Reviewer comments and responses

We thank the reviewers for their careful evaluation of our manuscript and for their constructive comments and suggestions. We have revised the manuscript extensively in response to the reviews. Our responses are provided in blue text. All figure, table, and section references refer to the revised manuscript unless otherwise noted.

REVIEWER 1 (Michael Darin):

I want to commend the authors for the revisions to the discussion and conclusions, which now appropriately temper the manuscript's central claims. The conclusions no longer assert that pre-6 Ma fault systems demonstrably carried Li-enriched fluids, and instead state that the data are "compatible with" but do not "uniquely identify" older Li source reservoirs. This is a more defensible framing than in the original submission, and I appreciate the authors' willingness to revise their interpretation accordingly. Several concerns remain, however:

Accept. Thank you for the second review of this manuscript. Responses to individual comments are below.

(1) Selective partition-coefficient fit — The claim that "relatively low lithium concentrations in calcite do not necessarily imply lithium-poor parent fluids" rests on a forward-modeling exercise in which only one of nine tested partition-coefficient scenarios (Fuger et al., 2019, intermediate) overlaps appreciably with the observed dataset. This should be stated explicitly wherever the argument is invoked, including in the conclusions, so readers can weigh the strength of the underlying support.

Accept. We have revised the text in Section 5.1 to state explicitly that only the intermediate partition coefficients of Füger et al. (2019) provide a close correspondence with the observed calcite Li concentrations, whereas the remaining partitioning scenarios show only partial overlap. We have also added text to Section 5.1 and Supplemental Material 8 to clarify that this is not intended as a model-selection exercise or to identify a uniquely applicable partitioning relationship for the Clayton Valley system. Rather, we compare the observed data with the range of experimentally derived partition coefficients currently available in the literature to evaluate whether they predict calcite Li concentrations compatible with our observations. We further clarify that Li partitioning is highly sensitive to growth rate and pH, that these variables are unconstrained for the natural samples, and that the intermediate Füger et al. (2019) partition coefficients are discussed because they provide the closest correspondence among the published experimental calibrations, rather than because they are uniquely applicable to the Clayton Valley system.

(2) Circularity of the fluid input — The partitioning argument uses modern Clayton Valley brine Li/Ca as a proxy for paleo-fluid composition in calcite up to ~15 Myr old. Modern brine composition reflects the cumulative product of basin evolution, potentially including post-6 Ma volcanic input, and cannot be assumed to represent pre-6 Ma fluid chemistry. This limits how much weight the partitioning framework can bear as support for any pre-6 Ma Li-bearing fluid scenario, including the newly proposed granitic source.

We agree that modern Clayton Valley brine compositions should not be interpreted as representative of pre-6 Ma fluid chemistry. However, this limitation is already explicitly acknowledged in the manuscript, where we state that the forward-modeling exercise is "not intended as a formal reconstruction of paleo-fluid compositions," that "modern Clayton Valley brines may not be representative of all paleo-fluid compositions present during basin evolution," and that the calculations provide only a "first-order test" of whether the observed calcite lithium concentrations are compatible with precipitation from Li-bearing basin fluids. We have nevertheless added a brief clarification in Section 5.1 to further emphasize that the modern brine compositions are used only as a first-order reference composition for evaluating the plausibility of the partitioning calculations, and not as a reconstruction of paleo-fluid chemistry.

(3) New, unsupported source hypothesis — The revised conclusions introduce Jurassic–Tertiary granitic basement rocks as a candidate older Li source. This is a new claim not evaluated elsewhere in the manuscript, as no Li concentration, isotopic, or spatial data are presented linking these granitic units to the five pre-6 Ma calcite samples. As written, this reads as speculative and should either be supported with data/citations or presented explicitly as a hypothesis for future work rather than as a conclusion.

Accept. This discussion was originally added in response to Reviewer 2's first-round request that we explicitly consider alternative pre-6 Ma lithium source reservoirs. Our intention was not to identify or demonstrate these granitic rocks as lithium sources, but rather to discuss plausible alternative hypotheses. We have revised the text in Section 5.2 to further emphasize that these remain speculative possibilities for future investigation, and that our dataset is better suited to evaluating the timing, temperatures, and pathways of lithium-bearing fluid circulation than to uniquely identifying lithium sources.

