

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 #2

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The authors display an interesting study on the use of molecular biomarkers to "apportion" the sources of particulate matter exported by storm events at catchment scale within two nested sub-catchments. The present study is complementary to a recently published paper by Rowland et al. (2017, cited by the authors), carried out on the same catchments but focussed on the bulk properties of particulate matter. Bulk organic matter approaches are not always successful as parameters such as total organic C - total N concentration and stable C and N isotope measurements, may provide equivocal

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information, due for example to overlaps of end-members' signatures.

Molecular fingerprinting of organic matter, the approach reported by the authors in the present paper, is another possible way to strengthen source identifications, either as an independent tool or as a complementary discriminating approach. It is, however, a major challenge and any achievement in this direction is most welcome. Difficulties arise from many factors such as the heterogeneous composition of organic matter in the source end-members, the dynamics of particles' detachment, transport and mixing along catchment slopes, the characteristics of the storm event responsible for particulate matter export (i.e., temporal change in source supply during storm flows), landuse in the catchment or the distribution of storm flow events. Another difficulty comes from the analytical approach "in itself" that generates hundreds of molecular compounds. Accordingly, apportionment of source contributions is a very difficult goal as some of these compounds may occur in the composition of several source end-members. The accuracy of the source identification in the sediments may also be bias by the representativeness of the molecules in the samples. At last one additional level of difficulty is the use of a composite sediment sampling procedure that smoothes out differences in suspended matter composition during sediment transport.

The manuscript is well written and the authors fulfil several of these queries. The use of all peak areas provides a realistic picture of the composition of total organic matter in the samples. Statistical treatment of the data "sounds" accurate and the conclusions are consistent with usual interpretations in storm flow studies, i.e. enhanced deposition of litter at the upstream location, increasing proportion of in-channel material downstream or sediment sorting with increasing catchment's size (slope length). I do not have any major comments, only minor remarks that are reported below.

- Although the term is widely used in literature I think that the authors could briefly define or provide a reference for what they consider as particulate organic matter (POM). Does the term refer to "pure particulate organic matter" such as vegetation, root or leaf debris derived from litter or to one of the "soil and sediment matter properties", for example the concentration of organic compounds adsorbed or bound to mineral matter (clays, oxides,...) or both. Showing the parameters used to characterize bulk endmember and sediment compositions (in Rowland et al., 2017) may help. They could be added in Table 1 or reported in a new figure.

- I suggest a sharp reduction of the number of acronyms used in the text and a systematic report of the definition of the remaining ones in the legend of the figures. It would really help the reader.

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