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Interactive comment on "Molecular fingerprinting of particulate organic matter as a new tool for its source apportionment: changes along a headwater drainage in coarse, medium and fine particles as a function of rainfalls" *by* Laurent Jeanneau et al.

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Dear colleague, Many thanks for having taking the time to review our manuscript. In the following, we tried to answer your comments and to explain how we will consider them. Your first comment was on the extensive use of acronyms. The two other reviewers and you are unanimous on this point; some acronyms will be removed to improve the readability. Then you questioned the selection of end-members and specifically

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our choice to consider bed sediments. Bed sediments were chosen as a potential end member because of the finality of the study. Among other, one goal is to determine the origin of POM exported during storm events. It would not imply the same conclusion in term of catchment management if the POM comes from bed sediments or from surface erosion. As a consequence it is necessary to consider it in the study. Moreover we generally agree that bed sediments could be a mixture of other end members. However, it needs to be recognized that end-member signatures could be further processed/modified while in the fluvial network since stream is not a passive pipe and thus acquire a unique signature? To account for this potential variability we considered the bed sediment as a potential separate source. Bed sediment could be a substantial store in the fluvial system, and to account for this large pool we have also characterized it separately. Âă About the description of the end-members, all details can be found in the publication by Rowland et al., 2017. Although we understand the interest for the reader to have this information in this article, since it has already been described, we prefer not detail it here to keep the size of the article reasonable. Âă Your third comment was on the description of the rainfall event. You suggest a figure could be a better option than the description (section 3.1). We choose in the paper to present the hydrologic – rainfall data with table 1 along with the description. Moreover a figure can be found in the paper published by Rowland et al., 2017. Âă About the end-member contributions, you ask if it makes practical sense to group forest floor organic horizon and wetland soil surface horizon. We think yes, there may be a practical sense because the vegetation is quite similar for those two areas, so the plant-derived contribution through roots will have similar composition. Moreover they have similar proportions of microbial chemical markers (13% of analyzed compounds in the wetland soil and 11% in the forest soil). Consequently their chemical compositions are close and they group in the statistical treatment. Âă Your next comment was on the figure 4 and the fact that some deposited sediments plot outside the triangle defined by end-members and you suggest that maybe litter end-member did not capture the full compositional diversity of the catchment. We agree with this point. The fact is that end-members molecular

and isotopic compositions were measured on the bulk sample. This is one limitation of the present approach, since during the erosive transfer of an end-member, there may be a size fractionation. This could explain why some samples plotted outside the area defined by end-members. To prevent from this limitation, end-members should be size fractionated and each fraction should be analyzed. Âă About the comparison between molecular and isotopic data, you asked more details about the modeling exercise. This exercise was simply an end-member mixing approach and we will precise at the end of section 2.4, line 154. Then you ask what are the values of d13C used for each end-members. They come from the sister study (Rowland et al., 2017), which is indicated line 154. About this comparison you mentioned a potential bias for more negative values. Looking at this figure, we can have this feeling. So I come back to the data and checked the residual from the linear regression model. The highest residual was for the extrem point (-29.76; -29.32 : measured; modeled) but the mean residual for d13C < -29.2 ‰ (n=6) was 0.287 $\pm$ 0.085 (mean $\pm$ SE) and for d13C > -29.2 ‰ (n=14), it was 0.220 $\pm$ 0.044. The deviation was not statistically different for lower d13C values. Âă On this comparison between molecular and isotopic/elemental data you ask why we did not try the elemental/isotopic data model alone. It was not performed because with four variables (d13C, d15N, TOC and N, [C/N being a linear combination of two variables]) it is not possible to differentiate between more than 5 sources using this type of statistical treatment (Walling, 2013). "As a minimum, n -1 properties are required to discriminate rigorously between n sources. Additional properties are frequently required to increase the reliability of the results." Âă Then you highlight the fact that the discussion in section 4.3 is based on fragile relationship because of only four events were investigated. We totally agree with this point that is the reason why two sentences have been inserted at lines 322 and 332 to precise that this part of the discussion is speculative. It is clear that future investigations are necessary to support this part of the discussion but we found it interesting enough to be mentioned. Âă Then you provide 7 specific comments: Âă 1.ÂăÂăÂăÂăÂă About the design of the sampler. Âă Thank you for this comment. You are right, this method induces modification of

the velocity profile around the sampler, which could result in grain size fractionation. A sentence will be added to precise this point. Âă 2.ÂăÂăÂăÂăÂă The word “mean” will be replaced by “intermediate”.

3.ÂăÂăÂăÂăÂă Could the stream bed sediment characterized by low amount of identified marker contain petrogenic OM? Âă To answer this point, we searched for petrogenic biomarkers such as n-alkanes, hopanes and steranes. Those compounds were not detected in this sample. Its isotopic fingerprint was -28.7 ‰ which is in the range of the values recorded for stream bed sediments in this catchment from -27.7 to -29.4 ‰ mean = -28.5 ‰ (n=5).Âă Âă 4.ÂăÂăÂăÂăÂă What is the source of benzoic acid? Âă The proportion of benzoic acid in soil profiles increased with depth which has been interpreted as a consequence of the humification process (Chefetz et al, 2000). However, the humification of organic matter as a biogeochemical process has been clearly questionned this last tenth of years. We can assume that, since its evolution is inversely correlated to lignin phenols (slope = -0.32; r^2 = 0.23; p-value = 0.001) it derive from the degradation of tannins and lignins. Âă 5.ÂăÂăÂăÂăÂă C16:0 and C18:0 alkanoic acids may be consider as plant-derived. Âă Yes, C16:0 and C18:0 are ubiquitous and may derive from microbial and plant-derive inputs. For this reason they are not used for the calculation of microbial markers (Jeanneau et al., 2014). A precision will be added in the text to mention it. Âă 6.ÂăÂăÂăÂăÂă The word “mandatory” will be replaced by “necessary”. Âă 7.ÂăÂăÂăÂăÂă We agree that data should be freely accessible and we will prepare the data to add them in supplementary materials. Âă Once again many thanks for your time and consideration in reviewing our paper. Sincerely Âă Laurent Jeanneau On behalf of the coauthors Âă Âă Chefetz, B., Chen, Y., Clapp, C. E. and Hatcher, P. G.: Characterization of Organic Matter in Soils by Thermochemolysis Using Tetramethylammonium Hydroxide (TMAH), Soil Sci Soc Am J, 64(2), 583–589, 2000.

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