Answer to comments of RC1 on egusphere-2026-3608

Review #1/A

We thank the reviewers for their helpful comments. Based on the reviewer's suggestions we have re-written many parts of the manuscript and have now based our approach to decipher the role of primary production on a more robust statistical approach using Al and Ba as proxies for lithogenic and biogenic export of Pb instead of total Si, respectively. Fe and Ca) were further used to identify basaltic tephra layers which showed distinct different Pb isotope signatures. Furthermore, we have tested the role of Fe/Mn oxides in sediments as carriers of authigenic Pb precipitated from the water phase. The new statistical approach shows that biogenic export of Pb appears to be of lesser importance at the sites St9 and St16 compared to lithogenic Pb phases, although both cores indicate high productivity based on Ba concentrations. However, St15 (deepest core) shows a relatively large proportion of biogenic Pb and a clear relationship between Pb isotope ratios and the amount of biogenic Pb in the sediments. This relationship suggests that different Pb pools exist in the water phase -possibly due to selective dissolution and scavenging of isotopically different Ph phases in dust or other / biogenic particles in the water phase. Fe/Mn oxides have been found to be separated and mobilized in our sediments due to high sedimentation rates and the relatively high input of organic matter in this productive area. We assume that redox processes in the sediments such as the dissolution of Fe- and Mn-oxides and the coupled mobilization and dislocation of Pb blurred the statistic relationship between productivity indicators and Pb concentrations and Pb isotope signatures.

In general, our data suggests that Pb isotope ratios and their relation to biogenic and lithogenic phases in deep sea sediments show large variations, which are difficult to decipher due to the manifold processes which alter sinking particles in the water phase and alteration of Pb bearing phases in the sediments. Thus, our data and the used statistical approach shows only a broad brushstroke but display the difficulties in using Pb isotope ratios extracted from deep sea sediments to reconstruct sources or paleo ocean currents.

A1: This manuscript presents Pb concentrations and isotope ratios from three sediment cores collected near South Georgia and the South Sandwich Trench and explores their relationships with sedimentary Si and organic carbon. The potential influence of biological productivity and particle cycling on Pb isotope signals is an interesting and important topic. The dataset is valuable, and the manuscript has the potential to make a useful contribution to understanding Pb cycling in the Southern Ocean.
However, the central interpretation that primary productivity and particle transformations in the water column alter the Pb isotope signatures ultimately preserved in deep-sea sediments is currently not sufficiently constrained by the bulk-sediment measurements.
In particular, the contributions of lithogenic, authigenic and biologically associated Pb have not been separated, while total sedimentary Si is treated as a direct proxy for biogenic silica and productivity.

In order to more clearly distinguish possible Pb sources to the sediment, in the revised version of this manuscript we followed an approach to distinguish lithogenic from biogenic Pb by

normalization to lithogenic tracers (Al and Ti) as suggested by the reviewer. We did not distinguish "authigenic Pb" exported by Fe-Mn oxyhydroxides as Fe and Mn phases in our sediments have been altered due to related redox conditions and likely have released/trapped Pb during dissolution and re-precipitation at the oxygen boundary layer. In fact, our data supports the assumption that biogenic Pb is similar to authigenic Pb as both phases are likely not to be separated but associated in marine particles (Chuang et al., 2014). Instead of TOC or Si, we now used Ba as a proxy for productivity, to account for processes of mineralization and dissolution in the water phase, especially at large water depth.

A2: The manuscript also does not adequately compare its results with existing authigenic Pb, Nd isotope and opal-flux records from the same general region.

Efficient Extraction of Past Seawater Pb and Nd Isotope Signatures from Southern Ocean Sediments (Huang, 2021) (The robustness of our improved leaching method was validated via direct comparison of Pb and Nd isotope signatures with actual seawater, porewater, and corresponding sediment.

