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 Dr. Hoffmann and Referee 2 for their time reviewing this manuscript and for their constructive and positive comments. They have pointed out several important aspects that we take into account and discuss further below. We also appreciate the valuable suggestions to clarify and improve the consistency and the structure of the text, which surely will improve the readability of our manuscript. Both reviewers comments are answered below. The topics and the comments of the referees are presented in *italic* and our answers in standard fonts.

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 will be changed accordingly.

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

OK

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).

No quantitative data are available for contribution on different processes, but a qualitative description and general geomorphological processes dynamics will be summarized in the study site. Besides, we can further illustrate the relative role of gully and bank erosion based on some of our previous field and modelling studies (Boix-Fayos et al., 2007; Nadeu et al., 2011; Quiñonero-Rubio et al., under review).

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. Accepted.

• 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 will be included in the manuscript.

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.
OK

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?

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. Organic carbon is mainly associated to the solid phase of sediment fluxes. At the plot scale, Mineral Associated Organic Carbon (MOC) is associated to the fine particles and it has been observed to be the main OC fraction mobilized by interrill erosion due to the size selectivity processes taking place during the detachment and transport phases of erosion (Martínez-Mena et al., 2008). It has also been observed how POC (Particulate Organic Carbon >0.053 mm) (more labile and with low density) is preferentially mobilized during low intensity storms where transport capacity is low (Martínez-Mena et al., 2012), all this at the plot and hillslope scale. In contrast, at the catchment scale, we have observed that mobilization of MOC is strongly associated to fine particles and transported over long distances, while POC is associated to coarser material and mobilized over short distances (alluvial wedges). 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.
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?

The use of 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 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 (for more details on this see response to reviewer 2).

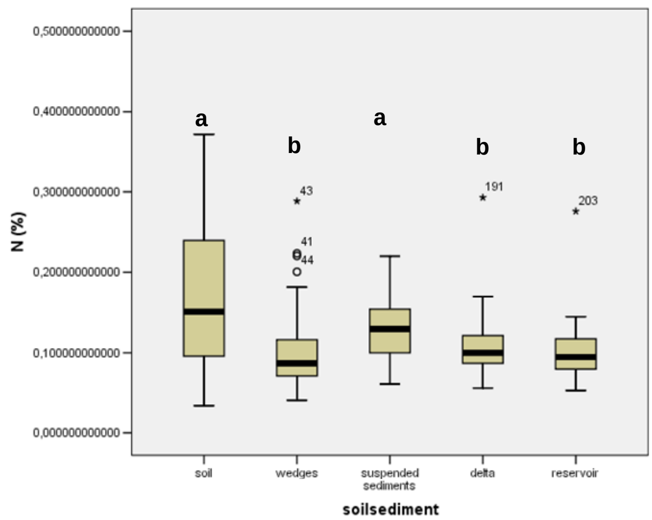

Figure 1. Box-whisker plots of the N concentration in soils and different sediment deposits Differences according to Kruskal-Wallis test ($p<0.05$).

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.
OK, we will restructure the text 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.

We will include few scatter plots that provide additional insights.

• 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 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 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. The comparison between the superficial soil and the whole sedimentary profile indicates the dynamics of mobilized OC in a wide range of erosional events.

• 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).

We have collected and published information about the in-depth distribution of OC in soils and sedimentary deposits in former papers on the same area (Boix-Fayos et al., 2009; Nadeu et al., 2011; 2012). In this paper we focus on the lateral processes and distribution. Nevertheless, where needed for understanding, we will add some more information and reference to our earlier work regarding the in-depth distribution of OC to support our interpretations of stability.

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 more strictly will separate aspects related to the selectivity of material sizes from those aspects related to its influence on the mechanisms of the OC cycle. Through this we aim to adopt the “sediment cascade structure” (as pointed out by Dr. Hoffman) for interpretation of the results from the beginning of the paragraph.

• 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 (OC in-depth poor sediments or richer OC superficial soil layers). On the other, OC concentration in sediments is also influeced by post-depositional carbon dynamics (stabilization upon burial, mineralization or new OC formation).

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. More attention will be given in the text to the discussion on those questions, calculating ratios between fractions and interpreting them.

• 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.

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.

Discussion on this subject will be extended in the text.

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 further data.

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 soil can be a good indicator of vascular plants and potentially different C:N ratios. The soils under different land covers in our study catchment do not show significant differences in the C:N ratio 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 2 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 are 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.

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. (Referee 2).

Ok, further discussion on this subject will be included in the text.

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).

The paragraph begins with "Among the processes involved …" so the idea was to talk about terrestrial sedimentation of eroded soil as one among other processes involved in the terrestrial carbon sink. It was not our intention to neglect replacement of eroded C as other of the processes contributing to the terrestrial carbon sink. We will rewrite the paragraph to clarify the main processes involved in the terrestrial carbon sink, including replacement of eroded soil carbon.

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).

In the studied sedimentary deposits lighter organic particles indicated by POC (Particulate Organic Carbon) content do not seem related to clay particles. Clay content is positively associated with TOC in sediments of alluvial wedges and suspended sediments and negatively associated to the lighter organic fraction (POC) in all data and in the reservoir sediments (Table

3). Also along the fluvial path the selectivity during transport shows that 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) have a 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.