



**Science Editor**  
**Prof. Shiling Yang**  
*Climate of the Past*

Topic: EGUSPHERE-2025-2619 Response to reviewers

Geneva, 12 January 2026

Dear Prof. Yang,

Thank you for handling our manuscript and for the opportunity to revise it. We have carefully addressed all comments from both reviewers and revised the manuscript accordingly. Below we summarise the main changes.

1. We revised and clarified the conceptual framework used to interpret detrital  $\delta^7\text{Li}$  records, correcting statements related to high  $W/D$  regimes and refining the discussion of grain-size effects, hydrodynamic sorting, and potential dilution by primary silicates.
2. We restructured the Discussion to compare the Southern Pyrenees  $\delta^7\text{Li}$  records directly with previously published PETM weathering datasets. This revision highlights the importance of the regional geomorphic and hydroclimatic context.
3. We strengthened the link between our geochemical interpretation and independent sedimentological evidence for increased physical erosion and sediment flux during the PETM in the Southern Pyrenees (e.g. coarse-grained depositional pulses and enhanced channel mobility). These references have been integrated into the main text.
4. To address questions regarding mineralogical influences on  $\delta^7\text{Li}$ , we added new Supplementary Figures (Figs. S1 and S2) showing cross-plots of  $\delta^7\text{Li}$  versus clay mineral abundances. These plots indicate no systematic correlation within analytical uncertainty for Rin, and highlight recovery-phase clustering at Esplugafreda, consistent with changes in clay detrital input.
5. We corrected and quantified the expected temperature effect on Li isotope fractionation using a conservative  $\sim 3$  °C PETM warming estimate for the region, showing that temperature-driven changes are too small to explain the observed  $\sim 1\%$  excursion. We also clarified the role of runoff and floodplain residence times in shaping the preserved clay  $\delta^7\text{Li}$  signal, avoiding any implication of a simple direct runoff– $\delta^7\text{Li}$  relationship.
6. We updated figures for clarity and consistency, including a new synthesis figure summarising the isotopic trends (as proposed by Reviewer 2) and correcting/clarifying Figure 1 legend and colour information (as requested by Reviewer 1).

We hope these revisions fully address the reviewers' concerns and improve the clarity and robustness of the manuscript.

Please find below the point-by-point response to the reviewers' comments and queries.

Color coded:

Reviewer comment / **Original text** / **Response and modification**

On behalf of all co-authors,

Sincerely,

Rocío Jaimes Gutiérrez (she, her)  
Postdoctoral researcher  
Earth Surface Dynamics Research Group  
Department of Earth Sciences  
University of Geneva  
Rue des Maraîchers 13  
1205 Geneva, Switzerland

RC1: ['Comment on egusphere-2025-2619'](#), Gaojun Li, 18 Jul 2025 [reply](#)

In this manuscript, the authors analyze the lithium and neodymium isotopes of two sections from the southern Pyrenees, deposited during the PETM event (~ 56 Ma ago). They report a positive excursion of ~0.8‰ in the clay-size fraction lithium isotopic ratios during the PETM in both sections, while the neodymium isotope data indicate stable sediment provenance. They suggest the positive excursions in lithium isotopes reflect a decline in weathering intensity and congruency, potentially linked to the enhanced formation of secondary clays during continental weathering. To better understand how Earth's climate regulates itself, it is essential to unravel the linkages among denudation, weathering and climate. I believe this paper improves our understanding of chemical weathering at hyperthermal events. The language of this manuscript is well-revised and clear enough, and the data broadly support the authors' interpretations. However, several sections of the manuscript raise important questions or require clarification, as outlined below. Overall, I recommend to accept this manuscript after minor revisions.

We sincerely thank Reviewer 1, Gaojun Li, for his constructive feedback, support of the manuscript, and thoughtful review suggestions. We have implemented his suggestions, which have certainly improved the quality and clarity of the paper. Below, we address each of the points raised.

First of all, the authors propose **increased denudation and weathering flux during the PETM**, while also suggesting that **weathering intensity (W/D) declined** (e.g. Lines 525-533). I am wondering are there any direct geological or sedimentological evidences for increased erosion or tectonic uplift in the southern Pyrenees during the PETM? Because in the absence of uplift, one would expect weathering intensity to rise with increasing temperature and precipitation.

**Original, lines 525-533:** “The Southern Pyrenees during the PETM was a relatively high-erosion regime, such that physical erosion dominated, increasing the sediment supply and exposing fresh minerals. When erosion exposes fresh minerals, weathering rates increase with total denudation, albeit less strongly than the increases in erosion, consistent with shared controls on chemical weathering and physical denudation rates (Riebe et al., 2004). Even though high-relief regions produce weakly weathered sediments, their high sediment yields and moderate clay formation rates result in elevated weathering fluxes (Gaillardet et al., 1999). Therefore, we propose that the Southern Pyrenean floodplains record a shift from a high-weathering intensity regime to a moderate-weathering intensity regime during the PETM (Fig. 7).”

**Response:** We agree that independent evidence for enhanced denudation during the PETM is essential. In the Southern Pyrenean foreland basin, several sedimentological records attest to rapid erosion and enhanced sediment supply during this interval. For example, the deposition of the massive conglomeratic megafan unit (Claret Conglomerate; Schmitz & Pujalte, 2003, 2007; Pujalte et al., 2015, 2016) reflects increased sedimentary fluxes that seem to be attributed to extreme precipitation events (Prieur et al., 2025). Additional indicators include enhanced channel mobility (Chen et al., 2018) and an increase in *Microcodium* abundance and export to the oceans (Prieur et al., 2024). Together, these records point to a marked intensification of erosion and sediment transfer, consistent with our interpretation of increased weathering fluxes. We integrated these references explicitly in the revised text to make the connection between the isotopic evidence and sedimentological records clearer.

We have modified the main text to reflect these references.

**Modified lines 553 to 569:** The Southern Pyrenees during the PETM was a relatively high-erosion regime, such that physical erosion dominated, increasing the sediment supply and exposing fresh minerals (Schmitz and Pujalte, 2007; Pujalte et al., 2015, 2016; Chen et al., 2018; Prieur et al., 2024, 2025). When erosion exposes fresh minerals, weathering rates increase with total denudation, albeit less strongly than the increases in erosion, consistent with shared controls on chemical weathering and physical denudation rates (Riebe et al., 2004; West et al., 2005). Even though high-relief regions produce weakly weathered sediments, their high sediment yields and moderate clay formation rates result in elevated weathering fluxes (Gaillardet et al., 1999). Therefore, we propose that the Southern Pyrenean floodplains record a shift from a high-weathering intensity regime to a moderate-weathering intensity regime during the PETM (**Fig. 7**). The pre-PETM conditions were characterised by a low reactivity of the parent lithology (Kump and Arthur, 1997; Caves Rügenstein et al., 2019), associated with the carbonate-rich, reworked sediments in the floodplain deposits, and hence low total weathering fluxes. The above scenario is also consistent with the "system-clearing" event documented in western North America (Foreman et al., 2012), where sediment transport surged in response to rapid climatic forcing, as well as with other Eocene warming events such as the Mid-Eocene Climatic Optimum, which saw a shift towards enhanced clay formation and a lower weathering intensity (Krause et al., 2023).

