Answer to REVIEWER #2 (author's answers are written in bold)

General comment

This paper focuses on the analysis of stream-riparian-hillslope hydrological connectivity in a humid catchment investigated through the use of the novel tracing technique provided by diatoms. The well-known Luxembourg-based research group was the first, as far as I know, to promote this experimental method a few years ago and this is one of the first applications to investigate hydrological processes at the catchment scale. Overall, the paper is clearly written, with a clear goal, sound analysis and interpretation, and good graphs. However, there are some points that I think the author should address and explain better. First of all, I agree with all comments by the first reviewer, and I'm avoiding to repeat them in my review. I encourage the authors to pay particular attention to the comments reported at the top of page C762 (the comments regarding P2403, L4-9 and P2404, L11-12) and in the central-lower part of page C762 (the comment regarding P2406, L2). In addition, I have also some major concerns and some minor corrections that are reported below and that, in my opinion, should be considered before acceptance of publication to HESS.

We acknowledge anonymous referee #1 for his/her constructive comments on the paper. The effort is highly appreciated. The reviewer agrees with all comments made by Reviewer #1. We kindly ask the reviewer to check how we have answered the comments of reviewer #1.

In the following lines we address the main comments outlined in his/her review.

Specific comments

-The title does not reflect very well the three objectives: indeed, the title suggest a more process-based study, whereas the objectives are more methodologically-oriented. Maybe the title could be changed into "Diatoms as indictor of hydrological connectivity through the riparian-stream system" or something like that.

The first concern of Reviewer #2 is that the title, in his/her opinion, does not reflect well the three objectives of the paper. The Reviewer suggests changing the title to highlight that this is a more methodologically-oriented paper to 'Diatoms as indicator of hydrological connectivity to the riparian-stream-system'.

We have changed the title to 'Hydrological connectivity inferred from diatom transport through the riparian-stream system', as we would like to claim that the paper is both process and methodologically oriented.

-I like the fact that the three objectives reflect well three subsections of the Discussion. However, when one reads the paper for the first time, he/she has a hard time to see the differences between the first objective and the third one. The authors should probably reformulate these two questions in a more univocal way.

Reviewer #2 argued that it is difficult to differentiate between the first and the third objective of the paper. We understand from the comment that the text reads well. It is only the research question that should be reformulated.

Research questions (1) and (3) have been reformulated:

PREVIOUS RESEARCH QUESTIONS:

1. Can terrestrial and aerophytic diatoms be used to reveal hydrological connectivity within the hillslope–riparian–stream system?

3. What are the advantages and limitations of the use of diatoms to infer hydrological connectivity in the HRS system?

NEW QUESTION:

1. Can aerial diatom transport reveal hydrological connectivity within the hillslope–riparian–stream system?

3. Can aerial diatoms be established as a new hydrological tracer?

-P2402, L1-7. These findings are not clearly showed in the manuscript and it's not clear if they come from previous research (in this case insert references). This behaviour should be showed by a new Figure or including a reference to an existing Figure. More importantly, I think that the observation that a second peak (mostly formed by pre-event water, as stated at P2410, L21) does not occur during dry conditions suggests that groundwater (which I assume is the most important component of pre-event water) levels are low and not contribute much to the hydrological response. But this would imply a small contribution of pre-event water, which is not the case (event water dominates during wet conditions). However, later in the manuscript, it's reported that when the catchment was wet there was a higher contribution of groundwater to streamflow. This is quite confusing, we need evidence of these observations, and I think that the author should do a better job to clearly show measurements and observations here and to discuss more in details in section 5 the process interpretation based on them. Finally, we have no clues of how large or small runoff coefficients are: they that should be reported in a Table somewhere (possible Table 4).

