Reply to Referee #1

The paper presents a comprehensive dataset on the presence of plutonium isotopes, Pu-239 and Pu-240, in eight sedimentary reservoirs from different regions of South America located within the 30–60°S latitudinal band. The authors also provide dating results based on the Pb-210 technique, complemented by Cs-137 data, which together allow the interpretation of Pu profiles in relation to periods of atmospheric nuclear testing.

A broad Cs-137 peak is observed in all cores, and most of them display both a primary and a secondary Pu peak. The most recent years, up to the 1970s, consistently show the lowest Pu-240/Pu-239 ratios, indicating a strong influence from the French nuclear tests conducted in French Polynesia up to 1974. The interpretation of the results is very thorough, the figures are of high quality, and the overall manuscript is solid. In my view, the paper could be published in its current form with only minor revisions and incorporating some suggestions from my side.

Many thanks for these constructive and encouraging comments! If the editor decides so, we will provide a revised version of the manuscript incorporating all the requested modifications.

General suggestions:

- I recommend adding a table summarizing the dates and yields of the French nuclear tests in French Polynesia, which would serve as a useful reference when interpreting the Pu profiles.

Thank you for your suggestion. A table or a figure will be added to the supplementary material of the revised version of the manuscript to help readers better understand the different periods of nuclear weapons testing, including the French testing period.

- The British tests in the premoratorium period in Australia and in the Christmas Islands should also be named and considered in the Introduction.

Thank you for your suggestion. We have mentioned the British tests in Australia in the method section (l.219-221): “British tests conducted in Australia approximately ten years prior to the French tests were also excluded due to their low yield and predominantly regional fallout deposition (Child and Hotchkis, 2013; Dicen et al., 2024) likely not reaching South America.”

We will also introduce the British tests conducted in Australia and on the Christmas Island during the pre-moratorium period in the introduction section.

- There is a recent paper reporting the Pu-239 signal in Antarctic ice cores with sub-annual resolution: **Shin et al., Sci. Adv. 11, eadv1172 (2025)**. This study provides complementary information to Koide et al. and could be used to support the authors’ conclusions and offer additional evidence regarding the influence of French tests in the Southern Hemisphere.

Thank you very much for this suggestion. We did not use this paper in our discussion, as the $^{240}Pu/^{239}Pu$ atomic ratios were not reported. However, we agree that given the high temporal resolution of ^{239}Pu measurements and the different locations of the ice cores compared to those in Koide’s paper, it would be interesting to include this paper in our discussion of the revised manuscript.

- I have serious concerns regarding the chemical procedure applied to the samples. It appears that, after the leaching or fusion step, no coprecipitation step was performed prior to Pu purification using TEVA resin. Such a coprecipitation step is critical for removing potential interferences in mass spectrometry and for achieving reliable chemical yields. The authors should clarify why this step was omitted. Additionally, quality control of the results, e.g., through the analysis of IAEA reference materials or other well-characterized samples with similar matrices and Pu concentrations, should be reported to validate the results.

We agree that a coprecipitation step is a critical step to improve the overall performance of the Pu separation from others actinide and reduce the matrix effect. However when measuring a large number of samples, this additional step is disadvantageous for routine analysis, as mentioned in a recent method paper published by one of the co-authors (Dicen et al. 2026):

"Coprecipitation is sometimes employed to improve ^{238}U separation factors [20, 36, 37], which were found to be one to three orders of magnitude greater than those achieved in the tested procedures (Fig. 3, Table 3). However, as it requires a series of additional steps needing additional pH adjustments, reagents, and solid-liquid separation, coprecipitation is not convenient for routine analysis of a large number of samples. For separation procedures for later measurement in an ICP-MS/MS, residual ^{238}U after the chemical separation step is addressed within the instrument during analysis [25, 36]."

In their study, the authors showed also overall good performance without the coprecipitation step:

"Despite using a minimal amount of sample and relatively simple procedures, the values that we have obtained are comparable with the recovery rates and separation factors reported in the literature (Table 3). Among the modified leaching and separation procedures tested in this study, method E achieved a lower recovery rate than FD (69 ± 8% vs. 84 ± 3%), but a higher mean 238U SF (1.29 × 10^4 vs. 1.00 × 10^4). These results indicate that HNO_3 leaching is sufficient to achieve acceptable recovery rates comparable to the values reported in the literature, and even a low sample mass does not require a coprecipitation step to achieve high ^{238}U separation factors." (Dicen et al. 2026, Ultra-trace 239Pu and 240Pu determination in soils using optimized leaching, vacuum-assisted TEVA separation, and isotope dilution ICP-MS, DOI: 10.1007/s00216-026-06614-y)

We can cite also different studies that did not use a coprecipitation step in environmental studies and obtained satisfactory results (Dowell et al 2023 DOI : 10.1039/D3AY01030A; Ketterer et al 2004 DOI : 10.1016/j.scitotenv.2003.09.016)

IAEA reference materials, procedural blanks, and internal standard results were provided in the online data files for each lake. We agree that presenting these QA/QC results in a dedicated supplementary table would provide a clearer overview of the analytical performance. We can summarize this information in a table in the Supplementary Information and refer to it in the manuscript.