(4) Feasibility vs. evidence — The core distinction we raised still applies: demonstrating that low-ppm calcite Li values are geochemically compatible with Li-bearing fluids is not equivalent to positive evidence that such fluids were present. The revised conclusions largely respect this distinction now, and we ask only that this framing be carried through consistently (e.g., avoiding language like "indicates" in favor of "is compatible with" at Line 503 in the revised text).

Accept. Text modified accordingly in the Conclusions and in the Discussion to consistently distinguish between geochemical compatibility and direct evidence for Li-bearing parent fluids.

(5) Discrepancy between response letter and revised manuscript text — The authors' response letter states that they "now interpret the dataset as recording evolving structurally controlled fluid systems through time, with the strongest evidence for significant Li enrichment associated with younger basin-fill and fault systems that are contemporaneous with, or post-date, emplacement of the major late Miocene silicic volcanic units." This is a substantive and welcome concession, but it does not appear anywhere in the revised text. Instead, the abstract retains language stating that "several veins containing measurable lithium yield U–Pb ages older than the ~6 Ma Rhyolite Ridge

tuff… suggesting that structurally focused lithium-bearing fluid circulation may have locally predated emplacement of this unit" — which foregrounds the pre-volcanic framing the authors state they have moved away from, without stating the inverse and, in my view, better-supported pattern: that the strongest Li enrichment in the dataset is associated with fluids contemporaneous with or post-dating the 6.0 Ma Rhyolite Ridge tuff and other latest Miocene silicic volcanic units. I implore the authors to incorporate this explicit statement, as described in their response letter, into the abstract and conclusions, so the manuscript's framing matches the interpretation they describe having adopted during revision.

Accept. We have revised the Abstract to acknowledge that the highest lithium concentrations are associated with younger basin-fill and fault systems, while retaining the evidence for earlier Li-bearing fluid circulation. We have also added a brief statement in Section 5.2 to reinforce this interpretation within the broader discussion of fluid evolution.

EDITOR (Nicolas Beaudoin):
Thank you for revising your manuscript, and complying with the reviewers comments. I think the manuscript still requires some corrections before being good for publications. These include the comments of the first referee, and my own that I detail hereinafter.

Accept. Thank you for the editorial handling and review. Responses to individual comments are below.
Among the remaining problems I pointed is the use of the SupMat 8. Although it is interesting that you provided a foreward geochemical model to better relate the [Li] of your paleofluids to the [Li] of the geological reservoirs, that exercise raises a couple questions that requires clarifications in the main text:

1) Why choosing the average upper continental cruste abundance for Li from Teng et al, 2004 that differs from the values fetched from Coffey et al. 2021, that were acquired in your area? (p. 24)

Accept. Teng et al. (2004) is not used as an input to the forward model presented in Supplementary Material 8. The forward model uses only modern Clayton Valley brine Li/Ca compositions from Coffey et al. (2021). The upper continental crust value from Teng et al. (2004) is included solely as a general geochemical reference in Figure 5 and is not used in any model calculations. To avoid this confusion, we have clarified this distinction in the main text and added basin-fill Li concentrations from Coffey et al. (2021) to Figure 5.

2) Why do you write l. 424 that the several modeled scenarios… compare favourably with the observed dataset? When looking at it, that writing is misleading. There are partial overlap (statistically insignificant) in a few models, but only one is favourably to your interpretation. I don't think it necessarily is a bad thing to propose alternate scenario, but it must be clear. Here, I would like you to discuss why the mid Fugler is more likely to be chemically/geologically convincing than the other ones. You need to justify soundly these two points that the conclusion of the paper does not look model-driven.

Accept. We have revised Section 5.1 to state explicitly that only the intermediate partition coefficients of Füger et al. (2019) produce a close correspondence with the observed calcite Li

concentrations, whereas the remaining scenarios show only partial overlap. We have also clarified that this is not a model-driven or model-selection exercise intended to identify a uniquely applicable partitioning relationship for Clayton Valley. Rather, we compare the observed dataset with the range of published experimental partition coefficients to evaluate whether realistic partitioning behavior can reproduce the measured calcite Li concentrations. The intermediate Füger et al. (2019) scenario is emphasized because it provides the closest correspondence, not because we infer that it uniquely represents the natural system. We further note that Li partitioning is highly sensitive to growth rate and pH, for which we lack independent constraints in these samples. Corresponding revisions have also been made in Supplemental Material 8.