As guessed by the reviewer, conclusions about event and pre-event water contributions during runoff events were also drawn from previous studies conducted in the catchment by Wrede et al. (2013). When the catchment was wet, a double peak occurred. The first peak was mainly formed of event-water (50%). We believe that this peak is mainly (but not only) controlled by saturation-excess overland flow in the near-stream areas. On the other hand, the second peak is mainly formed by pre-event water (event water contribution of 16%). The second peaks represent a much larger volume of water than the first peak (see Fig. 4a), resulting in a much larger volume of pre-event water. Pre-event water contribution in the catchment mainly refers to groundwater.

In contrast, during summer conditions, only the first peak occurred. We estimated maximum event-water contributions of 59.5% and 27% for two consecutive events occurred in June 2010 (see Fig 4b; note that there was a typo here, we apologize for this). These values were larger than the 18% event water contributions of the second peak occurred when the catchment was wet. We agree that the results of the second summer event are not in accordance with the general findings in the catchment. We have thus decided to avoid reporting on this event, and rather sustain our results by citing Wrede et al. (2013), which first described this single-double peak event by using silica to discriminate between event and pre-event water during events in the Weierbach catchment. The manuscript has been edited and we hope that this is now clearer. We have also edited Figure 4.

We computed event-based runoff coefficients for the sampled events (using the simple "straight line" separation of baseflow / event flow). However, we avoided giving numbers because it is not obvious (and it is not the scope of this paper) to estimate runoff coefficients for the double peak events that occur in the catchment during wet antecedent conditions. Due to the nature of these events we consider it difficult to determine when the events end. If we consider the end of the events as the return to the pre-event low flow conditions, then recessions might expand over many days resulting in runoff coefficients much higher than 100%. In other cases, rainfall occurs during the falling limb of the hydrograph and a new event starts. As we did not detailed how we estimated the runoff coefficients in the methods section, we have finally opted by removing the text in the results section.

-P2405, L19. I know very little about diatoms but I guess we can expect no valves in rainfall samples. Is it the same for groundwater? Would it be possible that rainfall infiltration processes during long or intense events facilitate percolation of diatom valves through the vadose zone and down to shallow groundwater? Please, add a few words on this here.

We can expect no terrestrial and aerophytic valves in rainfall samples. We did not find diatoms in groundwater samples. At this state of our research we are not sure if they can infiltrate down to the groundwater. As discussed in page 2409, lines 25-30, we tested if diatoms percolate though different types of soil matrix using fluorescent diatoms (Tauro et al., submitted) and we concluded that it was unlikely.

-P2409, L28. This is a critical point. The authors say that transport of diatoms from the riparian zone to the stream could occur via macropores in the shallow subsurface layers and/or overland in the riparian zone. In principle, I agree with the explanation. However, the authors found very little diatoms in overland flow (P2406, L2) and this seems to be in contrast with their second hypothesis of stream-riparian diatom transport. Moreover, PCA suggests only a minor role of overland flow for streamflow generation (P2404, L8-11). I think that some suggestions should be posed by the authors on this issue.

Indeed, we found very little amounts of diatoms in HILLSLOPE overland flow. We specified in the methods section that we only sampled overland flow on lower hillslope positions (page 2397, line 5-12). End-member mixing analysis was performed only considering hillslope overland flow. To avoid confusions we always refereed to 'hillslope overland flow'. We did not explicitly sample riparian overland flow. But, we sampled litter, moss and vegetation for diatom analysis in the riparian zone. Terrestrial and aerophytic diatoms were much more abundant (in absolute numbers) in the riparian zone than the hillslope. When looking at all the measurements together we hypothesised that the transport of diatoms from the riparian zone to the stream might take place either through (i) a network of macropores in the shallow soils of the riparian zone or (ii) overland flow in the riparian zone.

Minor comments and technical corrections

-P2404, L14-15. Higher contribution of throughfall compared to what? Compared to throughfall when the catchment was dry? Or compared to groundwater? Please, clarify, this is an important part to understand well how the catchment behaves.

We have reformulated the sentence: 'To the contrary, a much higher contribution of throughfall was estimated during summer (events 5-8), when the pre-storm catchment state was dry, than during winter (events 1-2)'.

