Answer to REVIEWER #1 (author's answers are written in bold)

The manuscript presents new information on diatoms as possible hydrological tracers. From that point of view it can be attractive to the readers and I recommend its publications. I have the following comments:

1. The formulations at many places (e.g. page 2393, l. 16-20, l. 20-23; page 2411, l. 23-25; page 2412, l. 6-12) point out that isotopic and chemical tracers have their uncertainties or that they cannot identify the hydrological connectivity. While it is true, it does not mean that isotopes and water chemistry do not provide useful information. I do not understand the reason of stressing the limitations of isotopes and water chemistry (which are well known). Each technique has its advantages and disadvantages and it holds also for isotopes, water chemistry and diatoms. The application of all these techniques in hydrology should be complementary rather than competitive. While isotopes and water chemistry can not provide information on hydrological connectivity, diatoms can not idenfity water sources, quantify hydrograph components or provide the information on transit times the way isotopes and water chemistry do. In fact, both isotopes and water chemistry were used to provide useful information also in this manuscript. Uncertainties with isotopes and water chemistry are not the reasons while we need to use the diatoms. On the contrary, all tracers can help to improve our understanding of hydrological cycle. Therefore, I recommend to skip formulations stressing the fact that isotopes and water chemistry have limitations. The objective of this manuscript is to explore the potential of diatoms as an emerging tracer, not to remind of the uncertainties of other techniques.

We acknowledge anonymous referee #1 for his/her constructive comments on the paper. The effort is highly appreciated. In the following lines we address the main comments outlined in his/her review.

The reviewer #1 stressed that it is not appropriate to justify the use of diatoms as new tracers by listing the limitations of 'standard' tracers in hydrology (i.e. water stable isotopes and chemistry). The reviewer refers to two different parts of the paper: (1) the introduction (page 2393, l. 16-20, l. 20-23) and (2) the discussion (page 2411, l. 23-25; page 2412, l. 6-12). We fully agree with the argument of reviewer #1, and do believe that all tracers –including diatoms- have advantages and disadvantages (as described in our paper). They should be complementary rather than competitive. We have rewritten the introduction of the paper accordingly. We have also deleted the sentence in the discussion that refers to the limitation of using water stable isotope to trace water flowpahts (page 2411, l. 23-25). Nonetheless, we do not think that the paragraphs referring to large scale tracing (i.e., page 2412, l. 6-12) should be changed. In this paragraph we aimed at pointing out that diatoms might be useful at higher catchment scales, whereas the usefulness of the other tracers have been proved to be limited (e.g. Uhlenbrook et al., 2003; Klaus et al., 2013).

2. Abstract line 20 "(n=11, 2010-2011)" should read "(n-11, 2011-2012)" because the study was conducted in water years 2011 and 2012.

The reviewer also suggested referring to water years by the calendar year in which they end. This has been change in all the manuscript (i.e. abstract, Page 2401, l. 17).

3. Findings summarized in the abstract (which are useful) do not fully answer questions raised at the end of page 2394, especially questions 2 and 3.

Reviewer #1 considered that the findings in the abstract did not fully answer research questions (2) and (3) raised in the manuscript (page 2394, lines 20-25). We have added two new sentences at the end of the abstract, which read like this: *"*Diatom transport data were compared to two-component hydrograph separation, and end-member mixing analysis (EMMA) using stream water chemistry and stable isotope data. Hillslope overland flow was insignificant during most sampled events. This research suggests that diatoms were likely sourced exclusively from the riparian zone, since it was not only the largest terrestrial and aerophytic diatom reservoir, but also water from the riparian zone was a major streamflow source during rainfall events under both wet and dry antecedent conditions. In comparison with other tracer methods, diatoms require taxonomy knowledge and a rather large processing time. However, they can provide unequivocal evidence of hydrological connectivity and potentially be used at larger catchment scales.*"*

4. Page 2395, l. 10 "… strong seasonality in baseflow exist …" – this is a little confusing, because when talking about baseflow, one would suppose that baseflow was determined using certain technique. Baseflow is a component of hydrograph (a very uncertain one with no universaly accepted definition) rather than a characteristic of flow conditions (high-low flow). Therefore, I would rather speak about "seasonality of low flow" of "streamflow seasonality".

