

Here we respond to comments of reviewer P.-H. Blard

This article provides an interesting method based on a sequential heating of olivines from surface lava flow samples. This technique permits to separate the cosmogenic ^3He and the mantle ^3He component. Their dataset obtained on 2 samples convincingly indicate that diffusion at 800°C preferentially release cosmogenic ^3He , permitting to determine this cosmic-ray produced ^3He with a better precision than using the standard crushing-melting method. This better precision is possible because the mantle component is not released at “low” temperature, reducing the impact of the uncertainty of the magmatic correction. However, although I think that this pilot study is interesting and should be published, I have major criticisms about the radicality of several conclusions of the authors, who write that the isochron method and the standard crushing-fusion methods yields imprecise or flawed ages. They cannot be so definitive and negative about these well-established methods, for two reasons: first, they didn't apply the heating step method on the same samples that those processed by the crushing-melting methods, and second, they build an isochron without using samples (or aliquots) that have been exposed to the same “dose” of cosmic rays. The most plausible conclusion is rather that their samples have various exposure ages (either because the lava have different ages, or because the sampled surfaces suffered differential erosion or shielding). I encourage authors to revise their manuscript taking into account this main criticism and also the other points listed below.

The major point of the paper is to a) indicate that step heating can be used to reasonably isolate cosmogenic He from mantle He in these olivines that are extremely He retentive during crushing and b) to estimate the most recent eruption age. Blard here makes two legitimate points, but neither directly impacts the key results of the paper.

We agree that it is likely the samples we plotted in the isochron violate the assumptions we stated: "This same fusion data can be cast as an isochron provided we assume the flows have the same exposure age and mantle component." Regardless, in the end we agree that the isochron plot as presented was not useful or well described. In a revised submission we will modify the discussion of the isochron approach, instead focusing on how even if the fundamental assumptions of the isochron approach are absolutely true, the high mantle He concentration and its limited range in our powder fusion analyses coupled with reasonable analytical uncertainties inevitably yields substantial uncertainties on cosmogenic He concentration. In other words, we test the isochron's use under ideal conditions for our real-world samples, and assess the best-case precision that could be obtained.

We will also provide newly obtained crush results on the lava flow subjected to step heating.

Line 40: "low" is rather imprecise. Provide actual range of U and Th concentrations."

Line 42: This belief is not really accurate: young lava (< 100 ka) may require correcting for radiogenic ^4He accumulation, because what matters is the $^3\text{He}_{\text{cos}}/^4\text{He}_{\text{rad}}$ production ratio, that is independent from the lava age (e.g. Blard and Farley, 2008; Blard, 2021). Olivines that bear significant amount of U (> 0.1 ppm) require more than 10% correction (R factor lower than 0.9, see for example figure 9 in Blard, 2021). In other words, a "young" lava does not necessarily imply that the radiogenic ^4He impact on the magmatic ^3He correction is negligible (Dunai and Wijbrans, 2000 = 5% correction; Blard et al., 2006 = up to 12% correction, even in lavas younger than 200 ka ; see R factors in Table 2 in Blard and Farley, 2008).

Line 42: What is a "young" lava?

All of this refers to our generic statements in the introduction that under certain limiting circumstances it is possible to reduce the nearly intractable 3 component problem (mantle + cosmogenic + radiogenic helium) to a solvable one that involves only the first two components. It would derail the logic of the introduction by digressing into the details requested here. Instead, we will add text to the discussion where we justify why, in these olivines with very high mantle concentrations and estimated eruption age of ~10 kyr, the limiting case is reasonably satisfied. The key conclusion is that there is so much mantle helium in these olivines that the radiogenic component in 10 ka olivines is not a factor of concern.

Line 49: Maybe add a caution here to mention that dry powdering under atmosphere may yield adsorption of non negligible amount of atmospheric helium on the phenocrysts/xenocrysts (Protin et al., 2016).

Easily done in revised version.

Line 51: Quote a source for the value of this atmospheric isotopic ratio.

Easily done in revised version.

Line 56: Figure 5 in Blard 2021 shows a modeling of the magmatic helium impact on the final $^3\text{He}_{\text{cos}}$ uncertainty.

We can point this out in revised version.

Line 68: Note that this approach involving a step heating to selectively release cosmogenic ^3He at around 1000°C is not a newly developed method. As written in Niedermann 2002: "The few papers which report stepwise heating data for cosmogenic He show the major release of ^3He from mafic and ultramafic minerals (olivine, pyroxene) below ~ 900 - 1100°C (Kurz 1986a; Staudacher and Allègre 1991, 1993a; Sarda et al. 1993; Schäfer et al. 2000)."

We are not claiming priority in demonstrating step heating He component resolution, we are just demonstrating its use for quantifying the cosmogenic ^3He concentration in an unusually problematic set of samples. We can add these references in revised version.

Line 75: Lava-dammed lakes are not shown on figure 1, could you please add them on this map? It would be very useful to clearly show the stratigraphic relationships between lavas and lakes.