Second, the manuscript suggests that increased kaolinite abundance reflects enhanced secondary clay formation (Lines 547–549). However, kaolinite abundance alone may not reliably indicate the total formation flux of secondary clays, as it does not account for other clay mineral phases. I think it's not easy to determine the total formation flux of secondary clays, but one can consider an extreme case: the observed kaolinite enrichment could also result from the dissolution of other clays.

**Original:** Hence, its enrichment suggests either intensified in-situ clay formation in the floodplains or increased transport of weathered material from the uplands to the lowlands.

**Response:** We acknowledge the reviewer's point that kaolinite abundance alone cannot serve as a straightforward proxy for enhanced secondary clay formation. Kaolinite enrichment could, in principle, result from processes such as the erosion and redeposition of pre-PETM clays. While we discuss this limitation later in the manuscript, we emphasize that with the currently available mineralogical data, it is not possible to unequivocally distinguish between neoformed clays and reworked/pre-PETM clays. Some Scanning Electron Microscopy images is available from the Esplugafreda section (Jaimes-Gutierrez et al., 2024, Supplemental Material), showing preservation of rose-shaped smectite and bookshelf structures in kaolinite, which have been proposed as evidence for pedogenesis.

Additionally, the positive  $\delta^7\text{Li}$  excursion observed in our dataset suggests an overall increase in clay formation during the PETM. In this context, the kaolinite data remain consistent with such an interpretation. Importantly, global mass-balance calculations indicate that the magnitude of the seawater  $\delta^7\text{Li}$  excursion during the PETM cannot be explained solely by erosion or recycling of previously formed clays; rather, it requires enhanced formation of new secondary clays (Pogge von Strandmann et al., 2021). This scenario supports our interpretation that the observed kaolinite increase reflects intensified continental weathering and clay neoformation, even though kaolinite alone does not capture the full spectrum of clay mineral phases involved.

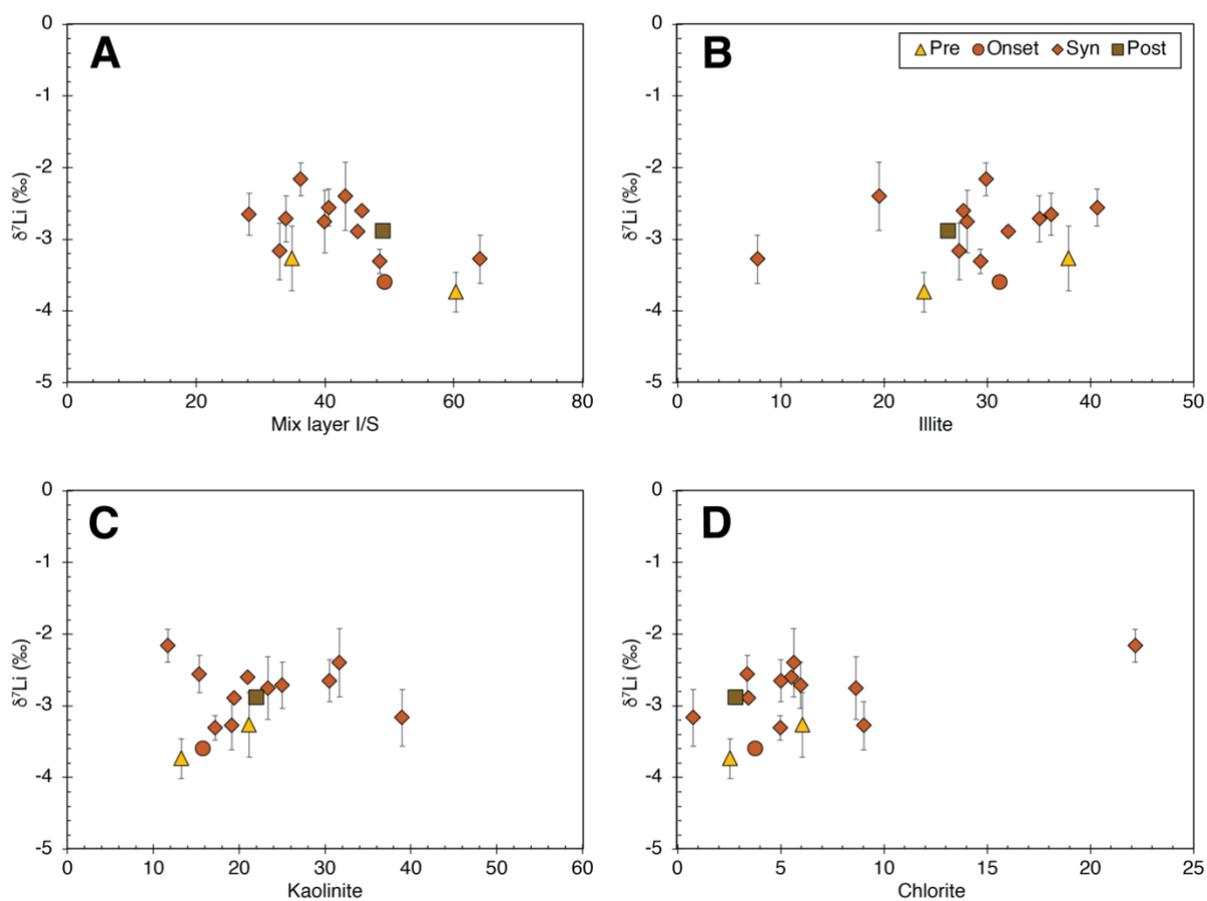
Third, several studies (e.g., Pistiner & Henderson, 2003; Golla et al., 2021) suggest that lithium isotope fractionation is mineral-dependent. Since the authors already present clay mineralogy data, it would strengthen the paper to discuss how variations in mineral assemblages might influence the lithium isotopic fractionation factor ( $\Delta^{7}\text{Li}_{\text{water-clay}}$ ) and, consequently, the observed  $\delta^{7}\text{Li}$  values.

**Response:** We thank the reviewer for this important observation. Indeed, Li isotope fractionation can be mineral-dependent, which may affect the interpretation of  $\delta^{7}\text{Li}$  variations. This idea is supported by both Pistiner & Henderson (2003) and Golla et al. (2021). However, neither paper derives mineral-specific equilibrium fractionation factors for different clay minerals. In Pistiner & Henderson (2003), the mineral-dependent behavior arises from sorption experiments, while in Golla et al. (2021) the clay-specific  $\alpha$  values used for kaolinite and illite are imposed as kinetic model parameters based on conditions outside our system (e.g., ambient batch experiments or high-temperature illitization extrapolations).

To our knowledge, experimentally validated, mineral-specific equilibrium fractionation factors for low-temperature weathering conditions are not yet available. Additionally, Experimental studies indicate that lithium isotope fractionation during secondary mineral formation is largely independent of clay mineralogy, with Li uptake into different secondary phases producing a broadly constant bulk fractionation factor (Hindshaw et al., 2019; Pogge Von Strandmann et al., 2022).