-P2406, L28. Are the bivariate plots built putting streamflow on the x or y axis? This has to be mentioned to correctly understand the direction of hysteretic loops.

Hysteretic loops have been done with streamflow on the x axis. The sentence in the manuscript has been corrected.

P2392, L11. Here and later in the manuscript: 'assemblages': is this a technical word used to describe biotic communities?

Yes, 'assemblages' is a technical word widely used in ecology. It usually refers to planktonic communities.

P2392, L25-28. These sentences should be modified according to possible changes in the results and discussion about the source of diatoms (role of hillslopes).

As previously explained, we only sampled overland flow on lower hillslope positions (page 2397, line 5-12). We did not explicitly sample riparian overland flow. But, we sampled litter, moss and vegetation for diatom analysis in there. Indeed, our results showed that (i) hillslope overland flow contribution to streamflow during events was minimum; and (ii) presence of terrestrial and aerophytic diatoms on the hillslope samples was really little or zero (in absolute numbers), (iii) the riparian zone was the highest terrestrial and aerophytic diatom reservoir. Our results suggested that diatoms were likely sourced exclusively from the riparian zone.

P2393, L11. Add 'of water' after 'stable isotope'.

It has been changed for 'water stable isotope tracers'.

P2398, L5-7. Skip this, it have already been mentioned.

The sentence has been removed.

P2399, L21. Here, and everywhere in the manuscript, I strongly suggest to avoid using the term 'concentration' when referring to the isotopic signature. Technically, it's not a concentration. I suggest to use 'isotopic composition'.

We fully agree with the reviewer. We have revised all the manuscript and all references to 'isotopic concentrations' have been replaced by 'isotopic composition'.

P2399, L26. Include 'isotopic' between 'bulk' and 'composition'

The sentence has been changed.

P2400, L2. Skip ‘end-member mixing analysis’ and use directly EMMA. The acronym has been already defined at page 2394.

The text has been changed.

P2400, L2. Add ‘that’ after ‘assumes’.

It has been added.

P2400, L10. It’s good that you have included reference but please shortly explain what is the difference between ‘physical mixing’ and ‘equilibrium mixing’ because this is an important concept here.

The EMMA approach is based on the assumption that it is the mixing of the different sources of water (with different geochemical and isotopic signatures) that control stream water geochemical and isotopic signatures. The method assumes linear mixing, and we refer to this as ‘physical mixing’. If equilibrium reactions among solutes of different charge occur (and are dominant) we would not expect linear mixing as equilibrium reactions among solutes of different charge are higher-order polynomials (Hooper, 2003). We refer to this as ‘equilibrium mixing’.

The sentence has been modified in the manuscript.

P2400, L21. Explicit ‘SD’ (I guess standard deviation).

Done.

P2401, L1. ‘was’ should be ‘were’.

This has also been corrected.

P2403, L13. What is ‘riparian water’? Groundwater? Overland flow? Please, clarify.

‘Riparian water’ refers to soil water in the riparian zone. We have replaced all references to ‘riparian water’ in the manuscript by ‘soil water in the riparian zone’.

We have moved the sentence to the methods section 3.4, and specified which sampling points are included in each end-member type: “Catchment end-members included shallow groundwater (GW1-4), soil water ($SS1_{20}$, $SS1_{60}$, $SS2_{60}$), soil water from the riparian zone (SSr), rainfall (R), throughfall (TH1-2), snow (SN) and overland flow (OF).”.

P2403, L16. Since here several solutes were mentioned, it’s not clear to which of them the correlation refers to. Please, clarify.

The text has been modified: ‘Ten out of the twelve tracers presented linear trends in the solute-solute plots of stream water samples with at least one other tracer (EC, Cl^-, Na^+, K^+, Mg^{2+}, Ca^{2+}, SiO_2, Abs, δ^2H and $\delta^{18}O$; $r^2>0.5$, p-value<0.01, Figure 6). These tracers were retained for further analysis.’

