

Response to reviewer's comments

Title: Factors controlling dissolved ^{137}Cs activities in Matsukawa-ura lagoon, a semi-closed estuary, after the Fukushima accident

The comments from reviewers are in black, and our responses to the comments are in blue. Additions and changes to the main text in response to reviewer comments are marked in blue.

I. Referee comments 1 (RC1) and reply on RC1

Major Comments

1. Figures and Visualization

- **Figure 1:** The current layout is difficult to interpret: the legends are too small, and colors are used simultaneously to represent ^{137}Cs deposition and river catchments. I recommend separating these elements:
 - Create a first panel where colors represent only ^{137}Cs deposition, and identify river catchments with labels instead of colors.
 - Zoom in slightly so that Fukushima Dai-ichi Nuclear Power Plant is clearly visible.
 - Include an inset map showing the study area within Japan to orient readers unfamiliar with the region.
 - Describe all acronyms in the figure caption.
- **Contextual location:** There is no figure linking the lagoon to its main ^{137}Cs source (Fukushima). A simple map indicating the relative position of the lagoon and nuclear plant would strengthen context for non-local readers.

The map has been adjusted to make it easier to see the relative positions of the study area and the Fukushima Daiichi Nuclear Power Plant, and river catchments have been identified with labels (Figure 1).

1. Methods vs. Results

- Some parameters discussed in Results and Discussion could be introduced in Methods:
 - K_d calculation: Please specify explicitly how K_d was obtained (e.g., Bq m^{-3} divided by Bq kg^{-1}).
 - Voronoi partitioning: Mention and briefly describe this procedure in Methods (currently introduced only in line 199).
- In general, all analytical and data-processing steps used to represent or interpret results should be described in Methods before they appear in the Results section.

The K_d calculation, Voronoi tessellation and mass balance calculation method mentioned in the Results and Discussion have been added in materials and methods (L120-L122). The water catchments basic information tables have been moved to the Supplementary (Table S1).

Representation of Results

- **Figure 2:** The statistical plot combines all river catchment data, but much of the underlying data are not shown elsewhere (only partially in Table S1). Consider a clearer, more illustrative figure:
 - For example, a four-panel plot—one per river—showing monthly dissolved and particulate ^{137}Cs concentrations (Bq m^{-3}) as column plots.
 - This would highlight the key findings: (a) Ume and Nikkeshi contribute most ^{137}Cs to the estuary; (b) fluxes peak in summer.
 - Remaining parameters (Bq kg^{-1} , g m^{-3} , K_d) could either share a secondary axis or be kept in a table placed in the main text rather than in the Supplementary Information.
- **Tables:**
 - **Table 1** seems to contain background or supporting data rather than primary

results. Consider moving it to the Supplementary Information and bringing **Table S1** into the main text.

Since no seasonal trends were observed in the ^{137}Cs concentrations in river water, the currently used box plots were retained (Figure 3), but the time series of dissolved and suspended ^{137}Cs and other parameters were added to the supplementary (Figure S1, Table S2).

1. Organization of Results and Discussion

- **Section 3.2 (Lagoon results):**
 - Subsection 3.2.1 is titled “Relationship between dissolved ^{137}Cs and salinity” but includes all parameters and does not first present the spatial distribution of ^{137}Cs itself. Begin with a clear presentation of particulate and dissolved ^{137}Cs distributions—perhaps using isosurface plots or panel figures similar to those for rivers (even a subset of months for clarity).
 - The investigation of relationships with salinity fits better in the Discussion rather than Results. Splitting Results and Discussion would improve flow and clarity.
- **Figure 3 (salinity relationships):** The panels suggest little to no correlation. Consider exploring:
 - Subsets of data (individual rivers, specific lagoon stations near river mouths, or specific seasons).
 - If no relationship exists under any subset, move these plots to the Supplementary Information or remove them.
- **Section 3.2.2 and Figure 4:** Similar concerns apply here—repeated salinity analysis without a clear signal. A deeper targeted analysis or removal is advised.
- **Figure 5:** This clearly shows seasonal variability (higher in summer, lower in winter) and could be placed under a dedicated subsection focused on temporal variability rather than under temperature–salinity relationships.