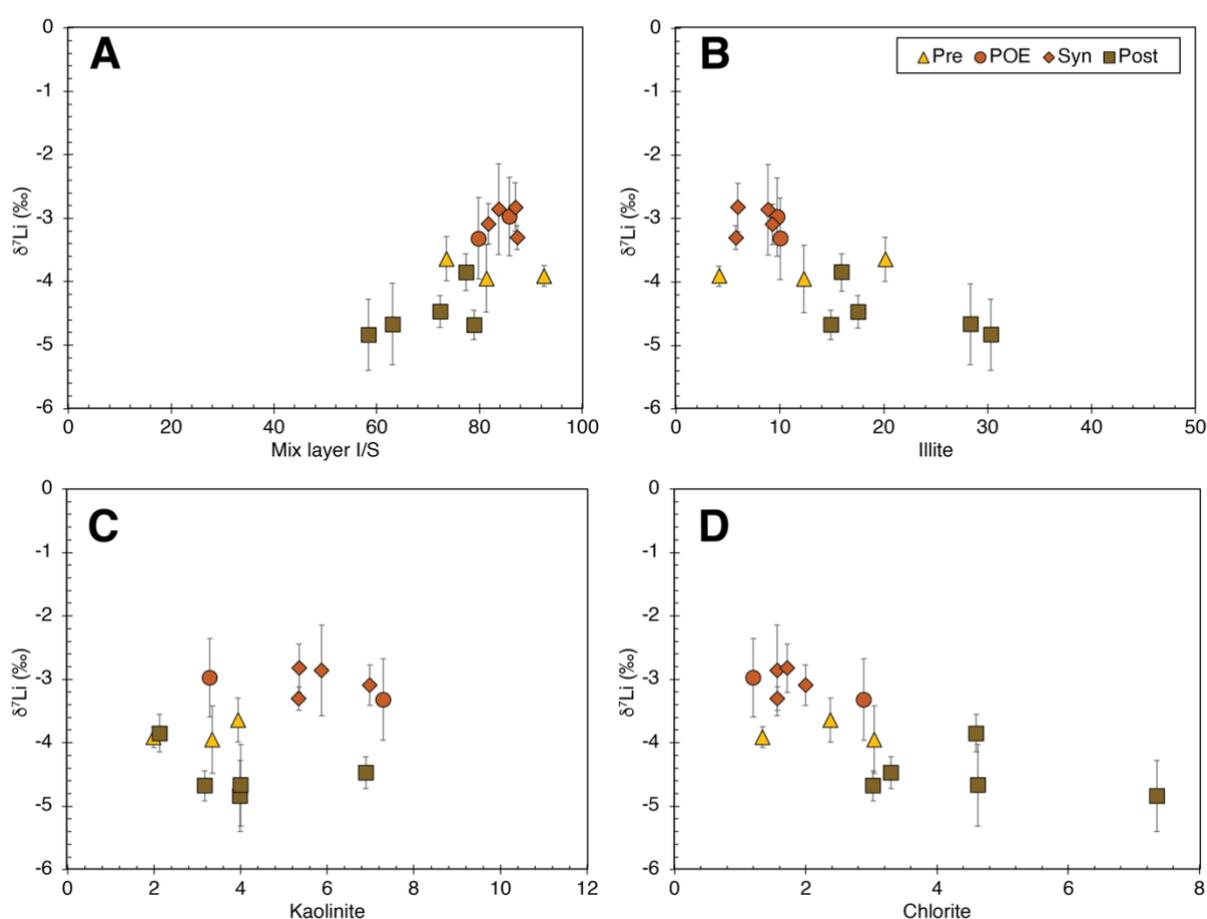
A possible approach to address this limitation is to examine whether mineralogy exerts any discernible control on our measured  $\delta^{7}\text{Li}$  values. To do so, we have generated cross-plots of  $\delta^{7}\text{Li}$  values versus kaolinite, mixed-layer illite–smectite, illite, and chlorite abundance. These plots show no statistically significant relationships (Fig. S1, Rin), suggesting that mineralogical controls on Li isotope fractionation—if present—are either minor or below the resolution of our dataset. Fig. S2, Esplugafreda does show some clustering, potentially relating to and increase in detritism during the Recovery phase (Jaimes-Gutierrez et al., 2024).

# Rin



**Figure S1.** Cross plots of lithium isotope composition ( $\delta^7\text{Li}$ ) in clays versus clay mineral abundances in the Rin section. (A) Mixed-layer illite–smectite (I/S), (B) illite, (C) kaolinite, and (D) chlorite. Clay mineral abundances are reported in wt%. No systematic correlation is observed between  $\delta^7\text{Li}$  and the relative abundance of individual clay minerals within analytical uncertainty.

## Esplugafreda



**Figure S2.** Cross plots of lithium isotope composition ( $\delta^7\text{Li}$ ) in clays versus clay mineral abundances in the Esplugafreda section. (A) Mixed-layer illite–smectite (I/S), (B) illite, (C) kaolinite, and (D) chlorite. Clay mineral abundances are reported in wt%. Samples from the pre-PETM, Pre-Onset Excursion (POE), and syn-PETM intervals cluster at high mixed-layer I/S abundances ( $\sim 80$  wt%) and display relatively heavier  $\delta^7\text{Li}_{\text{clays}}$  values. In contrast, recovery-phase samples form a distinct cluster characterised by lighter  $\delta^7\text{Li}_{\text{clays}}$ , coincident with increased abundances of illite, kaolinite, and chlorite.

### Modified lines 394 to 410:

The clays of the Rin section have a mean lithium isotope composition of  $-2.9 \pm 0.5\text{‰}$  ( $1\sigma$ ) (Fig. 5B, Table S2). Between 5.8 and 18.2 m, the mean composition is  $-3.4 \pm 0.2\text{‰}$ . Above this, from 20.3 to 39.5 m, the mean composition is  $-2.6 \pm 0.2\text{‰}$ , which corresponds to a shift towards more positive values of  $\sim 0.8\text{‰}$ . The minimum value of  $-3.7\text{‰}$  is seen before the Claret Conglomerate, and the maximum value of  $-2.2\text{‰}$  occurs immediately after the Claret Conglomerate, indicating a total range of up to  $\sim 1.5\text{‰}$ . The  $d^7\text{Li}$  values measured on carbonate nodules have a maximum value of  $8.6\text{‰}$ , a minimum value of  $-3.0\text{‰}$ , and a mean composition of  $0.9 \pm 4.0\text{‰}$  (Fig. 5C). **No clear temporal trend is observed in the  $d^7\text{Li}_{\text{nodules}}$  record. No systematic correlation is observed between  $\delta^7\text{Li}_{\text{clays}}$  and the relative abundance of individual clay minerals within analytical uncertainty (Fig. S1).**

At the Esplugafreda section, the clays have a mean lithium isotope composition of  $-3.7 \pm 0.7\text{‰}$  (Fig. 6B, Table S3). The pre-PETM samples (0-10 m and 21-28 m, Jaimes-Gutierrez et al.,

2024 and references therein) have a mean composition of  $-3.8 \pm 0.2\%$ . The POE samples (~15–21 m) have a composition of  $-3.2 \pm 0.2\%$ , and the syn-PETM sediments have a composition of  $-3.0 \pm 0.2\%$ , indicating a PETM shift towards more positive values of  $\sim 0.8\%$ . The post-PETM sediments have a composition of  $-4.5 \pm 0.4\%$ . **Samples from the pre-PETM, Pre-Onset Excursion (POE), and syn-PETM intervals display relatively heavier  $\delta^7\text{Li}_{\text{clays}}$  values, whereas recovery-phase samples form a distinct cluster characterised by lighter  $\delta^7\text{Li}_{\text{clays}}$  (Fig. S2).**

Finally, the manuscript assumes that  $\delta^7\text{Li}$  variations in the clay-size fraction reflect continental weathering processes exclusively, with negligible contribution from marine authigenic aluminosilicate clays. However, an increased fraction of marine authigenic aluminosilicate clays during the PETM could also elevate the clay  $\delta^7\text{Li}$  values. Could the authors provide some additional constraints to rule out this possibility?

