

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.

Anonymous Referee #3

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Jeanneau et al. present an interesting study of the molecular composition of particulate organic matter in a small, low-relief, forested watershed. They first characterize potential endmembers, then use statistical analysis to apportion their contribution in suspended sediments collected under various runoff conditions. They compare their results with those obtained using elemental and isotopic composition (from a study recently published in Biogeochemistry) and discuss the impact of changing rainfall parameters on the composition of particulate organic matter exported by the river. Over-

all this is an interesting, well written study presenting novel data and proposing a new source apportionment approach. I do have numerous questions and comments, listed below, which I hope will help the authors tighten their manuscript during the revision process.

General comments:

1) Although the manuscript is generally very well written, the (very) extensive use of acronyms is really annoying.

2) Selection of the endmembers: I find strange to consider bed sediments as an endmember as they are in the first place derived from erosion processes and as such should be composed (at least in part) of a mixture of the other likely endmembers. And that is indeed supported by the data presented in the manuscript, e.g. L275-276.

It would also be good to describe the potential endmembers in greater details. What is currently provided section 2.2. is extremely succinct!

3) It's currently very hard for the reader to get a good feeling for how rainfall and hydrology varied across the study period (section 3.1). A good figure would be worth all these words.

4) endmember contributions: the statistical analysis leads the authors to group the surface horizon of forest soils (FH) and wetland soils (W) into a single endmember. I understand why they're doing so from a pure statistical standpoint but I think it would be good to discuss this in terms of surface processes. In other words, does it make practical sense to group these two endmembers together?

Figure 4 has me perplexed by the composition of the coarse fraction in the 12ha catchment. It clearly lays outside the boundary of the mixing defined by the endmembers. It suggests that the composition of the litter endmember is likely not adequate to capture the full compositional diversity in the suspended sediment samples.

5) comparison of molecular and elemental/isotopic data: it would be good to provide

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more details regarding the d13C modeling exercise. For instance, what's the d13C value of each of the endmembers and how did the author choose these values? Looking at the supplemental figure it seems that the model does OK for measured d13C values higher than -29.2‰ or so, but not so well for more negative values (i.e. the modeled d13C values are very flat for measured d13C values < -29.2‰. This should be discussed.

Also, I wonder why the authors haven't tried to compare the molecular data model to the elemental and isotopic data model (instead of comparing molecular data to molecular data + elemental and isotopic data). I think it would be cleaner.

6) As it stands, section 4.3 isn't very convincing as the relationships between composition and rainfall parameters are very tenuous. I suggest dialing it back some such that it doesn't look as speculative as it currently does.

Specific comments:

L82: by design the sampler, although quite clever, isn't isokinetic and as such likely fractionates the suspended load with respect to grain size distribution. Perhaps add a note of caution in the text.

Section 3.1: the use of "mean" is a bit strange, e.g. in "mean median intensity". I know what the authors mean (no pun intended) but it sounds weird. Consider rewording.

L177-180: could this sample have unusually high relative proportions of petrogenic carbon? Is its stable isotope composition any unusual compared to the rest of the dataset?

L200-201: what's the source of Benzoic acid then?

L208-209: it seems to me that C16 and C18 could also be plant derived, at least in part. They're the most abundant FA in living plants.

L381: "mandatory" is weird in this context, consider rewording.

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Data availability: There is no reason not to provide the data as a supplement. I urge the authors to do so.

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