The title of the subsection 3.2.1 has been changed to "Changes in parameters due to salinity" (L203). The figures of salinity and each parameter have been left intact to clarify whether there is a relationship with salinity (Figure 4), but the figures showing the relationship with salinity at each location have been removed. Instead, figures showing the time series of dissolved and particulate ^{137}Cs has been added (Figure 5), which will lead to the next subsection, "Seasonal variation of dissolved ^{137}Cs in the lagoon". Other parameters have been added in the supplementary (Table S3, S4). In addition, the context has been restructured to make the results and discussion clearer.

A subsection focusing on seasonal variations has been added as subsection 3.2.2, and the effects of water temperature have been integrated into this subsection (L236-L262).

Section 3.3 (Modeling)

- Add a schematic diagram summarizing the model setup and assumptions for desorption/dissolution of Cs. Values adopted from the literature could be noted on the schematic for transparency.
- Include final model results within the main text (e.g., from Figure S1). Panels a–c of Figure S1 are particularly informative, illustrating that most ^{137}Cs is discharged in August and that the particulate fraction dominates.

We added column plots as Figure 7, showing estimated fluxes from the river, and Figure 8, showing the model results estimating the flux of dissolved ^{137}Cs supplied to the lagoon. These make the model results easier to understand visually. The original table showing the model results has been reorganized and moved to the supplementary (Table S5). Additionally, we added schematic illustration of the model for dissolved ^{137}Cs in the lagoon as Figure 2.

1. Broader Perspective and Implications

- Quantify, if possible, the flux of ^{137}Cs from the lagoon to the Pacific Ocean, addressing one of the paper's stated aims.
- Place findings in a broader context:

- How significant are lagoon discharges compared with other ^{137}Cs sources to the Pacific?
- Are there similar studies (in Japan or elsewhere) that could provide comparison?
- What are the implications of these results for understanding radionuclide transport or for monitoring strategies?
- Suggest potential next steps or research directions based on the conclusions.

We quantified the flux of ^{137}Cs from the lagoon to the Pacific Ocean (Figure 8a) and compared it with studies reporting fluxes from other rivers in Fukushima Prefecture (Niida et al., 2022) (L307-315). Niida et al. (2022) calculated the fluxes of ^{137}Cs flowing out the Pacific Ocean from rivers closer to the Fukushima Daiichi Nuclear Power Plant than Matsukawa-ura lagoon during high flow conditions and found that some of this flux was equivalent to the flux of ^{137}Cs flowing out the Pacific Ocean from Matsukawa-ura lagoon. This highlights the significant impact of the flux of ^{137}Cs flowing out the Pacific Ocean from Matsukawa-ura lagoon compared to other rivers.

Furthermore, because the impact of ^{137}Cs dissolution from bottom sediments is significant (Figure 8b), the report also proposed future monitoring strategies in the Fukushima coastal areas, such as strengthening analysis of marine sediments and their pore water (L329-336).

II. Referee comments 2 (RC2) and reply on RC2

Specific Comments

Mass balance section

L244: The authors estimated the supply flux from sediments by using the tidal prism to calculate “the volume of seawater inflow per 12 hours,” and assuming that this inflowing water uniformly reaches the lagoon’s mean concentration within 12 hours. The difference was then attributed to sedimentary fluxes. However, this estimation has several critical flaws:

- (1) The tidal prism does not represent a “pure inflow” of new seawater. In reality, inflow and outflow occur simultaneously, and the exchange efficiency is less than 100%.

As suggested by the reviewer, the seawater that flows into Matsukawa-ura lagoon is not 100% replaced, but I wanted to suggest a qualitative trend that there is a seasonality. We have addressed the lack of this as follows:

" Although the tidal prism does not fully represent the entire exchange of estuarine water with oceanic seawater entering from outside the lagoon, it is used here—based on the simplified assumption that pure seawater flows in from the open ocean—to help understand the monthly variation in the influx of dissolved ^{137}Cs and the key factors maintaining relatively high ^{137}Cs concentrations in the lagoon" in L166-169..