This can be done in revised version.

Line 85: "Cosmogenic sampling". Since "cosmogenic" means "produced by cosmic rays", I suggest to reword.

Easily modified in revision.

Line 90: A picture of a sample would be very useful here.

Easily modified in revision.

Line 91: What is the typical sample thickness?

Easily modified in revision.

Line 97: It would be useful to show field pictures of the sampled lava surfaces (either in Figure 1, or in a new figure). Field photographs would be very useful to show the vegetation cover.

We will mention the existence of field photos of these lava flows in published references, especially Jackson and Stevens 1992.

Line 102-103: Are these mm-olivine phenocrysts or xenoliths?

We analyzed olivine crystals that could be either ol phenocrysts or disaggregated xenoliths. The references we cite suggest that the majority of the olivines are the latter (disaggregated during eruption), as we already stated in the "Geologic Setting" section of the manuscript.

Table 1: Why don't you provide thickness for samples 8, 10 and 11?

Will be added in revision.

Line 119: Along Cox et al 2022, quote here the first article that identified this issue of helium adsorption (Protin et al., 2016).

Will be added in revision.

Line 121: 1200 °C is well below the melting point of Mg-rich olivine (that is above 1500°C). Are you sure of 1) the accuracy of the furnace T control, and 2) are you really reaching fusion (isn't it an He extraction by complete diffusion)?

In revision we will add text to the effect that we are using the term "powder fusion" to mean "complete extraction of He at high temperature". We verify complete He extraction using the re-extract method. We don't fuse the olivines because ~1800 C is beyond the range of our furnace.

Line 135: Please provide the blank levels for each T step, as you did in the previous section for 1200°C.

Will be added in revision.

Line 136: It is surprising that the contribution of the 1400°C blank is lower than the one of the 1200°C blank.

Why? The system is being cleaned up by the steps that occur earlier.

Line 153: It would be useful to give the value of this shielding correction here.

Corrections are listed in Table 5, but in revised version we can mention them here.

Table 2: I think this is not necessary to provide ^3He and ^4He with 2 different units (cc/g and at/g). This would be much more useful and informative to add a column with the computed cosmogenic ^3He concentration corrected for the magmatic ^3He for each sample.

Easily modified in revision.

Line 172-173: "mantle component": I guess you mean "magmatic $^3\text{He}/^4\text{He}$ ratios", not magmatic helium concentrations, that need to be variable to ensure the isochron method working. Maybe rephrase to avoid confusion.

Easily modified in revision.

Line 176: Given that this isochron is built from samples having different exposure histories, I think you should use this number with a huge caution.

Agree. The isochron section will be completely rewritten in revision, as described above.

Figure 3: For a better readability, you could consider removing the vertical lines between each T extraction step.

Figure will be revised.

Line 209: Replace "u" by "mu", the greek m.

Easily modified in revision.

Line 218-219: Burnard et al., 2015 also provided experimental evidence for the presence of a significant "reservoir" of mantle helium at the grain boundaries.

There are no grain boundaries in our analyzed olivines- we are analyzing individual crystals. We can mention in revision that we have crystals, not small polycrystalline xenoliths as this comment seems to be imagining.

Figure 4: It would be useful to add an enlarged zoom centered on these "trail" of fluid inclusions.

Figure will be revised.

Line 237 and 240: It would be useful to add these relative increases in cosmogenic ^3He in Tables 2 and 3.

We will do this.

Line 240: "34 and 39%": Why are these 2 numbers different from the 36-42% range mentioned line 237?

This will be clarified in revision.

Line 241: What do you mean by "detailed step heat"? Do you mean an initial heating at lower temperature?

It is not obvious what temperature the additional steps would need to be to obtain best component resolution. This will be clarified in revision.

Line 253: It is strange to compute a mean using a set of samples with heterogeneous ages.

In the end we agree this mean computation adds no value. This section will be reworked in revision.

Line 266: "7.7 +- 4 ka". I think there are too many significant numbers.

Easily fixed in revision.

Line 269 to 271: You should rather compare VM1 and 3 with VM2, 6 and 8, not with the isochron that is built from a dataset that breaks the required assumption of an homogeneous dataset.

Line 279: I disagree with this conclusion. You could conclude that if the same 3 methods were applied on the same samples. Here you applied different methods on different surface samples. The most plausible conclusions from your data is that some samples (9, 10, 11) have experienced less exposure at the surface than others (1, 2, 3, 6 and 8). Regarding the building of the isochron, you did not respect the necessary assumption of using aliquots with homogeneous cosmogenic ^3He concentrations, so you obtain an underestimate average concentration. Nothing else can be concluded from that.

Line 291-292: As already mentioned before, this conclusion is a false overstatement. You cannot provide such a general conclusion about the isochron and the crush-fusion methods since you did not apply them to the same samples.

As already mentioned, we will rework the discussion of the isochron and add a crush measurement in revision. These will address this entire set of concerns.