

Dear Editor and Reviewers,

Thanks very much for your useful comments and suggestions on our manuscript that have served to markedly improve the manuscript. We have discussed and modified the manuscript accordingly, and detailed corrections are listed below.

Response to Referee 1:

General comments:

The manuscript by Palleiro et al. presents field data on sediment and metal export from a rural catchment over a period of 3 years. Generally, the presented field-based approach is sound and the manuscript is written fairly well. The manuscript lacks, however, a more thorough discussion including all available data. In particular, chapter 4.5 needs some more in-depth discussion and could thus considerably increase the value of this scientific contribution and, finally, answer the question for the “hydroclimatic controls of sediment and metal export”.

Some of the main findings, e.g. that particulate metal loads are driven by runoff events and that sediment transport is variable over time, are maybe of only little novelty. The strength of this study is the comprehensive data base of 50 sampled discharge events along with meteorologic and hydrologic data over a period of 3 years. However, I have the impression that the potential of this data pool is not fully exploited. For instance, Tables 4 and 5 provide important information on statistical relationships that should be complemented by a process-based discussion of the causal connections. This is not done although the hydrometeorological and hydrochemical data series should provide loads of process information.

Is it possible to distinguish origins of metals or the involved runoff generation processes?

What is the role of the antecedent conditions? These data are shown but not really used for a thorough interpretation.

Do any information on organic carbon contents of the suspended matter exist? It is not clear if the particulate transport of metals occurs sorbed e.g. to organic matter or as part of the minerals themselves.

- ✓ The discussion of section 4.5 was completed.
- ✓ The possible origin of metals according to runoff processes was also included along the text.
- ✓ Yes, information on organic carbon contents of the suspended matter exists, but the data are not published. A comment about this was added in section 4.5.
- ✓ *“In this catchment, the organic carbon content of the suspended matter is low because the organic carbon is mainly exported as dissolved organic carbon*

*(data not shown), suggesting that particulate transport of metals occurs as part of the mineral fraction.”*

Specific comments:

p. 3758, l. 9: I do not understand these numbers: If 38 % of the total metal load was transported particle-bound, then 62 % should be transported in dissolved form. However, the range of the dissolved fraction is only up to 49 %.

- ✓ These percentages were not referred to the total metal loads, but rather to events. That means that if between 38-61 % of particulate load was exported in events, the remainder particulate load was exported in baseflow conditions. The same for dissolved loads, if between 27-49% was exported in events then between 51-73% of the loads was exported in baseflow.
- ✓ Now, in the text, we specified that the percentages are referred to events.

p. 3759, l. 4: How do runoff processes contribute to metal pollution? Do you mean dissolution of metal-bearing minerals?

- ✓ This sentence was unclear and was rewritten. The possible sources of metal pollution are listed: *“In particular, possible sources of metal pollution to rural catchments are domestic wastewaters, atmospheric deposition, soil erosion, and agricultural and livestock activities”*.

Section “2 Study area”: Since runoff events are decisive for metal transport you should give some information about hydrological characteristics, e.g. mean discharge, frequency and magnitude of flood events.

- ✓ These data were included: *“Consequently, most events occurred in autumn (26) and winter (17) followed by spring (4) and summer (3). The mean event rainfall was 39.9 mm, ranging from 12.4 to 101.5 mm. Peak discharge ranged from 0.4 to 21.2 m<sup>3</sup> s<sup>-1</sup>, the maximum increase of discharge (peak discharge/discharge at the beginning of the runoff event) being 6.3. The mean discharge of the 50 events was 1.7 m<sup>3</sup> s<sup>-1</sup>. A detailed study of the hydrological behavior of this catchment can be found in Palleiro et al. (2014).”*

p. 3761, l. 20: At which distance from the river bed was the inlet of the automatic sampler tubing?

- ✓ This information was specified: *“The sampling site was located at the midpoint of the channel cross-section. The inlet of the automatic sampler tube remained at about 1 m from the riverbed.”*

p. 3762, l. 25: How many samples were collected in total?

- ✓ This information was specified: “*A total of 753 water samples was collected during the study period.*”

p. 3765, l. 9: Were dissolved LOADS or CONCENTRATIONS high at low discharge? I can imagine that concentrations decrease when Q increases but loads should always increase with Q.

