## Dear Reviewer,

Thank you very much for your constructive comments. Below please find our responses to your comments.

The manuscript "Combining colour parameters and geochemical tracers to improve sediment source discrimination in a mining catchment (New Caledonia, South Pacific Islands)" by Virginie Sellier et al. presents a fingerprinting study in the Thio River catchment in New Caledonia using colour and element concentrations individually and combined as tracers, as well as a conventional fingerprinting approach and partial least square regression (PLSR) models based on the entire visible spectrum. The study includes interesting findings, is well described, and fits within the scopes of SOIL (soil and method/ degradation), although I think the soil part could be enhanced in the manuscript (please find detailed comments in the pdf and below). The manuscript represents a statistical approach to compare tracer performance and fingerprinting approaches. Artificial mixture samples help validate results and increases the validity of the paper. Methods are not new but the manuscript elicits well the different results obtained in one catchment and is worth being published after major revision. I attach the pdf with detailed comments (98).

| Reviewer's comments                               | Replies                                         |
|---------------------------------------------------|-------------------------------------------------|
| Generally, I would encourage to use less          | This syntax problem will be taken into account  |
| parentheses. In quite a few cases I had the       | when revising the manuscript.                   |
| feeling there is more information in the          |                                                 |
| parentheses than in the actual sentence. In my    |                                                 |
| opinion it disrupts the reading flow and the      |                                                 |
| information should be included into the text.     |                                                 |
| Abstract: The Abstract mentions the               | The abstract will be modified in line with this |
| methodological question of the paper and hints    | comment in order to further highlight the       |
| a management advice "focus on the                 | methodological aspect of the study.             |
| contributions of mining tributaries to reduce     |                                                 |
| sediment inputs", which is not really observed at |                                                 |
| the end and seems obvious knowing mining case     |                                                 |
| studies. I would suggest not distract from the    |                                                 |
| methodological focus of the paper with these      |                                                 |
| "lonely" and obvious statement.                   |                                                 |

The information provided subsequently will be integrated in the revised version of the manuscript.

| Introduction: The introduction seemed to be to   | The methodological aspect of the article is       |
|--------------------------------------------------|---------------------------------------------------|
| be a compromise between a case study, a          | crucial and will be further highlighted in the    |
| management advice, and a methodological          | introduction. Nevertheless, the 'management'      |
| exploration. I would focus on the latter. The    | aspect of the study should not be neglected, as   |
| potential extrapolation of the method is         | the objective of this type of study is to provide |
| repeated in one sentence in Abstract,            | guidance to carry out this environmental          |
| Introduction and Conclusion. I would suggest     | monitoring on-site. The choice of the selected    |
| stating it once (or twice with Abstract and      | methods was also based on the equipment that      |
| elaborating a bit more the criteria (comments in | could be deployed on site (e.g. choice of         |
| the pdf in Conclusion)                           | spectrocolorimetry, portable, fast and            |
|                                                  | inexpensive in terms of analysis).                |

|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  | The extrapolation potential of the method will<br>be more detailed in the conclusions of the<br>revised version of the manuscript.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                        |
|--------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Research Area: The lateral and longitudinal<br>sediment connectivity seems crucial for this<br>work and is not well addressed in the chapter<br>nor throughout the manuscript. How can you be<br>sure that the contributions you calculate are not<br>due the effect of different lateral and<br>longitudinal connectivity throughout the sub-<br>catchments instead of different erosion values<br>on the slope? You write about channel banks and<br>alluvial deposits (map) which let me doubt that<br>the connectivity is as good as believed. Please<br>provide information (text, maps, pictures etc.)<br>proofing your point! Furthermore, the whole<br>relevance of the study revolves around erosion<br>and there is not one picture displaying<br>the "extensive erosion processes" you mention.<br>Please give us some insights here. | Previous results found in the grey literature (e.g.<br>Immila project report, 2020) indicate that there<br>is a strong remobilization of sediments in New<br>Caledonian hydrosystems, particularly for low<br>intensity floods (<200 m <sup>3</sup> /s). However, the<br>magnitude of this process could not be<br>quantified. Tracers such as <sup>7</sup> Be (Le Gall et al.,<br>2017) could be used to trace the contributions of<br>"new" versus "old" sediments and provide more<br>precise indications on the dynamics of sediment<br>remobilisation in New Caledonian hydrosystems.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                |
|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                  | For consistency, the mining and non-mining sources will be referred to as « mining tributaries » and « non-mining tributaries » in the revised version of the manuscript.<br>Several types of samples can be considered for sediment tracing including soil or sediment samples. The sampling of lag deposits has the advantage of being more representative of the entire drainage area, compared to local point-based sampling of soils that will be characteristic of a given more local area (Haddadchi et al., 2013). Moreover, at each sampling site, five to ten subsamples of fine sediment were collected across a 10 m <sup>2</sup> surface which increases the representativity of the sampling scheme.<br>Sampling was based on the knowledge acquired in the catchment, on visual observations made during the two sampling campaigns (2015 and 2017) and on the conditions of accessibility to the sampling areas (i.e. restricted access, no access roads). Erosion zones were notably highlighted by Garcin et al. (2017) and sediment deposition zones were indicated by the inhabitants of Thio and mining engineers who accompanied us in the field to help us carry out the sampling. |

