Reply to Referee #2

The authors describe measurements of Pu isotopes in lake sediments on a longitudinal transect in South America, to resolve different eras of nuclear bomb testing. This is to produce a better time calibration for dating sediment cores from which to reconstruct environmental records.

Using the distinct Pu isotopic composition produced by different iterations of nuclear bomb production authors successfully constrain three periods of Pu deposition to the study region. I agree with the authors stated goals and conclusions that this can refine our use of fallout radionuclides to provide age constraints on environmental records.

Some specific line comments follow which may be helpful for the authors in revising a final version for publication,

We thank you for your time and for your overall positive assessment of our manuscript. We also appreciate your constructive suggestions which would help to clarify some statements if the editor invites us to resubmit the manuscript.

Abstract

Line 34: unsure of intent here, is it that such extensive environmental changes are particular to South America, or that reliable time markers are essential for South America? I would prefer the latter if this can be clarified

Thank you for your comment. If we are allowed to resubmit our manuscript, we propose the following modification: "Reliable time markers are essential for dating sediment sequences and reconstructing environmental degradation in regions that have experienced rapid and extensive environmental changes, particularly in South America."

Line 36: little unclear how we have gone from needing reliable tracers to identifying distinct fallout sources…
Line 38: and then continue to describe only the fallout sources, and no link back to how this helps towards reliable time marker and then to understanding environmental change.
We agree that the beginning of the abstract needs to be rewritten slightly, so that the link is clearer. Here are some suggestions:

"In this respect, the $^{240}Pu/^{239}Pu$ atomic ratio can be used to distinguish distinct sources of plutonium fallout and, consequently, to identify the chronological succession of different nuclear weapons testing periods, including radioactive fallout from French atmospheric nuclear weapons tests (NWTs) conducted between 1966 and 1974 at the Mururoa and Fangataufa atolls (French Polynesia)."

Introduction

Line 62: is broad Cs peak attributable to post-depositional mobility? Maybe clarify if this is the case? (I believe that it is).

Thank you for your comment, we agree that this process could explain the broad shape of the ^{137}Cs peak at deeper depth. However, it could also reflect an extended period of Cs deposition as illustrated by the atmospheric deposition record from Buenos Aires (Fig. S2) and could potentially be observed as elevated Cs activity extending to shallower depths, above the depth of ^{137}Cs maximum activity peak. We can discuss this phenomenon in the results and discussion section of the revised manuscript.

Material and methods

Line 147: geometry of vial/ volume, and typical sample masses should be reported as well.

The volume of material used to measure dry bulk density may have varied depending on the person who subsampled the core. Either a small syringe (1.5cm in diameter) or half of the core was used, with different sample thicknesses. The mass range used for each lake is reported in Table S2. We can add to the revised manuscript that between 1 to 4 g of material per sample.

Line 188: I've not seen this expression, surface activity, before. More commonly called inventory, stock, or areal activity? In line 196 you use inventories, or is this only for "total" inventory?

The expression "surface activity" allows us to describe the Pu accumulation within a given sediment layer, normalized to a surface area unit, whereas the "inventory" represents the total accumulated stock. This expression is probably more commonly used in soil studies (e.g, Alewell et al. 2017), but it has also been used for lake sediment (Foucher et al. 2021).

We will carefully consider your comment regarding the terminology in the revised manuscript.

Line 198: Can we calculate a reliable flux without taking account of dispersion? I.e., the core profiles are not true histories even if the peak position is reliable because dispersion broadens the peak, and reservoir effect (in-lake or watershed) leads to long tailing of fallout (often) into contemporary sediments.

Thanks for your comment. This is indeed an important point when calculating lake contaminant fluxes and comparing fluxes among lakes. When considered together with the $^{240}Pu/^{239}Pu$ atomic ratio (Fig. 5), Pu fluxes reveal a similar overall pattern among the lakes, although the absolute flux values differ. Despite different environmental context between La Estanzuela (lake surface: 0.06 km2 catchment area:1 km2) and Rincon del Bonete (lake surface: 465-1485 catchment: lake ratio: 25-33), both lakes show also similar trends in Pu flux and $^{240}Pu/^{239}Pu$ atomic ratio. In addition, Figure 5 does not show the fluxes from the top sediment (recent sediment) but only the flux associated with the Pu deposition peak.

We can discuss the potential post-mobility of Pu in sediments and to emphasize both the limitation and the advantages of using Pu fluxes associated in combination with the 240Pu/239 Pu atomic ratio when comparing lakes.

Line 204: please cite the number of reference soil sites.

We suggest adding these numbers to the Figure 5 as the soil inventories and associated study methodology are not part of this work. We only used these data for comparison with the inventories of the studied lakes.

Line 210: endmember is a key word that could be used here, some readers (including myself) would think about the distinct sources this way.

Ok, it will be included in the methodology section and in other parts of the text where necessary.

Line 231: brief justification for how 7% was arrived at? This is based on volume of the cylindrical sediment plug?