- ✓ This sentence was clarified in the text. “*By contrast,  $Fe_D$  and  $Mn_D$  load was higher in 2007/08, which was the driest year and with lesser streamflow but higher baseflow (Palleiro et al., 2014).*”
- ✓ Loads do not always increase with the increase of flow. For example, when there is an important dilution of concentrations with the increase of flow, charges may decrease.

p. 3765, l. 24 ff.: Comparisons with adjacent catchments are reasonable, but the value of comparisons of loads from catchments with different geologies and climates is limited unless you do explicitly focus on the different geochemical settings and processes.

- ✓ This comment was taken into account and the comparison of metal loads was focused on catchments with similar characteristics to the studied catchment.

p. 3767, l. 23: Is there any indication for the transfer of Zn to the soluble phase? I did not find an explanation in the cited reference as well.

- ✓ This sentence was confusing, and because of that it was deleted.

p. 3768, l. 10-13: This sentence is unclear – please reword. Which kind of runoff processes favor Zn transport? Surface runoff?

- ✓ This sentence was rewritten and now information about the kind of runoff which could favor Zn transport was included: “*On the other hand, Zn is more abundant in soils, but it is more retained than Cu (Adriano, 2001), hence, the transport of  $Zn_D$  is favored when runoff processes are active. The  $Zn_D$  is delivered to the river probably by subsurface flow, which is the dominant runoff process in this catchment (Palleiro et al., 2014).*”

Figure 4: Are the events shown chronologically or ordered by any variable, e.g. sediment load? This has to be explained somewhere.

- ✓ This was specified in the text of the figure: “**Fig. 4.** *Cumulative rainfall, runoff, sediment (SS) and metal loads during events. Events were ranked according to decreasing sediment and metal loads.*”

p. 3769, l. 10-12: Does the hydrological data give any indication that different particles were transported compared to other events? Or is it possible that particles and metals came from another source / another runoff component?

- ✓ More information about the possible sources of sediments was included: “*Visual surveys showed a strong laminar erosion as well as the formation of rills and ephemeral gullies in some agricultural fields of the Corbeira catchment (Rodríguez-Blanco et al., 2010c), adjacent to the Mero catchment. Probably, this could also happen in the Mero catchment because both basins have similar characteristics.*”

p. 3770, l. 17: “: : efficiency: : :” This is an awkward explanation. Sediment transport from the catchment’s surface means erosion, so the correlation between sediment load and runoff maybe reflects the proportion of surface runoff which is responsible for erosion from surfaces. Moreover, the correlation between sediment load and discharge reflects the concentration of suspended sediment.

- ✓ This sentence was rewritten as follows: “*The good correlation between sediment load and runoff reflects the proportion of surface runoff which is responsible for erosion from catchment surface.*”

p. 3771, l. 4-5: Loads are the product of concentration and flow rate, so it is clear that loads are correlated with flow rates. Because of this link also the loads of sediment and metals are likely correlated with each other. Here it could be interesting to correlate concentrations instead of loads. Have you tried this?

- ✓ This is an interesting comment. In fact, the correlations of flow rate and the remainder variables with sediment and metal concentrations will be discussed in a future paper.

p. 3771, l. 19-20: How much was the explained variability improved? Is it possible to give the same measure of correlation for tables 4 and 5 so that the improvement gets visible?

- ✓ The explained variability was slightly improved. The multiple regression allows us to know the combined effect of the hydrometeorological variables that influence the metal loads. They do not always coincide with the variables which show higher Pearson correlation.

p. 3772, l. 7-8: Loads of Fe(D) and Mn(D) are higher during low flow? Or do you mean concentrations (see also comment above)?

- ✓ As we explained above, this sentence was referred to the loads.

p. 3772, l. 22: I would reword this as follows, since this reflects the causal chain: Particulate metal loads were highly related with sediment load.

- ✓ We had rewritten this sentence as the referee indicated: *“Particulate metal loads were highly related to sediment load, indicating that in the study catchment particulate metal load may be estimated by sediment load.”*

p. 3772, l. 24: How can base flow govern sediment loads? Please explain.

- ✓ Baseflow is a proxy of antecedent conditions of the catchment. This variable could be important for the catchment loads during runoff events through its effect on runoff.

In particular figures 1 & 2 need to be in color or the grey shades should be adjusted. In the present state they are difficult to read.

- ✓ Figures 1 and 2 were changed, the colors were adjusted and Figure 1 includes more information about soil uses.
- ✓ All the technical corrections were taken into account.