P2403, L17. It’s not clear what the authors mean by ‘retained for further analysis’. Which? Why? Please, explain.

The sentence has been modified: 'These tracers were retained for the PCA analysis'. Moreover, the steps followed for the hydrograph separation analysis are listed in the methods section 3.3 (see P2400, lines 8 to P 2041, line 14).

P2403, L19. What is the 'pre-defined threshold of collinearity'? Pre-defined by whom? Please, clarify and possible include a reference.

The pre-defined threshold of collinearity is defined in the methods section 3.3. References are also listed there:' stream water concentrations and isotopic compositions (of all samples collected during storm events and low flows at the catchment outlet) were considered conservative when they exhibited at least one linear trend with one other tracer (i.e. r2>0.5, p-value<0.01) (James and Roulet, 2006; Ali et al., 2010; Barthold et al., 2011)'. We do not think that this should be repeated in the results section.

P2403, L21. Skip the definition and use only 'PCA' (acronym already defined earlier in the manuscript).

We have only retained the acronym.

P2404, L24. Change 'wettest' into 'wet'.

This has been changed.

P2405, L7. Is 230 a small or a high or a usual number of taxa? We, as simple hydrologists, have no solid idea.

The number of taxa might be associated to sampling efforts. The number might be higher or lower in different environments. We believe that in order to state if this is a rather high or low value it should be compared to other values.

P2405, L7. I suggest to delete 'catchment-wide'.

We have replaced 'catchment-wide campaigns' by 'seasonal campagins'.

P2405, L12. Replace 'Riparian' with 'riparian'.

This has been changed.

P2406, L4. Replace 'But' with 'However'.

'But' has been replaced by 'However'.

P2407, L15. 'Fig. 10b' should be '9b'.

This has been corrected.

P2407, L25. Although everybody knows what DOC is please explicit the acronym. Moreover, explain and/or give a reference supporting the statement that UV absorbance can be considered a proxy of DOC (it's not immediately intuitive to me).

We have moved the sentence to the 'Methods' to avoid having references in the 'Results' section. We have also added a reference: 'UV absorbance at 254 nm can be considered a proxy of DOC (Edzwald et al., 1985)'.

P2408, L7. Skip 'hillslope-riparian-stream' and use directly HRS, since it was already defined.

'hillslope-riparian-stream' has been replaced by 'HRS'.

P2408, L10-11. Remove (already mentioned).

The sentence has been removed.

P2408, L22. Typo in 'litter'.

The typo has been corrected.

P2410, L9. Use only 'EMMA', without the already mentioned definition.

This has been corrected.

P2411, L14-19. This sentence sounds as already said. Please, try to reformulate.

The sentence has been reformulated.

P2411, L26. Remove 'But'.

It has been removed.

P2412, L4. I think it's more common to use 'ecohydrology' or 'eco-hydroogy' instead of 'hydro-ecology'.

'Hydro-ecology' has been changed by 'eco-hydrology'.

P2413, L10. Replace 'hillslope-riparian-stream' with 'HRS'.

This has been replaced.

Table 2. In the caption, I suggest to remove the sentence in brackets (but keeping the sample size).

This has been done.

Table 3. I suggest to specify in the caption that the valves found on the hillslopes do not include the dry litter zone. Moreover what is the 'baseflow drift'?

The caption of the table has been modified to make it clearer to the reader: "Table 3. Relative percentage of terrestrial and aerophytic valves quantified at distinct zones of the Weierbach catchment. Streambed samples refer to epilithon samples. Riparian zone samples include litter, moss and vegetation. Hillslope samples include litter, moss and surface soil samples. Diatoms were absent on hillslopes covered by dry litter and samples were discarded."

Table 4. Replace 'storm runoff-events' with 'rainfall-runoff events'.

This has been changed.