We have now included the aforementioned references (Huang et al.,2020 and 2021) and a comparison with authigenic Pb isotope data in the manuscript. Our observed Pb isotope signal is almost identical to the one described in Huang et al. (2020 and 2021). However, we are seeing problems applying this approach to our sediments or in general at sites located in the South Atlantic diatom ooze-belt, which contain relatively high TOC concentrations, inducing Fe-Mn oxide dissolution. Moreover, many of these pelagic sediments contain basaltic tephra layers from the South Sandwich Trench. Those pyroclastic materials are Fe-rich and altered in the sediments, in our dataset they clearly show a diverging Pb isotopic fingerprint -. We have eliminated the tephra layers from our statistical analyses, which has furthermore indicated that the Fe and Mn phases in our sediments have been dislocated in the sediments.

A3: I therefore recommend major revision. The study is potentially publishable, but the authors should more carefully **distinguish observations from interpretations, consider alternative explanations and substantially qualify some of their mechanistic conclusions**.

As recommended by the reviewer, we have now avoided over-interpretation of our data regarding the influence of primary productivity. The now used normalization approach using Al or Ti for lithogenic and Ba for biogenic Pb phases provides a more robust interpretation.

Major comments

Pb isotope compositions of bulk sediment and the identification of Pb carriers

A5: The Pb isotope measurements were performed on total digestions of bulk sediment. Consequently, the measured Pb represents an unknown mixture of lithogenic Pb, authigenic or

seawater-derived Pb, Pb associated with biogenic particles, and potentially anthropogenic Pb. **Changes in the relative proportions of these components could produce the observed downcore Pb isotope variations without requiring alteration of Pb isotope signatures during particle sinking.**

We agree with the reviewer, in our data we see a mixture of lithogenic, authigenic and biogenic Pb which seem to show different isotope signatures. We have streamlined the discussion on water phase alteration of Pb isotope signatures during particle sinking in the revised manuscript. However, this topic needs some explanation/assumption of why the different Pb phases show different isotope signatures with likely just a single Pb source. Unfortunately, most of the literature data is based on dissolved Pb data only and data on Pb isotope signatures in marine particles is not yet available. Accordingly, most of the discussion on Pb isotope signatures in marine particles in the literature as well as our study is based on assumptions only. Recent studies (Lanning et al., 2023, Olivelli et al., 2024, Griffiths et al., 2026) show strong indication that alteration of Pb isotope signatures in marine particles (in the Southern Ocean) occur, although no particle data has been provided.

A6: The authors should clarify what information can and cannot be obtained from bulk-sediment Pb isotope measurements. In particular, the manuscript should distinguish among e.g., **mixing of isotopically distinct sedimentary Pb components; redistribution or isotope exchange between dissolved and particulate Pb pools; selective dissolution and scavenging**. These processes are sometimes discussed interchangeably, although they have different implications. Because mass-dependent Pb isotope fractionation has not been demonstrated by the present data, terms such as "fractionation" and "alteration" should be used cautiously. In many places, "redistribution," "isotope exchange," "selective scavenging," or "mixing of distinct Pb pools" would be more appropriate.

We thank the reviewer for this comment. The now used normalization approach allows to keep the wording in the revised version more consistent, separating the processes more distinctly while discussing limitations of the individual concepts.

Why was biogenic silica not measured?
A7: The interpretation of Si as a productivity proxy is central to the manuscript, but the authors measured total Si in bulk sediment. Total Si includes both biogenic opal and lithogenic silicates or quartz. Indeed, the authors acknowledge that Si may be partly lithogenic.

Biogenic silica is highly soluble and undergoes extensive dissolution during its descent through the water column and within the upper sediment layers which also includes release of Pb bound in diatom frustules (see Chuang et al., 2014). Only about 3–4% of surface-produced bSi escapes dissolution and is permanently buried in sediments. Due to this, we have not extracted bSi but now followed a normalization approach using lithogenic and biogenic elements (see comments above).