## Response to Referee 2:

### General comments:

This paper deals with an analysis of temporal variability in sediment and metal transport at different time scales (annual, seasonal and event). The work is well suited to the journal scope. The objectives are relevant, as they aim understand hydroclimatic factors affecting the transport of sediments and metals (dissolved and particulate) from an agroforestry catchment to a river. Overall the paper presents a large volume of data that could be much more exploited and discussed. The writing style is correct, as is in general the English standard. Tables and figures are suitable although they can be improved. In general a critical reading of the manuscript in order to correct editorial errors is necessary. Some suggestions for improving the manuscript are indicated in the following comments.

### Specific comments:

#### Abstract

Page 3758/L9: Please check the percentages. The total values of the metals transported in particulate form (38%) don't correspond with the metals transported in dissolved form (49%?).

- ✓ This was also commented by the referee 1, now we clarified that these percentages are referred to the exportation during events rather than total metal loads. We clarified that the percentages are referred to the total particulate and the total dissolved; it means that 38% particulate in events is indicating than 62% in particulate form is exported in baseflow.

#### 2 Study area

Include more catchment data as: average height, average slope and time of concentration. This information allows the reader to better understand the behaviour of the catchment.

- ✓ We have added more information about the study area.

#### 3 Material and methods

Page 3762/L7: Please include a sub-session call: chemical analysis. This will distinguish more clearly the data recorded in field, sample collection and laboratory analysis.

- ✓ This sub-session was included: **“3.2 Chemical analysis”**

Page 3762/L7: Enter the five metals species analyzed. In this section, it could be important to specify clearly again the metals determined, although they have already been mentioned twice before (abstract and objectives).

- ✓ The five metal species analyzed were included.

Page 3762/L23-27: Specify the total number of samples collected.

- ✓ This information is now specified in section 3.1 (Data collection): *“A total of 753 water samples were collected during the study period.”*

## 4 Results and discussion

### 4.1 Annual sediment and metal export

Page 3765/L13-26: Include more information on the studies with which you are comparing your results. Note that the climatic conditions, the characteristics of the catchments and geology can produce big differences. This information may help the reader to distinguish the similarities and differences in the results.

- ✓ As indicated above, the comparison of metal loads was focused on catchments with similar characteristics to the studied catchment.

### 4.3 Contribution of runoff events to total sediment and metal loads

Page 3768/L3-10: Given that some metals have higher affinity to form complexes with the organic matter, in the experimental design was taken into account the organic matter determination in suspended matter? If these data are available please include them.

This information could answer questions about how the transport of particulate metals may have occurred. Moreover, doubts raised in the objectives of the work would be clarified.

- ✓ A comment about the organic matter in suspended matter was added: *“In this catchment, the organic carbon content of the suspended matter is low because the organic carbon is mainly exported as dissolved organic carbon (data not shown), suggesting that particulate transport of metals occurs as part of the mineral fraction.”*

### 4.5 Factors affecting sediment and metal loads during rainfall–runoff events

Page 3770/L16-24: Please include more information about the results obtained from the analysis of antecedent precipitation. The information provided is reduced to "Antecedent rainfall 1, 3, 5, 7, 15 and 21 days before the event also affected sediment load during events". A more thorough analysis of these data, together with the information discussed in this paper, could answer questions about its importance in the analysis of the factors affecting the transport of sediments and metals load.

- ✓ More information about the antecedent rainfall influence was included (section 4.5).

#### Conclusions

Page 3772/L7-8: Concentrations or loads in the case of Fe (B) and Mn (D)?

- ✓ They are loads as it is reflected in the text.

Page 3772/L24: Please check this sentence: "Q<sub>b</sub> were the hydroclimatic factors governing the sediment", are you sure with this statement, if it is correct please give a explanation.

- ✓ This was better explained. Q<sub>b</sub> is a proxy of antecedent moisture conditions of the catchment.

#### References

Page 3771/L2, Page 3772/L1 and Page 3774/L18: Please check the correct name of the author: Kuterbanch or Kurtenbach?

- ✓ This mistake was corrected. The right spelling is Kurtenbach.

#### Figures

It is difficult to understand the figure 1, I recommend adjusting colours or increasing the size of the information that you want to highlight. Moreover, if it thinks fit, could superimpose the river on the map of land use.

- ✓ Figure 1 was modified and the colors were adjusted.

Figure 3: Please indicate in the caption of the figure: "Figure 3 Fractions of sediment (SS), particulate (p) and dissolved (D) metals....." Although this information may seem redundant, the figures have to provide the reader with all necessary information. The same indications are required for figure 4, 5 and all the tables.

- ✓ This information was included in all tables and also in figures 3, 4 and 5.

Finally, we remain at the disposal of the Managing Editor and the Referees in terms of making additional changes and improvements to the manuscript.

The authors.