### **Response:**

The reviewer raises an important question regarding the potential contribution of marine authigenic aluminosilicate clays. Based on the coastal but dominantly continental depositional setting of the Rin section, the formation of authigenic clays is possible but unlikely to have been significant. The Esplugafreda section is fully continental during the pre, syn and recovery of the PETM. In marine sediments, clay mineral assemblages dominated by kaolinite, illite, smectite, and mixed-layer illite/smectite are generally interpreted as mainly detrital in origin, reflecting continental weathering and fluvial transport rather than in situ marine precipitation (Thiry, 2000; Fagel, 2007; Velde and Meunier, 2008).

Authigenic formation of these clay minerals in marine environments is known to occur, but is typically restricted to specific conditions such as alteration of volcanic ash, evaporitic or hydrothermal settings, and is therefore not expected to be significant in shallow, near-shore depositional systems with high terrigenous sedimentation rates (Środoń, 2001; Wise et al., 2001; Meunier, 2005). In contrast to glauconite or other green clays, which may form authigenically under low sedimentation and long residence times, kaolinite and illite in coastal marine settings are widely regarded as inherited from continental sources (Thiry, 2000; Meunier, 2005; Fagel, 2007). Given the dominantly continental depositional setting of the Rin section and the large detrital clay input inferred for the near-shore environment, any marine authigenic clay contribution is therefore expected to have been minor relative to the terrigenous signal.

Below are some minor issues:

In Lines 111-112, the term “weathering efficiency” is ambiguous. I guess this refers to the  $\text{CO}_2$  consumption flux as suggested by Bufe et al., 2024. It would be better if the authors can make this clearer.

**Original:** However, weathering efficiency peaks at intermediate erosion rates, while extremely thin or thick soils reduce weathering fluxes (Gabet and Mudd, 2009; Dixon and Von Blanckenburg, 2012; Bufe et al., 2024).

**Response:** We have deleted this term as it could be unclear and it was not central to the argument.

In Lines 115-116, How can weathering rates increase with erosion under kinetically-limited regime? I suppose this is only the case for supply-limited regimes.

**Original:** In kinetically-limited regimes, which are typical of high-relief areas with thin soils, weathering rates are controlled by mineral dissolution kinetics and increase with erosion (Kump et al., 2000; Riebe et al., 2004; West et al., 2005).

**Response:** Thank you for highlighting this mistake. We have deleted the second part of the statement, as dissolution kinetics should control the weathering rates.

In Lines 493-495, the authors suggest enhanced clay formation in lowlands during the PETM. Is there evidence to support this? Could increased runoff instead dilute solute concentrations, thereby suppressing clay precipitation? Also, is it realistic to assume that enhanced clay formation in lowlands could compensate for reduced formation in uplands?

**Original:** These processes would lead to decreased clay formation in the uplands (Kump et al., 2000; Riebe et al., 2004), but increased clay production and accumulation in lowlands, where longer sediment residence times promote authigenic clay formation.

**Response:** We thank the reviewer for this question, which helps clarify our interpretation.

While increased runoff can indeed dilute solute concentrations during peak discharge, particularly in river waters, seasonal and floodplain settings can experience longer sediment and pore-water residence times following flood events. In such environments, continued alteration, reworking, and preservation of fine-grained material may occur without requiring sustained high solute concentrations or basin-wide increases in clay production. We intended to highlight a redistribution of weathering and clay-processing processes towards lowland environments under PETM hydroclimatic conditions. We have revised the text to clarify these processes.

**Modified lines 510 to 516:** These processes would lead to decreased clay formation in the uplands by shortening water-rock interaction times (Kump et al., 2000; Riebe et al., 2004), while shifting clay production and accumulation towards lowland environments, where longer sediment residence times promote authigenic clay formation, consistent with modern river floodplain processes (e.g. Dellinger et al., 2015; Maffre et al., 2020). We also note that weathering processes are highly heterogeneous, and therefore global variability during the PETM can be expected (e.g. Frings, 2019). Critically, A decrease in  $W/D$  can lead both to positive or negative  $\delta^7\text{Li}$  excursions, depending on the starting weathering regime (Krause et al., 2023).

In Lines 517-522, I might be wrong, but higher temperatures during the PETM would likely reduce the lithium isotope fractionation (i.e. decrease  $\Delta^7\text{Li}_{\text{water-clay}}$ ) during clay formation. The secondary clay formed at this warm period should have a closer  $\delta^7\text{Li}$  to the starting solution (or river water) ( $\delta^7\text{Li}_{\text{secondary-clay}} = \delta^7\text{Li}_{\text{water}} - \Delta^7\text{Li}_{\text{water-clay}}$ ). Assuming a constant  $\delta^7\text{Li}$  value of river water ( $\delta^7\text{Li}_{\text{water}}$ ) during the period of interest, the clay formed at syn-PETM should have higher  $\delta^7\text{Li}$  values, rather than lower values.

**Original:** Considering the mean annual air temperatures for the continental Pyrenees were a minimum of 23 °C pre-PETM and a maximum of 28 °C syn-PETM (Jaimés-Gutiérrez et al., 2024), the resulting maximum fractionation from pre-PETM to syn-PETM was -0.70 ‰ (Li and West, 2014). Although this fractionation towards more negative values may have had a

small effect in reducing the magnitude of the excursion, it is evidently unable to account for the positive  $\delta^7\text{Li}_{\text{clays}}$  shifts observed in the records. Hence, climatic and hydrological processes, rather than direct temperature effects, must dominate the  $\delta^7\text{Li}$  signal.

**Response:** We thank the reviewer for raising this point. We have revised the manuscript to explicitly consider the magnitude and direction of the temperature effect using a  $\sim 3$  °C warming across the PETM in the Southern Pyrenees (Jaimes-Gutierrez et al., 2024). Based on experimental and theoretical constraints (Vigier et al., 2008; Li and West, 2014), this temperature increase would result in only a small (few tenths of a per mil) shift toward heavier  $\delta^7\text{Li}$  values in clays, which is insufficient to explain the observed  $\sim 1\%$  positive excursion in  $\delta^7\text{Li}_{\text{clays}}$ .

