

Eboigbe et al. Response to reviewers:

We note our responses are in blue and we use the notations RxCx to define a specifically numbered comment (C) relating to a specifically numbered reviewer (R). RxARx refers to a specifically numbered Author Response (AR) that relates to a reviewer comment.

REVIEWER 2 (R2):

R2C1: The manuscript describes a study on the distribution of Hg in soil, plants and the atmosphere near a ASGM mining site in Nigeria. The authors determined total and methyl Hg as well as stable Hg isotopes in soil, the atmosphere (by passive samplers (MerPas) as well as different parts of three types of edible plants with the aim to evaluate Hg levels in the plant and to track pathways of Hg uptake by the plants. Samples were taken near the mining site and at two farm sites (one reference site) situated at different distances from the mining site. The authors can show that the atmosphere, soil, and all edible plants are clearly affected by GEM emissions from the ASGM site. However, total and methyl Hg concentrations in all plant tissues were below reference dose thresholds.

Hg isotope analyses coupled with a two-endmember mixing model reveal that most Hg in plants are derived from atmospheric GEM uptake via foliage although crop roots appear to be to a larger extent influenced by Hg uptake from soil.

ASGM is seen as the most important today's anthropogenic Hg emission source to the environment. Besides its role in the global Hg cycle, investigations on the exposure of local people to ASGM Hg emissions, especially through crops, has been rarely investigated. Thus, the presented study is timely and important.

R2AR1: We thank the reviewer for their thorough summation of the study and positive feedback.

The study presents a comprehensive data set. I like such multi-proxy approaches as they offer deeper insights into the local biogeochemical cycling of Hg and dispersion pathways. The manuscript is well written although I think that some parts esp. the abstract and the introduction could be shortened.

R2AR1: We appreciate the sentiments to reduce the length of the abstract and introduction with the aim of streamlining the manuscript. However, we are hesitant to do this as we believe we have worked hard to make these sections as concise as possible without removing context and background of the study.

We have already trimmed background from the abstract (384 words), and the remaining parts of the abstract list all the critical data and findings of the study. We believe any further cuts to the abstract will begin to reduce its impact and value.

With respect to the introduction (1086 words), it includes all the critical context of ASGM, Hg biogeochemical research into vegetation and crops, and Hg stable isotopes. Considering the breadth/interdisciplinarity of this work, we have to cover a broad background to give proper context for our work.

Furthermore, on June 9th we surveyed the abstract and introduction word counts of the 10 most recently papers published in *Biogeosciences*. The average word counts were 366±124 and 1213±221, respectively. We are close to the mean (within 1SD) of each; thus we do not deem the abstract or introduction to be of concerning length compared with other works in this journal. It is our preference to retain the abstract and introductions sections in their current form.

R2C2: The authors mentioned that Hg isotopes are used for both, evaluation of Hg species transformation processes and for tracking contamination or uptake pathways. In this sense, I believe that the interpretation of the Hg isotope data is probably not as robust as it seems. Especially because there is actually only a single Hg source which is GEM and Hg isotope fractionation processes in soils and plants are far from being completely known/understood. Maybe the authors could address this point in their discussion, although I don't think that this will change the overall message of the study.

R2AR2: We believe our assessment and treatment of both source and processing tracing is robust and both scenarios as essentially assessed in the 2-endmember mixing model. Since the burning of Hg-Au amalgams emits vast amounts of Hg(0) to air, we deem this the best estimate of our source. We have added the following statement to section 3.1:

“Considering the burning of Hg-gold amalgams emits Hg(0) directly into the atmosphere, we deem the mean stable isotope values for Hg(0) in air in the contaminated areas (PS and Farm1) to be signal most representative of the ASGM source.”

We know stomatal assimilation imparts a big negative $\delta^{202}\text{Hg}$ fractionation and by comparing foliage $\delta^{202}\text{Hg}$ to air $\delta^{202}\text{Hg}$ (our “source signature”) we have determined those fractionation factors for each plant ($\epsilon^{202}\text{Hg}$ values listed in the manuscript and abstract). We then use this value in foliage as our first endmember. Our second endmember becomes the value in soil adjusted for the sole study examining a $\delta^{202}\text{Hg}$ fractionation from soil to roots (as described in Section S4 and now in the updated Section 2.6 of the manuscript). We then consider any changes in $\delta^{202}\text{Hg}$ within the plant to be a result of mixing of these to uptake mechanisms. There could be some process-based fractionation occurring within the plant during translocation of Hg, but there has not been any studies that have identified any specific processes causing that or any fractionation factors associated with any of those potential processes. We cannot assume what we simply have no evidence for yet and we do not believe that we have made any such assumptions beyond what the literature has knowledge of (i.e., one study examining soil-to-root fractionation that is included in our analyses). We believe these two additions should suffice the request from the reviewer to more robustly describe source and process differences.