|                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                   | If we compare our study with sediment tracing<br>studies carried out in other catchments of<br>equivalent surface area, the number of source<br>samples taken in our study (i.e. 2 sources, $n1 = 8$ ,<br>n2 = 16) is consistent. For example, the study by<br>Evrard et al. (2019) indicates that 37 source<br>samples (3 sources, $n1 = 12$ , $n2 = 8$ , $n3 = 6$ ) were<br>sampled in the study area (450 km <sup>2</sup> ). The study<br>by Brosinsky et al. (2014) collected 152 source<br>samples (6 sources with a number of samples per<br>source between 10-36) across a catchment area<br>of 445 km <sup>2</sup> .                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            |
|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Results and Discussion: I would assume that gully<br>erosion is the dominant erosion for mining<br>areas, whereas non-mining areas with a dense<br>vegetation cover show other processes. Hence,<br>eroded sediments differ, which helps the finger-<br>printing of course. However, they probably also<br>differ in organic and inorganic carbon content<br>influencing your tracer properties. How was that<br>explored and taken care of?                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                      | Inorganic and organic carbon contents may<br>indeed be relevant for tracing sediment sources,<br>and more specifically for tracing the<br>contributions linked to areas affected by<br>contrasted erosion processes (surface vs.<br>subsurface, Laceby et al., 2017). Analyses of<br>organic and inorganic carbon contents in the<br>source samples were carried out. The statistical<br>results of these analyses showed that these<br>tracers did not provide any discrimination of<br>sources, which is why these tracers were not<br>further explored.                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                              |
| Furthermore, Ca and K is soluble in water. How<br>can it be a conservative tracer in New Caledonia?<br>Please elucidate more on the anthropogenic or<br>natural process that explain the tracer's<br>differences (e.g. K concentrations or colour<br>difference) between mining and non-mining<br>sources? I understand that these are the results<br>of the statistical analysis but please let the<br>reader know about your knowledge of the<br>environment. Your argumentation is purely<br>statistical which, as shown for the FDVS-PLSR<br>model,might not always make sense. What<br>processes, geological or paedogenetic<br>background values, etc. are responsible for these<br>differences?! Your manuscript focuses mainly on<br>the characteristic of the geology, which in case<br>of the eroded mining sediments make sense.<br>However, in case of the erosion of the non-<br>mining sites I believe the properties of cover<br>beds and soils should be the focus throughout<br>the chapters,since in this climate you might have<br>meters of soil development and cover beds<br>above the underlying geology (which did not<br>derive from the underlying geology itself but<br>most probably slope upwards) that are actually | Subsurface processes dominate both the sediment contributions from mining and non-<br>mining sources as demonstrated in our previous study (Sellier et al., 2020) so that using lithological tracers is relevant to characterize and trace the contributions from non-mining sources. These differences in terms of the contents in geogenic elements (K, Th, U,) is demonstrated in the study by Sevin (2014) who specifically analyzed the different lithologies found in New Caledonia. Ca is present in the Earth's mantle (3%) and K is also present although in trace amounts. During the processes of obduction that led to the formation of New Caledonia, the mantle rocks (peridotites) were hydrated/altered by water, which transformed the source rock and depleted it in highly soluble elements including Ca and K (Noël, 2018). These alteration processes continued thereafter and led in particular to the formation of the laterite profile including (from the bottom to the top) peridotites (unaltered rocks), saprolites, yellow laterites, red laterites, ferruginous crust. These different layers of the profile reflect an increasingly important stage of alteration of the |

| eroding and NOT the geology itself. It seems that<br>the few non-mining tributaries provide a lot of                                                                                                                                                                                          | initial source rock (peridotites) from the bottom<br>to the top of the profile resulting in particular in                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                       |
|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| sediment to dilute the dominantly mining contributions along the River.                                                                                                                                                                                                                       | an increasingly higher accumulation of iron due<br>to its low solubility in water: peridotites <<<br>saprolites < yellow laterites (goethite) < red<br>laterites (hematite) < ferruginous armour<br>(hematite).                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                 |
|                                                                                                                                                                                                                                                                                               | These minerals including goethite and hematite,<br>will respectively give a yellow colour to the<br>'yellow laterites' and a red colour to the 'red<br>laterites' (Trescases, 1973). These colours are<br>not found in soils derived from volcano-<br>sedimentary formations, because by definition<br>these formations have much lower metal<br>contents. Soils derived from these formations by<br>weathering are therefore less concentrated in<br>heavy metals and do not show the red and<br>yellow colours characteristic of lateritic soils.                                                                                                                                                                                                             |
|                                                                                                                                                                                                                                                                                               | Regarding the solubility of K and Ca, they do are<br>soluble to some extent, but this does not mean<br>that they were totally removed from the lateritic<br>profile in view of our results. The residual traces<br>of these elements allow us, as the statistical<br>approach showed, to trace the contributions of<br>both mining and non-mining sources. K in<br>particular has already demonstrated its<br>effectiveness for tracing the lithological<br>contributions of sediment (Zebracki et al., 2015).<br>The sediment tracing study by Sellier et al. (2021)<br>that estimated the contributions of mining<br>sources and non-mining sources to sediment<br>during floods confirmed that K is temporarily<br>conservative (analysis of sediment core). |
| I wonder how results look when you standardize<br>the contribution by area? Have you tried that ?                                                                                                                                                                                             | This will be calculated in the revised version of the manuscript                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                |
| Conclusion: In my opinion your discussed factors<br>influencing colour and element concentrations<br>are just one out of many possible ones. Please<br>think about other factors that are worth<br>mentioning. Furthermore, your limited criteria<br>(Ni ores and peridotite massifs) for the | The conclusion will be revised at the light of this comment                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                     |