Appleby (2001) recommended using 7% as "a reasonable value for p" when computing CRS model. This value was also used in the serac package (Bruel and Sabatier 2020) and we therefore used it as a representative estimate of the average uncertainty of the dry bulk density. It would have been more precise to calculate the uncertainty for each measurement based on the uncertainties associated with sample mass, volume, and thickness measurements.

We will therefore assess the uncertainty in dry bulk density based on our data and determine whether the 7% value should be revised in the revised manuscript.

Results and discussion

Line 240: comparison between Cs and Pu profiles is not articulated clearly. Should we expect two Cs peaks as well? Is the single peak then due to higher dispersion/diffusion of Cs?

Thank you for your comment, we agree that this part might need revision and agree to clarify Cs and Pu profile description. Based on the closest atmospheric record from Buenos Aires (Fig. S2) we should not expect two peaks but one broad peak. Please see also my reply to your comment on l.62 (Introduction).

Line 248: there should be citations available here I believe, consider …

We agree, we suggest adding Hancock et al. 2011 and Appleby et al 2003 to this statement "Additionally, although 137Cs is known to be mobile, it is unlikely that the depth of its maximum activity concentration has shifted significantly".

Line 250: the verification should be quantified. What are 210Pb ages of Cs maximum? Include this in Table 1?

We agree, we will add the 210Pb ages of Cs maximum in the table 1 to corroborate this statement "The 210Pbex-based chronology, verified by the 137Cs peak, allowed us to quantify the sedimentation rates across all lakes and estimate the age of sediment deposition using the CFCS model (Fig S6-S13) "

Line 251: why choose CFCS over CRS when it's assumptions would appear to be more restrictive?

We have detailed the choice of each model in the supplementary files. The CRS model was often not applicable because it was not possible to determine the total inventory of excess 210Pb, which required measurements to be taken at greater depths (Appleby et al al. 2001; Mabit et al. 2014). When applicable, the CRS model generally produced ages that were too old when compared with

137Cs peak depth, whereas the CFCS model produced better agreement between the 137Cs peak and the corresponding historical period of 1964-1965. We could provide more detail in the supplementary files.

Fig. 2 is a tough read. Consider label each pair a-h

Ok, we will label each pair of Fig 2.

Fig. 3: I like this Figure, it is effective. I wonder, while we see a shift to low Pu ratios corresponding to FF , most cores see a gradual increase in ratios to present. Why is this? Is this due to much larger GF total deposition and slow mobilization of Pu from watershed?

Yes, exactly. We tried to explain this phenomenon (l.416-422) :

"The higher deposition of FF in Uruguay is also reflected in the low 240Pu/239Pu atom ratios measured in recently deposited sediments (post-tests period), where the average did not exceed 0.10 for EST and RDB, indicating a recent FF contribution between 30-50% (Fig. 3). This suggests that the total abundance of Pu fallout from French tests in Uruguayan soils was not only higher than in other regions but also results in a persistent FF signature in soil particles, which are eroded and transported to the lake basins. This interpretation is the most likely explanation for the continued presence of mixed French and global fallout Pu signatures in sediments more than 50 years after the end of atmospheric nuclear weapons testing."

While for the other lakes outside of Uruguay, we can assume that the deposition of French fallout was lower in comparison to GF, as suggested by the lower fraction of the FF from the Ipeak, and that in the present day this is mainly GF labelled soil particles and sediment that are remobilised from the catchment terrestrial surfaces. This is something we would like to investigate better as top sediment surface can also be an indicator of the different fallout sources that reach each site.

Line 389: Peak inventories meaning only the Pu in the main peak, but not in the entire core? Is this intended to reduce noise from post-depositional reservoir effect i.e. long tailing of Pu into recent sediments? Maybe explain the significance of I_{peak}?

Yes exactly, the term has been explained in the method section, and as suggested to the Referee #1 we can clarify it as :

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 around 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.

Fig. 5: having trouble with the y-axis not being continuous.

Ok, we will try to arrange the Figure 5 to improve readability.

The Pu inventories might be corrected for sediment focusing, if the true atmospheric flux is desired, e.g. using 210Pbex if those numbers in soils are better known, or can be estimated from rainfall, i.e., Pu-corrected = Pu-lake*Pb-soil/Pb-lake.

In our study, we used Pu peak inventory as an indicator of the Pu deposition across the eight lakes, while acknowledging the limitations mentioned above. Our goal is not necessary to derive atmospheric fluxes from these inventories, as we do not have possibility of correcting the inventories for inputs from the catchment.

We have only one reference soil near by La Estanzuela pond that could be used to correct the Pu sediment fluxes of EST and RDB. We could also try to use the 210Pb data available in the database compiled by Dicen et al. (202) which include 137Cs and 239+240Pu inventories from reference soil site in the Southern Hemisphere but it would require a substantial amount of time to extract the 210Pb profile and calculate the corresponding soil inventories.

Line 420: re-evaluation?

Thank you for noticing this mistake.