**Modified 543 to 551:** Temperature effects on lithium isotope fractionation are minor in the Southern Pyrenees. While lithium isotope fractionation during clay formation is temperature-dependent (Vigier et al., 2008; Li and West, 2014), the fractionation factor ( $\alpha$ ) for incorporation in smectite is nearly constant across typical surface weathering temperatures (Vigier et al., 2008). Using the  $\sim 3$  °C warming estimated for the continental Pyrenees across the PETM (Jaimes-Gutierrez et al., 2024), the maximum temperature-driven change in clay  $\delta^7\text{Li}$  values is expected to be only a few tenths of a per mil (Li and West, 2014). This effect would shift clay  $\delta^7\text{Li}$  values slightly towards heavier values and could therefore contribute marginally to the observed excursion, but it is insufficient to explain the full magnitude of the  $\sim 1\%$  positive  $\delta^7\text{Li}_{\text{clays}}$  shift observed in the records. Hence, climatic and hydrological processes, rather than direct temperature effects, must dominate the  $\delta^7\text{Li}$  signal.

In Figure 1C, there are three different blue areas (from light to dark), but only two of them are explained in the legend. Additionally, the meaning of the white-striped area in panel B is unclear and should be clarified.

The figure has been modified.

We thank Reviewer 1 again for the detailed and constructive feedback. We have incorporate the suggested clarifications and additional discussions into the revised manuscript, which we are confident has improve its clarity and robustness.

**RC2:** '[Comment on egusphere-2025-2619](#)', Anonymous Referee #2, 16 Aug 2025 [reply](#)

General comments:

This manuscript reports new geochemical ( $\delta^7\text{Li}$ ,  $\epsilon\text{Nd}$ ,  $\delta^{13}\text{C}$ ) and mineralogical data on two well-preserved Palaeocene-Eocene sedimentary floodplain sections in the Pyrenees. The objective is to reconstruct the regional changes in chemical weathering and physical erosion during the PETM warming event and the aftermath to characterize the response of the Earth system to warming events. The authors found a positive  $\delta^7\text{Li}$  excursion in the clay fraction of both sections during the main body of the PETM. They interpret these results as reflecting a decrease of the weathering intensity (higher chemical erosion and even higher physical erosion) during the PETM because of the intensification of the hydrological cycle and a more extensive clay formation in the floodplains.

Overall, this is a neat study and a valuable scientific contribution to the research field on the PETM. It is well written, the data are of high quality, and the interpretation and arguments are convincing. There are only a few aspects of the manuscript that should be improved, in particular the presentation of the interpretative framework and the implications, before final acceptance.

We thank Reviewer 2 for the constructive and positive evaluation of our manuscript, and for the detailed comments and suggestions. We are pleased that the reviewer finds the study valuable and the dataset of high quality. Below, we provide a point-by-point response to each major comment. We have revised the manuscript accordingly.

My recommendation is publication after minor revisions. The main revisions I suggest are:

- Clarify the interpretative framework of Li isotopes in detrital sediments (section 1.2). In my opinion, some statements in this section are not in accordance with results from published studies (see more details below).

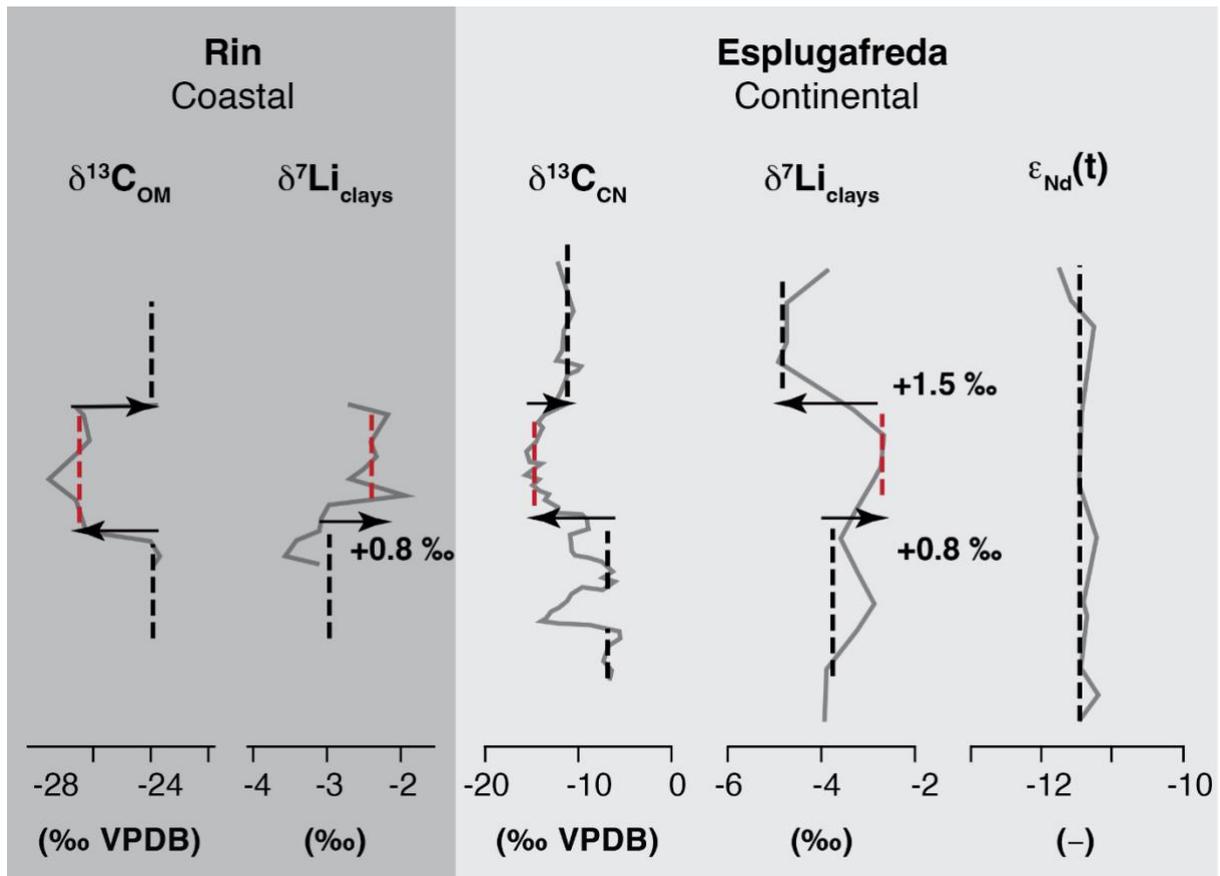
Applied (see below)

- Discuss in more details the comparison and implications with other Li isotopes PETM records. I suggest moving the paragraph lines 464-473 after discussing the results from the Pyrenees and modifying it to compare directly the results from various records and places. Why a positive excursion in the Pyrenees and a negative excursion in other settings? Is it because of the different geomorphic environment (deep regolith and transport-limited weathering in the pyrenees)? How does it compare with the other studied floodplain section, in the Bigorn basin (Ramos et al., 2022)?

We have modified the text accordingly.

**Modified lines 475 to 516:**

Our  $\delta^7\text{Li}_{\text{clays}}$  records from Rin and Esplugafreda both show a positive (~1‰) lithium isotope excursion in the continental Southern Pyrenees during the onset and body of the PETM (**Fig. 7**). The  $\delta^7\text{Li}$  values from carbonate nodules at Rin show greater variability (**Fig. 5C**), but remain inconclusive due to the potential clay contamination or cation exchange between clays and carbonates (e.g., Pogge von Strandmann et al., 2019). Given the high Li content in silicate minerals, even a minor clay particle content in the nodules could contaminate the signature. Critically, the invariant  $\epsilon_{\text{Nd}}$  composition of the samples in both size fractions throughout the Esplugafreda record supports a constant provenance of the sediments, which suggests that the  $\delta^7\text{Li}_{\text{clays}}$  records can be reliably interpreted as a reflection of weathering regime changes in response to the climatic perturbation.