| expansion of the method might work for New<br>Caledonia but it seems not sufficient to<br>extrapolate to the world! Please revise that<br>throughout the manuscript and in the<br>conclusion! |                                                 |
|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|-------------------------------------------------|
| Figures: Please insert in the map figures: - the                                                                                                                                              | This will be done when revising the manuscript. |
| tributaries' sub-catchments outline is shown, - a                                                                                                                                             |                                                 |
| light hillshade in the back to get a feeling for the                                                                                                                                          |                                                 |
| relief, - display the cake diagrams on the right                                                                                                                                              |                                                 |
| connected to the sample location with lines.                                                                                                                                                  |                                                 |

## References

Brosinsky, A., Foerster, S., Segl, K., Kaufmann, H. (2014). Spectral fingerprinting: sediment source discrimination and contribution modelling of artificial mixtures based on VNIR-SWIR spectral properties. Journal of Soils and Sediments, 14(12), 1949-1964.

Evrard, O., Laceby, J. P., Ficetola, G. F., Gielly, L., Huon, S., Lefevre, I., Onda., Y, Poulenard, J. (2019). Environmental DNA provides information on sediment sources: a study in catchments affected by Fukushima radioactive fallout. Science of the Total Environment, 665, 873-881.

Garcin, M., Gastaldi, Y., Lesimple, S. (2017). Quantification et évolution temporelle des apports miniers dans les rivières calédoniennes. BRGM/RP-66840-FR, 44 p., 23 fig., 5. Bur Rech Géol Min Mém.

Haddadchi, A., Ryder, D. S., Evrard, O., (Olley, J. (2013). Sediment fingerprinting in fluvial systems: review of tracers, sediment sources and mixing models. International Journal of Sediment Research, 28(4), 560-578.

Laceby, J. P., Evrard, O., Smith, H. G., Blake, W. H., Olley, J. M., Minella, J. P., Owens, P. N. (2017). The challenges and opportunities of addressing particle size effects in sediment source fingerprinting: a review. Earth-Science Reviews, 169, 85-103.

Le Gall, M., Evrard, O., Foucher, A., Laceby, J. P., Salvador-Blanes, S., Manière, L., Lefèvre, I., Cerdan, O., Ayrault, S. (2017). Investigating the temporal dynamics of suspended sediment during flood events with 7 Be and 210 Pb xs measurements in a drained lowland catchment. Scientific reports, 7(1), 1-10.

Noël, J. (2018) Etude pétro-structurale et géochimique des processus de serpentinisation et de carbonatation des péridotites de l'ophiolite d'Oman. (Doctoral dissertation, Université Montpellier)

Sellier, V., Navratil, O., Laceby, J. P., Allenbach, M., Lefèvre, I., Evrard, O. (2020). Investigating the use of fallout and geogenic radionuclides as potential tracing properties to quantify the sources of suspended sediment in a mining catchment in New Caledonia, South Pacific. Journal of Soils and Sediments, 20(2), 1112-1128.

Sellier, V., Navratil, O., Laceby, J. P., Allenbach, M., Lefèvre, I., & Evrard, O. (2021). Reconstructing the impact of nickel mining activities on sediment supply to the rivers and the lagoon of South Pacific Islands: lessons learnt from the Thio early mining site (New Caledonia). Geomorphology, 372, 107459.

Sevin, B. (2014). Cartographie du régolithe sur formation ultrabasique de Nouvelle-Calédonie: Localisation dans l'espace et le temps des gisements nickélifères (Doctoral dissertation, Nouvelle Calédonie).

Trescases, J. J. (1973). Weathering and geochemical behaviour of the elements of ultramafic rocks in New Caledonia. Bureau of Mineral Resources, Geology and Geophysics, Canberra, Extract from Bulletin, 141, 149-161.

Zebracki, M., Eyrolle-Boyer, F., Evrard, O., Claval, D., Mourier, B., Gairoard, S., Cagnat, X., Antonelli, C. (2015). Tracing the origin of suspended sediment in a large Mediterranean river by combining continuous river monitoring and measurement of artificial and natural radionuclides. Science of the Total Environment, 502, 122-132.