Fig. 2. In the second panel, use the same label used in Fig. 3, for consistency. I suggest to move the discharge series in the upper panel. I also suggest to change the caption in 'Time series of precipitation, discharge, groundwater depth, volumetric water content…' Also mention what the numbers indicate.

A new figure has been created using the same label for the second panel as in Fig. 3. We tried to move the discharge serie to the upper panel. However, it is then less visual to number the events that were sampled in summer (mainly due to the relative small change on discharge during the dry season compared to the wet season). The caption has been changed following the reviewer recommendation.

Fig. 4. Change the label 'O-18' into 'd18O' or at least '18O'. Change 'winter' response into 'fall-winter' response. Change 'Two components' into 'Two-component'. Delete all that comes after 'using d18O'.

The caption and the label have been corrected. We have changed 'summer and winter response' to 'a) Wet antecedent conditions' and 'b) Dry antecedent conditions'.

Fig. 5. As mentioned above, indicate what 'riparian water' means. Moreover, add if the median or the mean is displayed in the box-plots, as well as percentiles/standard deviation etc.

'Riparian water' refers to soil water in the riparian zone. We have replaced all references to 'riparian water' in the manuscript by 'soil water in the riparian zone'. We understand that, by default, the bottom and top of the box in a boxplot are always the first and third quartiles, and the band inside the box is always the second quartile.

Fig. 6. The part in brackets can be deleted (but keeping the overall sample size). Add 'The' Before 'upper'.

The caption has been corrected.

Fig. 7. Where is the vertical error bar in panel b? Too small to be displayed? SS3R is not in the legend and it's not clear what it indicates. Moreover, how can it be an end-member if some samples (e.g., event 2) fall outside it?

In panel (b) we zoomed in the middle of panel (a) and plotted peakflow stream water samples instead of all the samples. The OF vertical error bars just falls outside the plotted range in panel (b), the reader has to refer to panel (a) to see it displayed. We have better explained this in the new caption.

SS310 refers to soil water in the riparian zone. We have replaced SS310 in all the manuscript for SSr and stated that we refer to 'soil water in the riparian zone'.

Fig. 8. Would it be better to split the Figure in two? Moreover, change '%' into 'percentage'. The last 9 words could be deleted.

We have split the Figure in two and edited the caption.

FINAL NOTE:

We have replaced the term 'terrestrial and aerophytic diatoms' for 'aerial diatoms' in the manuscript. We considered aerial diatom communities as those communities living exposed to the air outside of lentic and lotic environments, following the definition of Johansen (2010), instead of using other classifications such as those of Petersen (1915, 1935) or Ettl & Gärtner (2014). Therefore, it seems now more appropriate to use the term 'aerial' as most species are not strictly terrestrial. We thus considered 'aerial diatoms' as those species listed with values 4 and 5 (Van Dam et al., 1994), which includes diatoms "mainly occurring on wet and moist or temporarily dry places" and diatoms "nearly exclusively occurring outside water bodies", respectively.

References:

Ettl, H. & Gärtner G. (2014) Syllabus der Boden-, Luft- und Flechtenalgen. 2. Auflage. Springer: Berlin Heidelberg.

Johansen JR. (2010). Diatoms of aerial habitats. In The Diatoms: Applications for the Environmental and Earth Sciences, 2nd Edition, Smol JP, Stoermer EF (eds). Cambridge University Press: Cambridge; 465–472.

Petersen J. B. (1915): Studier over danske aerofile alger [Studies on Danish aerophytic algae]. – Det Kongelige Danske Videnskabernes Selskabs Skrifter, Naturvidenskabelig og Mathematisk 12: 272–379.

Petersen JB. (1935). Studies on the biology and taxonomy of soil algae. Dansk Botanisk Arkiv 8(9): 1-183.

Van Dam, H., Mertens, A., and Sinkeldam, J.: A coded checklist and ecological indicator values of freshwater diatoms from The Netherlands. Neth. J. Aquat. Ecol., 28, 117–133, doi:10.1007/BF02334251, 1994.

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