**Figure 3.** Isotopic results from the Esplugafreda and Rin sections, with  $\delta^7\text{Li}_{\text{clays}}$  excursion from syn to PETM.

In many other PETM records, both marine and terrestrial  $\delta^7\text{Li}$  values show a negative excursion from pre-PETM to syn-PETM conditions. Pogge von Strandmann et al. (2021) documented a negative  $\delta^7\text{Li}$  excursion of  $\sim 3\text{‰}$  during the PETM in several marine carbonate sections and in detrital shales, indicating intensified global erosion rates (by 2–3 $\times$ ) and a 50–60% increase in silicate weathering fluxes, which was proposed to have contributed to climate stabilisation. Ramos et al. (2022) also found a negative  $\delta^7\text{Li}_{\text{clays}}$  excursion, albeit with a smaller magnitude of  $\sim 1.5\text{‰}$ , in fine sediments of the Bighorn Basin, North America during the PETM, and this change was sustained during the recovery phase. Consistent with these results, Chen et al. (2023) found a negative  $\delta^7\text{Li}_{\text{clays}}$  excursion of  $\sim 3\text{‰}$  in the Nanyang Basin, which, together with the negative  $\delta^7\text{Li}$  excursion in carbonates, was interpreted as recording a doubling of the regional silicate weathering intensity during the PETM.

Why, then, do the Southern Pyrenees floodplains record a positive  $\delta^7\text{Li}_{\text{clays}}$  excursion during the PETM? We interpret the positive  $\delta^7\text{Li}_{\text{clays}}$  excursion during the PETM as a shift towards increased incongruent weathering (moderate weathering regime), characterised by enhanced clay formation in the floodplain deposits. This regime would be characterised by increased chemical weathering, but relatively greater increases in physical erosion and sediment transport (e.g. Pujalte et al., 2015; Chen et al., 2018; Prieur et al., 2024), due to enhanced runoff causing short water residence times and rapid sediment export (i.e. lower  $W/D$ ; Fig. 8). Hydrological changes during the PETM are widely documented in the Southern Pyrenees (Schmitz and Pujalte, 2007; Pujalte et al., 2015; Chen et al., 2018; Rush et al., 2021; Prieur et al., 2024, 2025;

Jaimes-Gutierrez et al., 2024) and likely played a central role in driving those weathering changes. A shift towards more intense, episodic rainfall, without an increase in mean annual precipitation (Rush et al., 2021), could have reduced infiltration, increased runoff, and shortened water–mineral interaction times. These processes would lead to decreased clay formation in the uplands by shortening water-rock interaction times (Kump et al., 2000; Riebe et al., 2004), while shifting clay production and accumulation towards lowland environments, where longer sediment residence times promote authigenic clay formation, consistent with modern river floodplain processes (e.g. Dellinger et al., 2015; Maffre et al., 2020). We also note that weathering processes are highly heterogeneous, and therefore global variability during the PETM can be expected (e.g. Frings, 2019). Critically, A decrease in  $W/D$  can lead both to positive or negative  $\delta^7\text{Li}$  excursions, depending on the starting weathering regime (Krause et al., 2023).

- Discuss in more detail the reasons for the very low  $\delta^7\text{Li}$  values in the Esplugafreda section during the recovery period. This does not appear to be a simple return to initial conditions, but rather a further increase in weathering intensity (lower  $\delta^7\text{Li}$ ) associated with regolith deepening and soil shielding relative to pre-PETM conditions. If this is the case, what are the implications for  $\text{CO}_2$  removal through silicate weathering, given that such highly weathered settings are characterized by a weak weathering response and therefore act as inefficient sinks for atmospheric  $\text{CO}_2$  (Penman et al., 2020)? The timescale of the PETM recovery and the role of silicate weathering remain debated, and this study could provide useful new constraints.

**Response:** During the Recovery phase (in Esplugafreda), we observe a coeval increase in illite, kaolinite, and chlorite abundances in the Esplugafreda section. This period has also been interpreted as more arid, although this interpretation has been debated by Basilici et al. (2022), who suggested that the gypsum may be of secondary origin. Regardless, the clay mineral assemblage strongly suggests an increase in detrital input. For this reason, we consider that the observed excursion cannot be straightforwardly interpreted as a change in weathering intensity.

#### **Modified lines 537 to 541:**

In the Esplugafreda section, the Recovery phase sees a shift towards more negative values than the pre-PETM conditions. During this interval, we observe a coeval increase in illite, kaolinite, and chlorite abundances in the Esplugafreda section Jaimes-Gutierrez et al. (2024). This shift in the clay mineral assemblage strongly suggests an increase in detrital input. For this reason, we consider that the observed excursion cannot be straightforwardly interpreted as a change in weathering intensity, but rather reflect the clay mineral assemblage (Fig. S2).

- Add a figure with the average values ( $\delta^7\text{Li}$ ,  $\epsilon\text{Nd}$ ,  $\delta^{13}\text{C}$ , mineralogy) during each period (pre, POE, onset, body and recovery) for both sections (on the same figure). This would really help the reader to have a global overview of the main results and evolution through time.

Applied for isotopic proxies (new Fig. 7, see above).

In addition, the potential influence of marine clay formation (reverse weathering) on Li isotopes is not mentioned or discussed. Do you have any evidence to dismiss it for the Rin section? The study from Zhang et al., (2022) on the Ainsa paleo-delta shows that marine clay formation results in lowering of the  $\delta^7\text{Li}$  of sediments.

**Response:** We thank the reviewer for raising this important point, which connects to Reviewer 1's fourth comment on the potential role of marine authigenic clays. Based on the coastal but dominantly continental depositional setting of the Rin section, the formation of authigenic clays is possible but is unlikely to have been significant. Furthermore, even if authigenic clay formation occurred, its abundance would be minimal relative to the large detrital clay input in the near-shore environment.

Specific comments:

- Line 111: define « weathering efficiency ». Note that this hypothesis of maximum weathering rate at moderate erosion rate (the “speed limit”) proposed by some authors (Dixon and von Blanckenburg, 2012; Gabet and Mudd, 2009) has been challenged by others (e.g. Larsen et al., 2014; West, 2012).

**Original:** weathering efficiency peaks at intermediate erosion rates, while extremely thin or thick soils reduce weathering fluxes (Gabet and Mudd, 2009; Dixon and Von Blanckenburg, 2012; Bufer et al., 2024).

**Response:** This point has also been raised by Reviewer 1, and we have deleted the statement which was unclear and not essential for the argument.

- Lines 139-145: This does not appear to be consistent with findings reported in previous studies. At high  $W/D$  both clay formation and clay dissolution processes are taking place, either in different part of the weathering profile (top regolith vs deep regolith; e.g. Chapela Lara et al., 2022; Clergue et al., 2015) or different part of the catchment (e.g. see figure 10 in Dellinger et al., 2015 ; figure 3 in Henchiri et al., 2016; Fries et al., 2019). Therefore, at high  $W/D$ , rivers do transport modern neoformed clays from weathering profiles, and hence there is NO evidence for “minimal clay neoformation” (on the contrary).