We also agree with Reviewer #1 in that we should avoid referring to 'baseflow' when we have not use any technique for hydrograph separation. Hence, all references to 'baseflow' in the manuscript have been changed to 'low flow'.

5. Page 2396, end it would be useful to characterize the wells a little, e.g. their position on the slope, total depths, depths of screen and aquifer type (I suppose they represent unconfined groundwater… of the same aquifer?). If the soils are not more than 1 m deep, it is interesting to know about the wells. Do they capture also the bedrock groundwater (I suppose the upper bedrock might be weathered, not completely impermeable).

Extra information to characterize the wells was requested (page 2395, lines 12-15). In order to better characterise the subsurface. We have added extra information in the study area to mention that below the soils there are Pleistocene periglacial slope deposits (Juilleret et al., 2011), which exhibits high infiltration rates and high storage capacity (Wrede et al., 2014). Detailed information on the location, position on the slope and screening depths of the piezometers was also added in the manuscript. GW1 was located on the catchment plateau, GW2 near one of the springs, and GW3 and GW4 on the break in slope between riparian and hillslope positions. Wells were around 2 m deep and were screened at least for the lowest 50 cm up to a meter.

6. Page 2397, l.20 – please use 2H/H instead of D/H (to be consistent with notation for oxygen and with line 23)

This was corrected, as well as in page 2397, line 25.

7. Page 2401, l. 17 – Fig. 2 shows water years 2011-2012, not 2010-2012; line 20 instead of "water year 2010-2011" I would write about "water year 2011". While annual precipitation for water year

2011 is mentioned, the same information for water year 2012 is missing. It might be useful to mention it.

The sentence has been corrected and we have added rainfall values for water year 2012.

8. Page 2402, l. 4-5 "…the discharge response represented an ever increasing higher fraction of event rainfall" - How do you know it? If it was a result of some study, please give the reference, otherwise it may not be necessary to mention it in this part of the text.

A major concern of Reviewer #1 was the reference to the estimation of runoff coefficients in the results section (page 2402, lines 4-5). Indeed, 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.

9. Page 2403, l. 4-9. If event water contributions were 50%, 27% and 45%, I would not say that the peaks were formed "mainly" by event water.

The text has been reformulated.

10. Page 2404, l.11-12 "when the catchment was wet, there was a higher contribution of groundwater to streamflow than when the catchment antecedent condition was dry". It is an interesting finding which may not be incorrect. However, is it not in contradiction with statements on page 2403, l. 6 and 9? Pre-event water contributions were larger in June (dry conditions, event water contributions 27% and 45%) than in November (wet conditions, event water contribution 50% and 16%). While not all pre-event water is formed by groundwater, groundwater is certainly not an event water.

The reviewer also noticed that the statement in page 2404, line 11-12:

"when the catchment was wet, there was a higher contribution of groundwater to streamflow than when the catchment antecedent condition was dry",

might be in contradiction with what it was stated in page 2403, lines 6-9:

"in winter, when the catchment was wet and flow response was double-peaked, the first peak was formed mainly by event water. This contrasted with the delayed peak that was dominated by pre-event water. For instance, the first peak of the November 2010 event showed a 50% event water contribution, whereas the second delayed peak only 16% (Figure 4b)."

In November, 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). Wrede et al. (2013) first described that pre-event water dominates during the second peak of double peak events, whereas event water dominates when single peaks occur. The manuscript has been edited and we hope that this is clearer.

11. Page 2405, l. 10 I propose using "low flow" instead of "base flow" (see comment 4).

The text has been changed.

12. Page 2405, l. 16-10 – important seasonal changes were not observed. In my opinion Table 3 shows the seasonal differences for the streambed samples. Are they not significant? What is "n" in Table 3? If it is number of samples, is it possible to come to definite conclusion when the numbers of samples for different environments were different?