A8: Why was biogenic silica or opal not measured directly? Without separating biogenic and lithogenic Si, correlations between Pb isotope ratios and total Si cannot be uniquely interpreted as evidence for a productivity control. Such correlations could also result from variations in lithogenic input, sediment mixing or dilution by biogenic material.

We agree with the reviewer that total Si cannot be uniquely interpreted as evidence for a productivity control. As described above we are now following a normalization approach using Ba concentrations, which have frequently been used in other studies before as a measure of paleo productivity.

A9: Ideally, the authors should provide direct opal measurements. If this is not possible, they should at least: estimate excess or biogenic Si using Al, Ti or another lithogenic indicator. Sedimentary Si and TOC should also not be described as direct measures of primary production because they are additionally affected by export efficiency, preservation, dissolution, dilution and sediment focusing.

We agree, see comments above. We used Al and Ti for lithogenic and Ba for biogenic Pb.

A10: Comparison with nearby authigenic Pb isotope records
I suggest the authors to compare their results with the nearby Southern Ocean sediment record of Huang et al. (2021): Huang et al. (2021) extracted authigenic Pb isotope signatures, whereas the present study measured bulk sediment. Importantly, both studies appear to indicate only a limited contribution from anthropogenic Pb. The consistency between the two records should be discussed because it may strengthen the authors' conclusion that anthropogenic contamination is minor in this sector of the Southern Ocean.

We have cited the above-mentioned reference. We agree that both studies indicate a minor influence of anthropogenic Pb and discussed these findings in the revised version. As mentioned above, we are not sure how stable Fe-Mn bound Pb (authigenic Pb) in these sediments is, but a comparison of the data was included nonetheless as it is valuable to compare the Pb isotope signal of such a close region. Fe-Mn rich basaltic tephra, with a different Pb isotope signature, (see discussion on St15 in revised version) as well as the Fe-Mn redox mobility in our sediment, however, make it difficult to draw unambiguous conclusions from this comparison.

A11: At the same time, the different analytical phases provide an opportunity to evaluate the origin and carrier of Pb. The authors should compare the isotope ranges of the bulk and authigenic records and discuss whether their similarity or difference can constrain the relative contributions of: **lithogenic Pb derived from Patagonian dust; authigenic Pb derived from seawater; and Pb scavenged or transported by biogenic particles**.

See answer above. We have compared and discussed the Pb isotopic ratios of Huang et al 2015 and our study in the revised version with ambiguous results.

A12: One of the principal findings of the manuscript is the relationship between Pb isotope ratios and productivity-related parameters. A highly relevant comparison is provided by Huang et al. (2020). In that study, glacial–interglacial Pb isotope variations show a close relationship with opal flux, whereas they do not follow the authigenic seawater Nd isotope record, which reflects changes in water-mass provenance. This comparison is highly relevant to the present manuscript because it supports the possibility that sedimentary Pb isotope variability is related to biological particle export rather than being controlled solely by changes in water masses.

We agree with the reviewer and have discussed this important accordance in the revised version.

Additional comments
Lines 92–93: Revise "as assumed by (Cheng and Hu, 2010)" to "as assumed by Cheng and Hu (2010)."

Has been changed

Lines 255–290: The three sites do not constitute a controlled water-depth transect because they also differ in geographic location, productivity, sedimentation rate, lithogenic input and oceanographic setting. Differences among the cores should therefore not be attributed primarily to water depth or pressure.

We agree with the reviewer and have addressed this point in the revised version. An overview table of all differing parameters is offered in the SI for further clarification.

Lines 269–273: Higher Pb and Al concentrations at St15 may reflect stronger lithogenic input or weaker dilution by biogenic material, rather than enrichment of Pb during particle sinking. Please consider these alternatives.

This was considered in the revised version and connected to a distinct tephra-layer present in the sediments of St15.

Lines 273–278: The proposed ~1 cm offset between Si and Pb/TOC is unlikely to be explained simply by different particle sinking velocities. An offset in sediment depth represents a much longer timescale than typical water-column sinking differences. Sediment mixing, chronological uncertainty, sampling resolution and post-depositional processes should be considered.

