Review : New Developments in Incremental Heating Detrital 40Ar/39Ar Lithic (DARL) Geochronology using Icelandic River Sand, Okwueze et al. (2024).

Overview :

This contribution attempts to improve on the DARL method by overcoming the inherent limitations in previous applications that employed exclusively K-Ar methods. This is done using a detailed 40Ar/39Ar step heating approach.

Much of the data are of high quality and show excellent release spectra. Data from some of the more complex step heating spectra are reasonably discussed.

Application of non-atmospheric trapped compositions for correcting plateau ages is also explained and justified. However, in the case of apparent non-atmospheric trapped components, I would use the isochron ages as these will be less affected by the trapped component issues.

The proposed method of 'partial fusion+averaged trapped component' is very poorly explained and it's implications are not clear. As read in the text, it appears to give a different, but similarly blurred picture of the age distribution of clastic materials as would the K-Ar method.

Most importantly, there is no raw data provided with the manuscript so age calculations cannot be verified or explored. **The manuscript cannot be accepted without this information.**

Specific comments linked to line numbers:

73 : Use consistent units throughout (Ma).

77: Figure 1 - is low resolution and scale bars cannot be read.

126: Table 2 - Need to express to the precision to the correct number of significant digits, e.g., RHRDV01-4 : 0.42±0.23 Ma.

126 : Table 2 - Why are the plateau and isochron ages of RJKBR01-h so discordant? Especially considering the total fusion age is also older than the plateau.

141: What does 'first order' mean in this context? Are you implying something about precision or accuracy requirements? I think this is important as it's at the heart of the matter – the balance between the data volume required by provenance studies versus the efforts to obtain the best precision and accuracy with the technique.

151: MSWD should be listed for both age spectra and isochrons. p for isochrons. Ages and uncertainties for isochrons.

151: Since you discuss K/Ca below, it should be illustrated on the plots, along with an indication of the average (integrated) K/Ca that the reader can reference.

155 : Describe what the gray points are.

157 : Figures - Resolution is low making figures difficult to read. Needs to be brought up to publication quality. Could resize the isochron diagrams to be the same dimensions as the age spectra without consuming extra page space. The isochrons should be expanded (for example RHRDV01-b, RHRDV01-d or RPJRS01-a), that is, not plotted to the 39Ar/40Ar intercept, but rather to show maximum detail of the data and their relation to the regression line.

Should reconsider the scale on the age spectra. Most of the negative age range is unused and simply compresses the apparent scatter. Some plots, for example RJKBR01-g should be plotted on a finer scale as detail in the spectra cannot be seen.

189 : "discordant plateau with steps in the same general age range...". A discordant plateau isn't really an age.

193 : Confusing as written - the ages are not consistent with the proximal terrain, but instead the terrain that borders it. According to your watershed map, the lithic ages should not exceed 5.5 Ma. But this is an important observation; because you've produced age spectra, rather than K-Ar type ages, you can use these reliable dates to ask questions about what transport mechanisms could bring older detritus into this basin. If these were single-step fusions (i.e. K-Ar), you could simply dismiss them as having excess 40Ar. These older clasts effectively highlight why this is a much more robust approach.

237 : I think you mean to say that K/Ca will be affected by various processes including, but not limited to, ...degree of source partial melting, etc.

241 : Surely at 1400°C the sample is fused and largely degassed. What does the 37Ar release curve look like?

245 : This comparison should be with bulk rock data since they would be equivalent to what you measured.

252 : Or you could simply split the grain, c.f., Ellis et al. 2017
https://doi.org/10.1016/j.chemgeo.2017.03.005

254 : "This coupled petrological analysis and age determination on single..."

265 : Interesting point as you would then expect that using a non-atmospheric trapped component (as outlined below) should (could?) bias your ages relative the K-Ar ages in the literature which would have used an atmospheric correction.

267 : You should show these in the table since you plot them in figure 8.

275 : Do you mean temperature range? or a singular temperature? multiple steps?

278 : This should be in the table for reference

281 : Unclear what's being referred to here - do you mean t(0) regressions? Why would these be different as compared to a standard analysis? Why would the blanks be different? Because this is a single step? Please explain more carefully.

282 : I'm not at all sure what you're proposing. Are you taking about fusing at a single temperature? Or doing steps only between 680-1140? What happens to the gas from the lower temperature steps? Do you discard it? Have you calculated how much time your method saves relative to a full step-heating experiment, especially as furnaces usually require high temp. burnout between samples?

288 : Panel B should say "Partial fusion age calculated with..."

288 : This sample in panel B, RHRDV01-b, becomes even less compatible with the upper age limits indicated by the watershed boundaries (5.5 Ma).

288: You should put an actual 1:1 reference line in here for comparison. The line plotted here has no meaning.

298 : Fifteen. 4 of them were rejected – at least that is what the table indicates.

Additional comments (recommendations, not requiring action for resubmission):

1) The approach could be significantly improved if the samples were pre-treated more rigorously, i.e., crushed, phenocrysts and alteration phases removed and remaining groundmass leached to obtain pristine groundmass fragments as is usual for basalt dating. Obviously clast sizes limit the amount of datable material that can be processed this way, but this pre-treatment will limit the interfering effects of alteration and excess argon in phenocrysts, making for more robust data. Having a smaller amount of high purity material is better.
2) A CO2 laser-based approach would have lower blanks and can be run 24-7 for greater throughput. But even then, step-heating takes time.
3) I think this approach will struggle to be applied as a high-N type of technique (like U/Pb zircon) because initial conditions and assumptions (initial daughter composition, closed system behaviour) have significant impacts and cannot be

easily assessed without detailed analysis. Single-step analysis will not alleviate this problem.

4) Assuming a ‘regional’ trapped component composition will only blur the picture of sample ages, giving this technique even less resolution.
5) A better approach would be to pre-screen the materials for key geochemical data and then use the geochemistry as guide to select grains for high precision and accuracy step-heating analysis. Step heating analysis is the greatest strength of the Ar/Ar method and so should be leveraged for maximum benefit.