**Original:** Finally, in supply-limited regimes with high  $W/D$ , such as rainforests, there is no remaining primary rock material to weather, so pre-formed clays are re-dissolved, which drives solution  $\delta^7\text{Li}$  to low values, but with a very low weathering flux (e.g., Dellinger et al., 2015). Clays take up their Li from solution with an approximately constant fractionation factor, so they are also expected to mimic this boomerang curve (Dellinger et al., 2017; Pogge von Strandmann et al., 2023; Ramos et al., 2024).

**Response:** We thank the reviewer for this detailed and constructive comment and for pointing out the relevant literature. We agree that at high  $W/D$  ratios, clay dissolution and clay neoformation can occur simultaneously, either at different depths within the weathering profile (e.g. top versus deep regolith) or in different parts of the catchment, as documented in previous studies. Our original wording implying “minimal clay neoformation” in supply-limited regimes was therefore overly restrictive. The key point is the relative importance of clay dissolution. We have revised the lithium isotope framework subsection to remove this statement and to better reflect the coexistence of clay formation and dissolution processes under high  $W/D$  conditions.

- Lines 150-154: what is the evidence for “fewer of the coarser-grained primary minerals carried in rivers are expected to be transported into offshore sites, the clay weathering signal may generally be recorded more clearly in marine sites”? I see several problems here:

First, the Dellinger et al., 2017 relationship has been observed for fine sediments (< 63 microns) not “coarse-grained primary minerals”.

Second, several studies on the Yangtze estuary (Yang et al., 2021, 2025) show the opposite sediment transport process to the one suggested is suggested by the authors, i.e. preferential transport of coarse-grained primary minerals offshore relative to clay minerals (“the offshore transport of SPM in the Changjiang Estuary may result in the preferential flocculation and deposition of clay minerals during the flooding season, while primary minerals or other fine-grained particles tend to be resuspended and carried seaward by currents” (Yang et al., 2021)).

Third, even if there are fewer primary minerals in the <2  $\mu\text{m}$  fraction relative to the <63  $\mu\text{m}$  fraction, primary clays and other minerals (feldspar, micas) are still present in the <2  $\mu\text{m}$  fraction (e.g. Liu et al., 2025)

I suggest revision of the presented interpretation framework (Fig. 2) in accordance with existing publications.

**Original:** Because fewer of the coarser-grained primary minerals carried in rivers are expected to be transported into offshore sites, the clay weathering signal may generally be recorded more clearly in marine sites. However, such biases resulting from mixing with primary silicate grains in bulk sediment samples can potentially be significantly reduced by analysing the clay size fraction (<2  $\mu\text{m}$ ).

**Response:** We thank the reviewer for this detailed and constructive comment and agree that the original wording was overly simplified. The reviewer is correct that the relationship discussed by Dellinger et al. (2017) applies to the silt-grained fraction (<63  $\mu\text{m}$ ), rather than coarse-grained primary minerals, and that primary silicate minerals can still be present even within the clay-size fraction (<2  $\mu\text{m}$ ).

Our intention was not to imply a universal or unidirectional offshore loss of primary minerals, but to highlight that the representation of weathering signals depends strongly on the grain-size fraction analysed and on hydrodynamic sorting during river-to-marine transport. As shown empirically by Dellinger et al. (2017), coarser fractions tend to be strongly diluted by primary silicates, whereas finer fractions increasingly reflect secondary clay contributions. At the same time, we agree that even the <2  $\mu\text{m}$  fraction may contain inherited primary minerals such as feldspars or micas, which must be considered when interpreting clay-based isotope records. For Esplugafreda, Jaimes-Gutierrez et al. (2024) examined the mineralogy across different size fractions (<0.5  $\mu\text{m}$ , 0.5-2  $\mu\text{m}$ , 2-16  $\mu\text{m}$ , and bulk detrital fraction), with the clay mineral fractions (<2  $\mu\text{m}$ ) being depleted in primary silicates.

With respect to sediment transport processes, studies from the Yangtze Estuary show that estuarine processes such as flocculation can promote preferential settling of clay minerals nearshore, while some fine primary particles may be transported further offshore, leading to a decoupling of dissolved and particulate Li isotope signals (Yang et al., 2021, 2025). Other systems, such as the Amazon, show that mechanical sorting related to particle size and settling velocity can favor offshore transport of sheet-like clay minerals (e.g., Gibbs, 1977; Liu et al., 2023). These contrasting behaviors highlight that downstream redistribution of clay and non-clay minerals is strongly system-dependent and cannot be assumed a priori.

We have therefore revised the text and the associated interpretative framework to clarify that (i) the fidelity of clay-based Li isotope signals depends on the size fraction analysed, (ii) hydrodynamic sorting and flocculation can variably redistribute clay and non-clay minerals during river-to-marine transport, and (iii) analysing the  $<2 \mu\text{m}$  fraction reduces, but does not eliminate, dilution by primary silicates.

**Modified lines 145 to 153:** In detrital sediment archives, only part of this boomerang trend is typically observed because of mixing of the neoformed clays with primary silicate material, especially at low  $W/D$  conditions (Dellinger et al., 2017). Therefore, continental and marine detrital records may need to be interpreted differently (e.g., Pogge von Strandmann et al., 2021; Ramos et al., 2022, 2024; Jones et al., 2023; Jaimes-Gutierrez et al., 2025; Rush et al., 2025; Wei et al., 2025). Because finer sediment fractions tend to be preferentially transported further offshore due to hydrodynamic sorting during river to marine transport, clay-sized records may be more clearly expressed in some marine sedimentary records (e.g., Gibbs, 1977; Liu et al., 2023). Such biases resulting from mixing with primary silicate grains in bulk sediment samples can potentially be reduced by analysing the clay size fraction ( $<2 \mu\text{m}$ ).

We also refer the readers to Jaimes-Gutierrez et al. (2025) who discusses the interpretative frameworks:

**Lines 163 to 164:** A detailed discussion on the lithium isotope interpretative framework, including the roles of grain size, hydrodynamic sorting, and lithology, is provided in Jaimes-Gutierrez et al. (2025, Supplemental Material).

- Section 4.1: is there any correlation between  $\delta^7\text{Li}$  and the mineralogy of clays? I think this should be discussed somewhere in the manuscript since different clays could have distinct fractionation factor during process of Li incorporation or adsorption into clays.

**Response:** We thank the reviewer for this important point and agree that mineral-specific fractionation during Li incorporation or adsorption into different clay phases could potentially influence  $\delta^7\text{Li}$  signatures. As also noted by Reviewer 1, we have explored this possibility by examining cross-plots between  $\delta^7\text{Li}_{\text{clays}}$  and the relative abundances of the main clay mineral groups (illite/smectite, illite, and kaolinite).

These comparisons do not reveal a systematic correlation between  $\delta^7\text{Li}$  and clay mineralogy within our dataset, suggesting that the observed  $\delta^7\text{Li}$  variations are not primarily driven by changes in mineral assemblage.

We agree, however, that better constraints on mineral-specific fractionation factors remain an important open question, particularly for interpreting subtle  $\delta^7\text{Li}$  variations in sedimentary records.