We could actually not resolve this observation in the revised version. The suggested complications through sediment mixing, chronological uncertainty, sampling resolution and post-depositional processes would affect most other elements in a similar manner, which we could not observe. However, the overall effect of this offset on the data interpretation is only small.

Lines 370–375 and 390–394: The proposed pressure-induced dissolution of Pb-bearing mineral phases and biogenic silica requires stronger evidence. Otherwise, it should be clearly identified as a hypothesis.

We clearly identified this statement as a hypothesis, although the dissolution of opal in the deep ocean is quite well known (undersaturation of deep water etc.). We have clarified this point in the revised version and supported it with relevant citations.

Lines 402–407: The assumption that nearly all mineral-associated Pb was dissolved during sinking is not demonstrated by the available data and should be removed or substantially qualified.

We agree with the reviewer. This assumption has been removed.

The correlation analyses are based on a relatively small number of ordered downcore samples. Because common depth trends and autocorrelation may inflate statistical significance, the authors should test whether the correlations remain after detrending or using first differences.

In order to determine correlation coefficients and p-values, due to the small sample size ($n = 17$ per station) and the presence of non-normally distributed variables (Shapiro-Wilk test, $p < 0.05$), Spearman rank correlation was used. Additionally, a PCA was performed after detrending to verify the obtained results. The use of ratios (Al, Ti, Ba) provides another independent trend analysis.

Lines 489–495: The seawater data used for comparison were obtained substantially farther north and may not represent seawater directly overlying the sediment sites. This limitation should be acknowledged.

We have acknowledged this point and included the data published by Huang et al, 2020. However, the data published by Olivelli et al. 2024 does not show that much variation in dissolved Pb isotope signatures throughout the N-S transect which is why we believe that water phase Pb isotope signatures are similar in our sampling areas, although influence of particles attributed to high productivity has been reported here. The data published by Huang et al. (2020) is more difficult to compare as it shows only the authigenic Pb extracted from Fe-Mn oxyhydrates in the sediment. Although it is assumed that the authigenic Pb fraction reflects the average Pb isotope signature of the entire water column, this assumption is poorly validated yet as it disregards the biogenic Pb component in the sediment, which might be important especially in productive areas. In general, Pb binding to particulates in the oceans' water phase is still poorly understood and partly discussed controversially in the literature (see e.g. Chuang et al, 2014 and 2015). Recent studies on water column Pb isotope signatures show strong indications that high particle fluxes in productive areas have significant influence on water phase Pb isotope signatures as well as Pb export to the sediments which seem to be mainly related to biogenic particles rather than to Fe-Mn hydroxides. We have discussed this point in the revised version of the manuscript (Chen et al., 2026, Lanning et al., 2023, Olivelli et al., 2024, Griffiths et al., 2026).

Line 492: The reported sedimentary 206Pb207Pb range of 1.12–1.21 appears inconsistent with the range of 1.195–1.212 reported earlier. Please check this value.

Has been changed in the revised version

The conclusion should more consistently use wording such as “consistent with” or “suggests,” rather than presenting selective scavenging and water-column modification as uniquely demonstrated mechanisms.

Will be adapted in the revised version

Please check the use of “absorb” and “adsorb” throughout the manuscript. For surface binding to particles, “adsorb” is generally appropriate.

Has been corrected in the revised version.

Reference:

Huang, H., Gutjahr, M., Eisenhauer, A., and Kuhn, G. (2020). No detectable Weddell Sea Antarctic Bottom Water export during the Last and Penultimate Glacial Maximum. Nature Communications, 11, 1–10.

Huang, H., Gutjahr, M., Kuhn, G., Hathorne, E. C., and Eisenhauer, A. (2021). Efficient extraction of past seawater Pb and Nd isotope signatures from Southern Ocean sediments. Geochemistry, Geophysics, Geosystems, 22, e2020GC009287.

References have been included in the revised version.