- (2) The assumption that inflowing water reaches the lagoon-wide average concentration uniformly within 12 hours is unrealistic. Observations clearly show higher concentrations in the inner lagoon, demonstrating strong spatial heterogeneity.

The following texts were added to L242-L248 in the main text.

“The water depth of Matsukawa-ura lagoon is very shallow, averaging about 1m, and it has been indicated that about half of the water is exchanged during one tidal cycle (Kohata et al., 2003). For the simplicity in clarifying the seasonality of dissolved ^{137}Cs in the lagoon, we used the average concentration of dissolved ^{137}Cs among several sampling stations which could might represent the dissolved ^{137}Cs levels in the area of the lagoon. Actually, in February 2022 and 2023, dissolved ^{137}Cs concentrations ranged 2.5-9.0 and 3.5-8.3 Bq m⁻³ with small variation, respectively, with weighted mean dissolved ^{137}Cs concentrations of 5.3 and 5.5 Bq m⁻³, but, weighted mean dissolved ^{137}Cs in June 2022 were 17.0 and 15.0 Bq m⁻³, respectively, about three times higher than winter season. These results suggest that seasonal variations are more influential than variations in concentrations between areas within the lagoon.”

- (3) The authors assume a constant tidal prism exchange volume throughout the year, yet still discuss seasonal (summer vs. winter) differences. This is logically inconsistent.

For these reasons, the flux estimation is not valid and should be considered inappropriate.

This study focused on the seasonality of dissolved ^{137}Cs in Matsukawa-ura lagoon, and the calculations showed that dissolved ^{137}Cs in Matsukawa-ura lagoon is seasonal and would like to suggest the influence of water temperature. In this study, we measured the water depth at the station WKO, where connects with Pacific Ocean, from December 29, 2021. The mean water depth during the study period was 1.9 m, with a maximum of 2.2 m and a minimum of 1.7 m, and the difference between the maximum and minimum values was about 50 cm (Figure S2). Therefore, the exchange volume in Matsukawa-ura lagoon was assumed to be constant throughout the year, making it possible to compare between seasons (L283-287).

L260: The method used to estimate the decrease of ^{137}Cs in bottom sediments is unclear. Given that the mass flux calculation itself is subject to large uncertainties, presenting the results as quantified values—and further highlighting them in the abstract—risks giving readers an impression of unwarranted accuracy. Considering the likelihood of these numbers being cited in future work, the current presentation lacks scientific integrity. This section should be deleted or, at the very least, thoroughly rewritten.

We will remove the section on estimating the decrease of ^{137}Cs in sediments, suggest only seasonality. The abstract will be revised accordingly to exclude quantitative values and instead emphasize the seasonality.

L31 : Sanial et al. (2017) is not a study of sedimentary fluxes and its citation here is inappropriate. The positioning of this reference should be reconsidered.

Introduction : Kambayashi et al. (2021) is a highly relevant prior study and should be cited in the Introduction.

We remove Sanial et al. (2017) as a citation in the introduction and add Kambayashi et al. (2021) as a new citation in the introduction and discussion for the comparison with our study (L59-L65, L291-302). Kambayashi et al. (2021), studied in Matsukawa-ura lagoon in 2014-2016, calculated the mass balance of dissolved ^{137}Cs in Matsukawa-ura lagoon, and suggested that supply from porewater in bottom sediments increased in the summer, and the fluxes of dissolved ^{137}Cs from bottom sediments were 287-293 MB day⁻¹ in summer. The estimated fluxes of dissolved ^{137}Cs from bottom sediments during the summer were consistent with our results. These results suggest that the amount of ^{137}Cs deposited in marine sediments in the early stages of the accident and dissolved and dispersed into the seawater may not change significantly, whether three or ten years have passed since the accident. Thus, as more time passes since the accident and the supply of ^{137}Cs to coastal waters from rivers decreases due to the decommissioning of contaminated soils in their catchment areas, the contribution of the dissolution of this nuclide from bottom sediments may have continued.

Fig. 4 : The spatial arrangement of sampling sites is difficult to understand. I recommend improving the map by adding scale bars, landmarks, or other visual cues to make the locations more intuitive.

We have done (Figure 5).