- Another issue concerns the nature of Pu contamination in the samples. Several studies reporting Pu concentrations in samples from South America and Africa have shown that contamination can be highly heterogeneous, meaning that duplicate aliquots of the same sample may yield different Pu concentrations (see, for instance, (Chamizo et al., 2011, https://doi.org/10.1016/J.NIMB.2011.04.021) and (Chamizo et al., 2020, https://doi.org/10.1016/j.scitotenv.2020.139993). Have the authors investigated the nature of contamination in their sediment profiles by analyzing duplicate samples, particularly in those layers exhibiting lower Pu ratios?

We agree that duplicate aliquots analyses would have strengthened the study but one of its main limitations was the small quantity of material available for each lake sediment sample (1 to 4 g). To achieve the highest temporal resolution of the records, we chose to keep the smallest sampling resolution possible (0.5 to 2 cm). Most of the time, there was either no remaining sample mass or insufficient mass to perform duplicate analyses.

We adopted this approach because we considered that maintaining a continuous and high-resolution record was more important for accurately capturing temporal variations in the $^{240}Pu/^{239}Pu$ atomic ratio and for minimising the influence of sample heterogeneity, particularly along the Pu and Cs activity peaks. In addition, lake sediments are generally well mixed and can be considered relatively homogeneous at the scale of the analysed sample.

Nevertheless, duplicate analyses were performed for selected samples from two lakes out of the eight lakes during the completion of Pu activity profile, which allow us to compare measurements obtained at different

radiochemical procedure/analytical dates. If the editor permits resubmission, these duplicate results can be included in the online dataset or in our replies to the referees.

A further question relates to the consistently low Pu-240/Pu-239 ratios associated with French tests, even those with the highest yields. Low ratios are observed across different sediment cores throughout the entire French testing period. Is there an explanation for this? Tests conducted by other countries generally show a relationship between yield and Pu ratios, with higher yields corresponding to higher ratios. A comment on this would be welcome.

Thank you for your comment on this important issue. There are probably different signatures between low-yield and high-yield thermonuclear tests conducted by the French government in Polynesia, but very few published papers or grey literature documents that could indicate a different signature. The value range of 0.035 ± 0.015 for $^{240}Pu/^{239}Pu$ was established based on lagoon sediments (Chiappini et al. 1996; Chiappini et al, 1999) and on topsoils and coral bedrocks from Mururoa and Fangataufa (Hrnecek et al. 2005). Samples collected from both atolls mainly represent Pu deposition from barge tests and safety tests (Chiappini et al. 1999; Hrnecek et al 2005), as well as potentially local fallout from the other low-yield tests.

Higher ratios could be expected for the high-yield tests (thermonuclear tests, 1968-1970, max. 2.6 Mt) as seen in tests conducted by the US (max. 15 Mt), the former Soviet Union (max. 50Mt) or China (max. 4Mt). The design of French thermonuclear bombs may also have limited the time window of the neutron capture, which may have reduced the production of ^{240}Pu. In addition to the "lower "yield of the French thermonuclear tests compared to those conducted by the US or the URSS (>> 2Mt), this would have resulted in a lower $^{240}Pu/^{239}Pu$ atomic ratio (between 0.03-0.07 ?).

As a precaution, we used a source with an end-member value of 0.035 ± 0.015 for the French tests. We acknowledge this limitation and can address better this issue in the revised version of the manuscript.

Minor remarks:

Graphical abstract: What is the meaning of the grey bar in Lake Natri and Lake Laja plots?

The grey bars represent instantaneous event deposits identified during core logging or XRF analyses and do not represent continuous sedimentation. We will remove them from the graphical abstract.

Line 95: When naming previous works on the presence of FF in South America, please include: 10.1016/J.NIMB.2011.04.021

We agree and will include this paper, which is one of the few studies to present Pu measurements for the South American continent (*Presence of plutonium isotopes, ^{239}Pu and ^{240}Pu, in soils from Chile, Chamizo et al. 2011, doi:1016/j.nimb.2011.04.021).*

Line 165: Was Am241 detected in the cores by gamma spectrometry? What activity levels were measured?

The activity concentration of ^{241}Am , when detected, is indicated in the supplementary figures of each lake's age-depth model alongside ^{137}Cs and ^{210}Pb. The activity of ^{241}Am does not exceed 2 $Bq.kg^{-1}$. These values are also available online in the data file.

If the editor authorises us to resubmit our manuscript, we can add a sentence to the results section to discuss the ^{241}Am activity associated with the ^{137}Cs or Pu activity peak.

