

We use the following format for comments and responses:

R2C1 = Reviewer 2, comment 1. R2AR1 = Reviewer 2, Author Response 1.

R2C1 - I first congratulate the lead scientist and the entire team. The manuscript offers interesting findings providing valuable information for Hg research, especially on Hg mobility. The manuscript is well-documented and also well-written, particularly in explaining the experimental methods. I recommend this manuscript to be published in SOIL with minor suggestions.

R2AR1 – These are very kind comments and we thank the reviewer for their time and support of our work.

R2C2 - Abstract: The conciseness of the abstract can be improved. Some details can be moved to the conclusion section, ensuring the main finding remains highlighted.

R2AR2 – Briefly, the reason a conclusion section was not included as it is quite often repetition of an abstract, which serves to summarise the manuscript, and statements and suggestions made in the results and discussion section. Thus, it is our preference to not include a conclusion section for conciseness and to prevent unnecessary repetition. If the associate editor insists on a conclusion section it can be added.

Nonetheless, the abstract will be improved for conciseness and to remove some redundancies in the resubmitted manuscript.

R2C3 - Lines 53-66: The first paragraph can be re-arranged, some of the sentences are too broad and have little link to the research.

R2AR3 - [This is the same comment as R1AR3 and comes from leading author] I thank the reviewer for their suggestion (and this is a similar suggestion to reviewer 1). However, I deem this to be more related to preference of writing style. I prefer to introduce my papers with something different that provides unique information that some readers may learn and benefit from rather than repeating the same information that is stated in most every paper on a particular topic. For instance, I would ask how many papers has the reviewer read on mercury that have included some form of “mercury is a persistent, bioaccumulative, and toxic contaminant...” (or some other very well noted anecdote on mercury) within the first few sentences? Here, I have introduced information on why mercury is such a unique metal (its quantum/electronic structure), information that is not well known even to many mercury scientists.

Moreover, I would argue that this has quite important “links to the research” as the behaviour of mercury in such saturated environments is influenced greatly by the quantum/electronic structure. For instance, the volatility of elemental mercury, which we link as a potential loss process of Hg from groundwater (and surface soils and water) is directly linked to elemental mercury not forming metal-metal bonds. It is my stylistic preference that the introduction remain in this form; I do feel strongly that individual writing styles still very much have a space and a place within scientific writing.

R2C4 - Lines 122-124: is there any information on geologic materials and the site description where the samples were taken?

R2AR4 – The following sentence will be added to the methods section:

“The geology and structure of the soil/aquifer profile has been described in detail in previous works (Schöndorf et al., 1999; Bollen et al., 2008; McLagan et al., 2022).”

R2C5 - Line 140: put a space, "8 x 60 mL ..."

R2AR5 - will be changed.

R2C6 - Line 143: DI = deionised?

R2AR5 - will be changed to “deionised”.

R2C7 - Line 148: what is the desired volume or target density?

R2AR7 – We followed guidelines presented by Lewis & Sjöstrom (2010) for these aquifer materials which have been defined as sandy-gravel. The following sentence has been added to the methods:

“According to Lewis and Sjöstrom (2010), the average bulk densities range from 1.2 – 2.0 g cm³ for sands and 1.6 – 2.0 g cm³ for gravel. Thus, we deem the achieved bulk density of the columns to be appropriate for these materials, particularly as densities of the removed coarse materials are higher (solid densities are estimated at 2.65 g cm³; Lewis and Sjöstrom (2010)).”

R2C8 - Lines 171-173: The difference between EXP1 and EXP2 can be placed at the beginning of the paragraph, so readers can directly point out the difference between the two experiments.

R2AR8 – This is a good suggestion. This long paragraph will be broken up and a new paragraph will be started at “Stock solutions...”. As reviewer 2 suggests, the difference between the experiments will be better highlighted at the beginning of a paragraph in the updated manuscript.

R2C9 - Lines 172-173: The amount of HgCl concentration used in the experiment was estimated by considering the current concentration (after 55 years). Is there any data or information about the loss of Hg concentration (about 1000x) over 55 years?

R2AR9 – [this comment is the same as R1AR4] The original concentration of the solution applied during the kyanisation process was 0.66% or 6600 ppm HgCl₂ solution. Contamination at the site was from spills of this solution, which contains the very soluble HgCl₂. Thus, we know solution ~50x stronger the EXP2 and >100x stronger than EXP1 were entering the top of the soil profile and this site operated from 1904-1965 with 10-20 T of Hg being lost to the soils and aquifer. These data are stated in the study and other referenced work examining this site. Without question, experimental constraints (time) was a major factor in selecting these experimental stock solution THg concentrations, but considering the history of the site we do deem them to be applicable to this site and others. This is supported by the fact that Miretzky et al., 2005 (the only other study Hg column experiments) applied similar concentration to a similar column experiment design. This paragraph has been updated as follows to better describe these considerations:

“Stock solutions were 46.1 ± 0.1 mg L⁻¹ in EXP1 (n = 6) and 144 ± 6 mg L⁻¹ in EXP2 (n = 12) and were selected for (i) time considerations (see Figure S1.6) and (ii) these values remain between HgCl₂ concentration applied during industrial activities (6600 mg/L; spillages of this solution to the top of the soil profile) and recently measured groundwater concentrations up to 164 ± 75.4 µg L⁻¹ observed 55 years after cessation of the industrial activities at the site (McLagan et al., 2022).”

R2C10 - Figure 2: The blue and red dashed lines are not clearly seen.

R2AR10 – Figure 2 quality and clarity will be updated to reflect this request. Similarly, at the request of the associated editor, the quality/resolution of all figures has been improved.

R2C11 - Line 364: should be EXP1

R2AR11 - will be changed.

R2C12 - Lines 369: Reference of Miretzky et al. (2005) is missing

R2AR12 - will be added.

R2C13 - Lines 377-379: The authors pointed out the potential role of clay minerals or Fe/Mn oxides as an important solid-phase for the sorption of soluble Hg. In this case, the authors provide the properties of the solid-phase aquifer, such as Fe, Mn, clay, silt, sand (Table 1), and metal cations (S2). The pH was neutral to slightly basic, which support the adsorption of Hg into inorganic material. However, the clay content is low (table 1). Perhaps the authors could also provide a direct

link/evidence between the Hg sorption and the specific minerals or metals involved in this process.

R2AR13 – The clay content is 13.5%, which, along with Fe and Mn oxides present in sand/silt materials would provide sufficient mineral surfaces for sorption of all Hg applied within the columns. We do not have details on specific mineralisation of these materials and adding such here would be purely speculative. We do not believe such speculation would benefit the manuscript.

R2C14 - Conclusion: I suggest the authors include a conclusion section. The conclusion can contain a more comprehensive summary and suggestions for future work.

R2AR14 – Please see R2AR2.

REFERENCES:

Lewis, J., & Sjöström, J. (2010). Optimizing the experimental design of soil columns in saturated and unsaturated transport experiments. *Journal of contaminant hydrology*, 115(1-4), 1-13.