Concerning the seasonal changes of diatom communities in the catchment, we have now used the non-parametric Mann-Whitney U-test to determine if the samples of the riparian zone and hillslopes collected in summer and winter could come from the same population. However, we could not perform the test for the stream water at low flow and streambed samples due to the small number of samples. As far as we know, this test does not require the same number of samples in the two populations to compare them.

The test was not significant in both cases (two-tailed Mann- Whitney U-test) and thus the null hypothesis was not rejected:

	Z-score	**p-value**
Riparian samples	**1.252**	**0.21**
Hillslope samples	**0.325**	**0.73**

13. Page 2406, l. 2. "Almost no diatom samples were found in overland flow samples". This seems to be an interesting finding given that hillslopes had the highest mean % of terrestrial diatoms. It could be assumed that overland flow should mobilize them, although the overland flow occurred rarely. Do you think that the intensity of the overland flow was too small to mobilize the lively diatoms which have certain resistance? Could there be any other reason?

Reviewer #1 wonders if 'the intensity of overland flow was too small to mobilize the lively diatoms which have a certain resistance'. We found the highest mean % of terrestrial diatoms on the hillslope samples. However, when looking at the absolute numbers the 'the quantities of terrestrial and aerophytic diatoms found on the hillslopes covered by moss and in the overland

flow gutter samples were small and sometimes not sufficient to fully characterize the zone' (page 2408, lines 20-25). Thus, we believe that we did not find diatoms in the overland flow samples because the diatom reservoir in the hillslope was really small. Moreover, Coles et al. (2015) performed two rainfall simulation experiments over (1) a leaf litter hillslope and (2) a bryophie hillslope of the Weierbach catchment, and overland flow did not occur. The authors simulated 1 in 10 year rainfall events, with high rainfall intensities (40 mm/h).

14. Page 2406, l. 18-20 – systematic increase in terrestrial and aerophytic diatoms. Fig. 8 does not show strong increase for some events, especially the largest ones.

We did not mention that the increase was strong, but systematic.

15. Page 2407, l. 5 "low flow" instead of "base flow"?

The text has been changed.

16. Page 2409, l. 7 – no significant seasonal differences… see comment 11 (streambed diatoms seem to show the difference between summer and winter)

See answer to comment 11.

17. Page 2410, l. 11-12 – conclusion that "hydrological response in spring and summer is largely composed of event water" is not fully in agreement with Fig. 4 (see comment 8), similarly line 20 (the first peak was mainly event water)

See answer to comment 10.

18. Page 2412, l. 6-12. Since this text is not related to exploration of the usefulness of diatoms, I recommend deleting it.

The mentioned paragraph discusses on the concept of hydrological connectivity across catchments scales. As section 5.3 deals with the advantages and limitations of the use of diatoms to infer hydrological connectivity in the HRS system, we do think that the text is relevant as there might be potential to be use at larger catchment scales.

19. Page 2413, l.14-15 "-…riparian zones appear to be the largest diatom reservoir…:" Table 3 seems to indicate that also hillslopes are a large reservoir. Should it not be mentioned as well?

Table 3 shows relative percentages of terrestrial and aerophytic valves, not absolute values. Even though we found the highest relative percentages of terrestrial and aerophytic valves in the hillslope samples, the valves found in absolute numbers were lower.

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.

Klaus, J. and McDonnell, J. J.: Hydrograph separation using stable isotopes: Review and evaluation. J. Hydrol., 505, 47–64, doi:10.1016/j.jhydrol.2013.09.006, 2013.

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.

Uhlenbrook, S. and Hoeg, S.: Quantifying uncertainties in tracer-based hydrograph separations: a case study for two-, three- and five-component hydrograph separations in a mountainous catchment. Hydrol. Process., 17, 431–453, doi:10.1002/hyp.1134, 2003.

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.

Wrede, S., Fenicia, F., Martínez-Carreras, N., Juilleret, J., Hissler, C., Krein, A., Savenije, H. H. G., Uhlenbrook, S., Kavetski, D., and Pfister, L.: Towards more systematic perceptual model development: a case study using 3 Luxembourgish catchments. Hydrol. Process., doi:10.1002/hyp.10393, 2014.