according to data from~~ tropical-semiarid and dryland areas~~lakes and rivers~~ (Kunz et al., 2011; Medeiros and Arthington, 2011) ~~(Kunz et al., 2011)(Medeiros and Arthington, 2011)~~. The ~~variations~~fluctuation ~~of~~in ~~the~~ autochthonous organic matter production in riverine ecosystems depends on the input from terrestrial land uses in the drainage area, with thresholds in which terrestrially derived C is replaced by in-stream algal productivity (Hagen et al., 2010). However, it can also be influenced by ~~But also it depends on the~~ river hydrodynamics (Cabezas and Comín, 2010; Devesa-Rey and Barral, 2012). In particular, in Mediterranean river ecosystems there is an important seasonal shift between inputs of allochthonous and authochtonous organic matter, with high primary production in spring and alloch~~o~~tonous organic matter inputs in autumn~~m~~ (Romaní et al., 2013). Given that C:N ratios of ~~A~~allochtonous organic matter (more recalcitrant and resistant to degradation by microorganisms) ~~C/N ratios~~ tend to be higher than *in situ* produced organic matter ~~due to more recalcitrant organic components~~ (Devesa-Rey and Barral, 2012), shifts between one and the other will change OC decomposition rates and dynamics.

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

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

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

Comentario [E2]: format

Con formato: Fuente: Cursiva

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

5.3. Implications for the fate of eroded OC

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

6. Conclusions

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

The~~se~~ processes that determine~~ing~~ OC concentration at different pools are related also to the different phases of erosion (detachment, transport and deposition):~~distribution by the sediments in catchments seem also to be associated with the erosion phase~~ (i) during detachment: size selectivity, type of erosion process and source of material; (ii) during transport: size selectivity, protection of OC in micro-aggregates and transport distances, and (iii) during deposition and in the post-deposition phase: size selectivity, protection of OC from mineralisation by stabilisation of micro-aggregates and burial and new OC formation are important.
The OC mobilised in catchments is associated very closely with the sediment dynamics and can have long residence times, linked to the fate of the sediments. In addition, it can be increased by ecological processes and by replacement in eroded areas, converting catchments into relevant sinks for C budgets.

Acknowledgements

This work has been financially supported by the project ESUMA (11859/PI/09) of the Seneca Foundation, Regional Government of Murcia (Spain) and by the project SOGLO

(P7/24 IAP BELSPO) from the Belgian Government. Joris de Vente was supported by a contract 'Juan de la Cierva' (JCI-2011-08941). Elisabet Nadeu acknowledges financial support from an FPI predoctoral fellowship of the former Ministry of Science and Innovation (BES-2008-002379). We also thank the MIRAGE project (EC, FP7-ENV-2007-1) and Gonzalo G. Barberá, for facilitating the installation of the suspended solids sampling devices. Gonzalo G. Barberá adviced also about the statistical approach. Rosa Gómez Cerezo, María del Mar Sánchez Montoya and Marisa Arce are thanked for providing a part of the samples of the fluvial bars group. Many thanks to Pedro Pérez Cutillas for producing Figure 1.

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

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

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

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

*Different lower case letters mean significant differences among sediments and soil groups.

Different capital letters mean significant differences within sediments and soil groups, with regard to micro-aggregated class sizes

(Kruskal-Wallis test, $p < 0.05$)

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

	~~Micro-aggregated particles~~					~~Primary particles~~		
~~Groups~~	~~250-63 µm~~	~~63-20 µm~~	~~20-2 µm~~	~~IA~~	~~ASC~~	~~Sand~~	~~Silt~~	~~Clay~~
~~TOC~~					-			
~~All data~~	~~**-0.356**~~*	-	-	~~**0.477**~~	-	~~**0.701**~~	~~**-0.264**~~	~~**-0.642**~~
~~Soils~~	-	-	-	-	~~-0.512~~	~~0.574~~	-	-
~~Wedges~~	-	~~**-0.504**~~	-	-	-	~~-0.494~~	-	~~**0.497**~~
~~Suspended sed~~	-	~~-0.351~~	-	-	-	-	~~**-0.480**~~	~~**0.474**~~
~~Reservoir~~	-	-	~~-0.694~~	-	-	~~0.636~~	-	~~**-0.713**~~
~~POC~~								
~~All data~~	-	-	-	~~**0.488**~~	-	-	-	~~-0.273~~
~~Soils~~	-	-	-	-	~~-0.618~~	-	-	-
~~Wedges~~	-	-	-	~~**0.529**~~	-	~~**-0.720**~~	~~0.450~~	-
~~Suspended sed~~	-	-	-	-	-	-	-	-
~~Reservoir~~	~~0.582~~	-	~~-0.664~~	-	~~-0.674~~	~~**0.825**~~	-	~~**-0.874**~~
~~MOC~~								
~~All data~~	-	~~**-0.379**~~	-	-	~~-0.281~~	-	-	-
~~Soils~~	-	-	-	-	-	~~0.588~~	-	-
~~Wedges~~	-	~~**-0.577**~~	-	-	-	-	-	~~**0.724**~~
~~Suspended sed~~	-	-	-	-	-	-	-	-
~~Reservoir~~	-	-	-	-	~~-0.655~~	~~0.629~~	-	~~**-0.727**~~
~~C:N~~								
~~All data~~	~~**0.438**~~	~~**-0.549**~~	~~**-0.403**~~	-	~~**-0.552**~~	~~**0.414**~~	~~**-0.269**~~	~~-0.229~~
~~Soils~~	~~**0.747**~~	~~**-0.980**~~	~~**-0.902**~~	~~0.597~~	~~**-0.742**~~	-	-	-
~~Wedges~~	~~**0.505**~~	~~**-0.791**~~	-	-	~~**-0.533**~~	-	-	-
~~Suspended sed~~	~~0.345~~	-	~~**-0.519**~~	-	~~-0.374~~	-	~~-0.259~~	-
~~Reservoir~~	-	-	-	-		-	-	-

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

Table 3

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

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

[a] This correlation has $p<0.06$

Figure 1. Location of the study area and sketch representing the morphological positions selected for sampling of the soil and sediments. Soils were sampled all around the catchment in all the different land use classes: alluvial wedges behind check-dams predominantly in the upper and middle parts of the catchment, while alluvial bars and suspended solids were sampled in the middle and downstream areas. Delta and reservoir represent sampling areas downstream, at the entrance and in more central parts of the reservoir.

Figure 2. Median, percentiles (10th, 25th, 75th and 90th) and error of the total organic carbon concentration (TOC), particulate organic carbon (POC), mineral associated organic carbon (MOC) and C:N ratios in soils and different types of sedimentary deposits within the Rogativa catchment. Different letters (a-e) indicate significant differences according to the Kruskal-Wallis test ($p < 0.05$). ND= No data available, * = suspended solids measured contained only the MOC fraction.

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

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