About the vein geochemistry:

3) Figure 8 A plots the d18O water vs d13C carbonates. That does not really make a lot of sense, because there is a few geological factors that can makes C fractionation during precipitation (temperature, organic matter, microgravity). So what is it you showing there? Why not showing the usual D18O fluid vs T47 and D18 O CaCO3 as oblique? The interpretation d13C value higher than typical mantle signature is not convincing at all, -5‰ being very classical in basinal fluids (see Beaudoin et al., 2021, for a review). This figure and paragraph must be rewritten.

Accept. We have revised Figure 8A to the conventional presentation of reconstructed fluid $\delta^{18}O$ versus precipitation temperature with calcite $\delta^{18}O$ contours. We also revised the accompanying text to focus on the reconstructed fluid compositions and temperatures, removed the comparison with mantle $\delta^{13}C$ values, and clarified the interpretation of the reconstructed fluid compositions in the context of precipitation temperature and fluid source.

4) You need to explain how you obtain the D18O fluids from the clumped isotopes: the D47 need to be reported in the text, at least for the range, not only in temperature. It must be made clear that D18Owater is interpreted using a fractionation equation. Which one have you used? It should appear at least l. 347, and there is no information in your sup mat.

Accept. We have added text to the Methods (Section 3.2) describing how clumped-isotope temperatures and reconstructed fluid $\delta^{18}O$ values were calculated, including the use of the Kim and O'Neil (1997) calcite-water fractionation equation. We have also expanded the Results (Section 4.4) to report the measured Δ_{47} values alongside the reconstructed fluid $\delta^{18}O$ values where these data are first presented. Additional methodological details have also been added to Supplemental File 7.

5) You write part 5.1 that Li concentration are highest in v-s basin fill units with a value at 460 ppm. This one sample is the only one that does not have a meteoric signature for the paleofluid (d18O fluid = 3 permil SMOW), so it requires to be discussed.

Accept. We have added to text to Section 5.1 to explicitly acknowledge and discuss the anomalous fluid isotopic composition of the highest-Li sample relative to the dominantly meteoric signatures of the remainder of the dataset.

6) I also do not understand why the two ages 6.8 and 4.2 are interpretated on the "morden day" schematic cross section and not on the 6 My one?

Accept. The placement of the 6.8 Ma sample has now been moved to the ~6 Ma reconstruction. The 4.2 Ma sample, however, is intentionally shown on the present-day panel because it post-dates the ~6 Ma reconstruction. Placing it on the ~6 Ma time slice would incorrectly imply that the vein was already present at that time, whereas the present-day panel is intended to represent the cumulative geological record following the ~6 Ma reconstruction.

Some more technical considerations that need to be addressed:
7) The 2sigma related to analytical uncertainty (D13C, D18O, D47ISE) should not be in the table 2 because you can put them in the analytical procedure. It'll make the table easier to read.

Accept. Text added to the methods regarding analytical uncertainties. Associated columns have now been removed from Table 2.

8) L. 225, no tilde here is required, just the correct values of T47 with their uncertainties.

Accept. We believe you are referring to Line 225. The text has been revised here and elsewhere to report the calculated clumped-isotope temperatures with their associated uncertainties rather than approximate values.

9) it is usual to thank the reviewers in the acknowledgment section of a paper.

Accept. Text added.

10) The zenodo link only host 4 sup files, I think it needs to be updated.

Accept. Zenodo repository updated and new URL provided.

You will find some of my remarks to be in line with the report from the referee #1. To be clear, there is a possibility that your model is right, but there is a little bit of logic bending in the way it is presented. With a few lines of discussion, either to convince that your choices are clearly supported, either to clearly set the limitation of your interpretation (e.g. 'we can't exclude an important part of the Li comes from volcanic events as already shown in the literature') then it will be good for publication.

We thank the Editor again for the careful and constructive review of our manuscript. We hope that the revisions made throughout the manuscript have addressed the concerns raised and clarified the scope, interpretation, and limitations of our study.