Line 211: The GF average value for the Southern Hemisphere should be used here. As far as I know, this integrated value for the Pu240/Pu239 ratio includes also the contribution from the premoratorium period. This is just the integrated signature from soils that are assumed to represent and average value for the whole nuclear era.

Thank you for your comment, we will clarify this in the revised version of the manuscript.

Lines 225-230: The uncertainties formula should be moved to SI. On the other hand, I think a subscript is missing in the last "Ds" of the formula in line 230.

We agree to reduce the number of formulas to make the manuscript more readable. We will move uncertainties formulas to the supplementary file. We are unsure whether we understand the last "Ds", a subscript *i* might be missing form the last D as the sampling resolution may change across the lake profile.

Line 240: Are the Cs137 profiles corrected by decay using the dating of the core?

Yes, the ^{137}Cs activity profiles presented in Figure 2 were decay-corrected from the gamma measurement date to 2025, whereas the ^{137}Cs activities presented in the age-depth models of supplementary file are not decay corrected and correspond to the measurement data (available as table in the online database). We will precise "decay-corrected to 2025"in the Figure 2 caption.

Figure 2: "R-FR" should be changed by "R-FF" for consistency.

We agree and this will be changed in the next version of the manuscript.

"Grey horizontal bands mark instantaneous events, which do not represent continuous sedimentation". What is it the meaning of this?

An event deposit in lake sediments reflects the input of a detrital fraction that differs from the "continuous" (background) sedimentation. It commonly takes the form of turbidite deposits, which correspond to material transported or remobilised by various geodynamic or climatic events such as floods, glacial lake outburst floods, avalanches, hurricanes, earthquakes, tsunamis, volcanic eruptions, etc. Flood deposits are usually the most common type of event deposit observed in lake sediment records.

Although these deposits are episodic and represent a short time period (hours to days), they are included in the continuous sedimentary record. Therefore, they should not be considered when reconstructing the history of contaminant deposition.

It would be helpful including the latitude of the cores below the names in each plot, to facilitate the interpretation of the data.

Thank you for your comment. It will be added in the revised versions of Figure 2 and Figure 3.

Line 389: It is not clear the meaning of the Ipeak. Does it mean the integrated inventory for the peak in the 1970s?

In the method section, we have described the meaning of the I_{peak} (l.200-202): "We defined I_{peak} (Bq·m^{-2}) as the partial inventory representing Pu deposition during the nuclear weapons testing period, restricted by the endpoint depths of the $^{239+240}$Pu and ^{137}Cs peaks"

In the revised manuscript, we can add that it should "be indicative of the period between the first tests being detected in the southern hemisphere in 1950 and the end of the atmospheric nuclear weapon testing conducted in French Polynesia in 1974". We should also warn the reader that it doesn't only represent the atmospheric Pu deposition over this period (such as reference sol site), but this is an integration of atmospheric Pu deposition and catchment Pu input.

Line 391: Does the Ipeak appears in Table S2?

The I_{peak} does not appear in Table S2. However, we can add the I_{peak} and I_{total} to Table S2 in addition to the Figure 5.

Fig. S3: Please, change the unit of liter by L (not "l").

Yes, it will be changed in the revised supplementary file.

Table S2: How is it calculated, the SF factor for Th and U? Why is it so different between samples? Is there a correlation between bad SFs and Pu ratios?

In the same recent paper, published by one of the co-authors, the authors summarizes the distinction between U and Th separation factors (SFs) for the two methods:

"^{238}U separation factors were calculated as the quotient of the ^{238}U to ^{242}Pu recovery rates after separation on TEVA columns. The initial ^{238}U concentrations were obtained by measuring the samples by gamma spectrometry prior to leaching, or by analyzing aliquots of the solutions resulting from fusion digestion by ICP-MS. It should be noted that for the modified leaching procedures (methods A–E), partial reduction in total ^{238}U concentration already occurs due to the non-leaching of silicate-bound U. Consequently, the separation factors of leaching procedures reflect the combined effect of incomplete U extraction during leaching and U removal on the TEVA column. On the other hand, fusion digestion (method FD) dissolves all matrix phases including all U in the soil, so the SF reflects only the TEVA separation performance. Thus, the separation factors of the modified leaching and fusion digestion procedures are not directly equivalent, although both represent the net U reduction achieved relative to Pu in the final solution after the TEVA separation."

(Dicen et al. 2026, DOI: 10.1007/s00216-026-06614-y)

The U and Th SFs serve only as qualitative indicators of the effectiveness of the Pu separation from U for the leaching procedure, whereas they provide a quantitative measure of separation performance for the fusion procedure. In the fusion procedure, the SFs depend on the initial U and Th concentration in the sample, whereas in the leaching procedure it depends on the fractions of U and Th that are extracted during the leaching procedure.

There is no direct correlation between poor separation factors and Pu ratio values. However, lower separation factors can increase the uncertainty of the ^{239}Pu concentration measurements because of residual ^{238}U in the final solution. We can add a short explanatory paragraph to the revised table S2.