**Modified lines 395 to 410:**

The clays of the Rin section have a mean lithium isotope composition of  $-2.9 \pm 0.5\text{‰}$  ( $1\sigma$ ) (**Fig. 5B, Table S2**). Between 5.8 and 18.2 m, the mean composition is  $-3.4 \pm 0.2\text{‰}$ . Above this, from 20.3 to 39.5 m, the mean composition is  $-2.6 \pm 0.2\text{‰}$ , which corresponds to a shift towards more positive values of  $\sim 0.8\text{‰}$ . The minimum value of  $-3.7\text{‰}$  is seen before the Claret Conglomerate, and the maximum value of  $-2.2\text{‰}$  occurs immediately after the Claret Conglomerate, indicating a total range of up to  $\sim 1.5\text{‰}$ . The  $d^7\text{Li}$  values measured on carbonate

nodules have a maximum value of 8.6‰, a minimum value of -3.0‰, and a mean composition of  $0.9 \pm 4.0$ ‰ (Fig. 5C). **No clear temporal trend is observed in the  $\delta^7\text{Li}_{\text{nodules}}$  record. No systematic correlation is observed between  $\delta^7\text{Li}_{\text{clays}}$  and the relative abundance of individual clay minerals within analytical uncertainty (Fig. S1).**

At the Esplugafreda section, the clays have a mean lithium isotope composition of  $-3.7 \pm 0.7$ ‰ (Fig. 6B, Table S3). The pre-PETM samples (0-10 m and 21-28 m, Jaimes-Gutierrez et al., 2024 and references therein) have a mean composition of  $-3.8 \pm 0.2$ ‰. The POE samples (~15–21 m) have a composition of  $-3.2 \pm 0.2$ ‰, and the syn-PETM sediments have a composition of  $-3.0 \pm 0.2$ ‰, indicating a PETM shift towards more positive values of  $\sim 0.8$ ‰. The post-PETM sediments have a composition of  $-4.5 \pm 0.4$ ‰. **Samples from the pre-PETM, Pre-Onset Excursion (POE), and syn-PETM intervals display relatively heavier  $\delta^7\text{Li}_{\text{clays}}$  values, whereas recovery-phase samples form a distinct cluster characterised by lighter  $\delta^7\text{Li}_{\text{clays}}$  (Fig. S2).**

- Section 4.3: were the Li concentration measured on those samples? Any weathering-related changes in Li/Ti or Li/Al in both sections?

**Response:** Li concentrations were measured, but trace elements were not. This would be a good approach for future work to evaluate the feasibility of using pedogenic carbonate nodules and weathering archives.

- Lines 494-495: see also all the studies that report increase dissolved Li isotope composition and clay formation in rivers when crossing large floodplains (e.g. Bagard et al., 2015; Dellinger et al., 2015; Maffre et al., 2020; Pogge von Strandmann et al., 2017; Pogge von Strandmann and Henderson, 2015). The proposed interpretation here, i.e. “the shift towards increased incongruent weathering, characterized by enhanced clay formation in the floodplain deposits” is consistent with all these studies on modern rivers with floodplains.

**Original:** These processes would lead to decreased clay formation in the uplands (Kump et al., 2000; Riebe et al., 2004), but increased clay production and accumulation in lowlands, where longer sediment residence times promote authigenic clay formation.

**Response:** Thank you for highlighting this point. We have added references to some of the mentioned studies.

**Modified lines 510 to 514:** These processes would lead to decreased clay formation in the uplands by shortening water-rock interaction times (Kump et al., 2000; Riebe et al., 2004), while shifting clay production and accumulation towards lowland environments, where longer sediment residence times promote authigenic clay formation, consistent with modern river floodplain processes (e.g., Dellinger et al., 2015; Maffre et al., 2020).

- Lines 497-499: see also the study from Xu et al., (2021)

**Original:** Increasing evaporation, as recorded by gypsum lenses in Esplugafreda (Baceta et al., 2011; Khozyem, 2013; Jaimes-Gutierrez et al., 2024 and references therein), could also result in oversaturated waters, which would favour clay formation, with higher clay  $\delta^7\text{Li}$  compositions reflecting equilibrium with the water composition (Pogge von Strandmann et al., 2023).

**Modified lines 518 to 522:** Increasing evaporation, as recorded by gypsum lenses in Esplugafreda (Baceta et al., 2011; Khozyem, 2013; Jaimes-Gutierrez et al., 2024 and references therein), could also result in oversaturated pore waters, favouring clay formation. Experimental and field-based studies show that enhanced evaporation and reduced water availability increases dissolved  $\delta^7\text{Li}$  values in soil and pore waters, leading to higher  $\delta^7\text{Li}$  compositions of clays forming in equilibrium with these fluids (Xu et al., 2022; Pogge von Strandmann et al., 2023).

- Lies 502-504: This is not clear to me, why faster runoff results in a positive  $\delta^7\text{Li}$  excursion in the clays?

**Original:** Hydrological controls would also potentially enhance a positive  $\delta^7\text{Li}$  clays excursion with a weathering starting point in a supply-limited weathering domain (Fig. 7), as faster runoff would result in less time for water-rock interaction, leading to less time for clay formation (Zhang et al., 2022).

**Response:** We thank the reviewer for this clarification. Faster runoff does not directly result in a positive  $\delta^7\text{Li}$  excursion in clays, and we did not intend to imply a simple causal relationship. Indeed, modern observations show that increased runoff leads to shorter water–rock interaction times and lower dissolved  $\delta^7\text{Li}$  values in rivers (Zhang et al., 2022). The positive  $\delta^7\text{Li}_{\text{clays}}$  excursion we discuss instead reflects an indirect, system-scale effect. Increased runoff reduces secondary mineral formation in upland settings, but simultaneously enhances sediment export and storage in floodplains. In these lowland environments, fine-grained sediments can experience longer residence times and continued interaction with pore waters, during which clay alteration and lithium isotope fractionation may proceed. As a result, the clay fraction preserved in lowland deposits can record higher  $\delta^7\text{Li}$  values, even though instantaneous river waters are isotopically lighter during periods of high runoff. We have revised the manuscript text to clarify this distinction and to avoid implying a direct runoff– $\delta^7\text{Li}$  relationship for clays.

**Modified lines 522 to 528:** Hydrological controls can also influence lithium isotope signatures by regulating water–rock interactions, sediment transport, and secondary mineral formation (Fig. 8). In supply-limited weathering regimes, increased runoff shortens water–rock interaction times and lowers dissolved  $\delta^7\text{Li}$  values in river waters (Zhang et al., 2022), while enhanced sediment transfer promotes sediment storage and prolonged water–sediment interaction in floodplains, where continued clay alteration and isotopic re-equilibration can yield relatively higher  $\delta^7\text{Li}$  values in the clay fraction preserved in lowland deposits.

- Figure 2 & 3: could be combined (panel A and Panel B)

This would be a good idea given that they illustrate the evolution of Li isotopes, but Fig. 3 text would become too small, so we decided to keep it in its current state.

- Figure 4: I suggest indicating the different periods (pre, POE, onset, body and recovery) on the right of the figure to help the reader not so familiar with the PETM timing and evolution.

We have modified Fig. 4 accordingly.

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