We (Brueseke, Benowitz, Trop) found, the currently under discussion manuscript, *New Developments in Incremental Heating Detrital $^{40}Ar/^{39}Ar$ Lithic (DARL) Geochronology using Icelandic River Sand by* Odinaka Okwueze, Kevin Konrad, and Tomas Capaldi well written and a good contribution to the continued use of the DARL (Detrital Argon Lithics) geochronology approach. We agree the magmatic history of the glaciated Iceland magmatic province will benefit from applications of the DARL technique, as will other relatively remote and glaciated area such as the Cascades Arc of Northwestern United States.

We graciously recommend some key adjustments to the text, given our and others past work doing both $^{40}Ar/^{39}Ar$ step-heating and modified single grain fusion on ground mass, whole rock chips, and discrete mineral grains from gravel- and sand-sized volcanic-lithic clasts.

We first reported $^{40}Ar/^{39}Ar$ ages on volcanic-lithic grains from modern river sands in the Wrangell Volcanic arc at a 2014 conference (Benowitz et al., 2014), where we demonstrated that a modified heating schedule of sand-sized volcanic lithics was more efficient and accurate for DARL analyses. This was based on incremental heating single sand-sized volcanic-lithic grains and then modifying our fusion schedule based on these results. We also recommended when applying DARL to other regions standard step-heating be performed before developing a fusion or modified (shortened) step-heat schedule. At the time we were concerned about excess ^{40}Ar not excess ^{36}Ar (which Okwueze et al. document). We agree that excess ^{36}Ar is an underappreciated aspect of $^{40}Ar/^{39}Ar$ geochronology (Benowitz et al., 2018). These method details were explained in a subsequent Geosphere article (Trop et al., 2022; relevant aspects are copied below) and inasmuch, should be noted as where the DARL technique originated and was first published. Furthermore, Kenny et al. (2022) also performed $^{40}Ar/^{39}Ar$ incremental step-heats on detrital sand volcanic-lithic grains. We also performed and published (Trop et al., 2022) incremental step-heats on volcanic-lithic grains when results were questionable or were of key age spans as one of our goals was to determine the age of initiation for the Wrangell Arc. VanderLeest et al (2020) also applied step-heats to detrital clasts.

Thus, we kindly suggest that Okwueze et al. revise their text and clarify that $^{40}Ar/^{39}Ar$ step-heats and modified fusions were done previously on modern river volcanic-lithic grains, consequently the contribution here builds on these prior studies. This key fact should be made clearer in this manuscript; as-is, the DARL technique as described is not new or particularly novel, especially given that it is centered on n = 15 grains (vs. n = ~2600 grains; Trop et al., 2022). Additionally, Kenny et al., 2022 performed modified step-heats on 50 grains with step counts varying from 2 to 15 (?) steps to optimize number of grains vs. diffusion profile information. See their supplemental files.

We understand there is so much literature out there, that it is easy to miss aspects of past research and take no offense and based on conversations with the corresponding author know none was meant. We are genuinely excited to see more DARL work reported from this research team and others.

Specific recommended changes:

Something like the following for their introduction: *Following previous combination $^{40}Ar/^{39}Ar$ incremental step-heating and informed modified fusion procedure on modern river volcanic-lithic grains (Benowitz et al., 2014, Trop et al., 2022), we developed a new DARL partial fusion procedure specific to the magmatic products of Iceland*.

Below are additional changes and information re: relevant past work we recommend the authors consider during their revision.

Around line 15 (Benowitz et al., 2014; VanderLeest et al., 2020; Kenny et al., 2022; Trop et al., 2022 did ^{40}Ar/^{39}Ar incremental step-heats on detrital cobbles and/or sand). Here we present a new methodology for capturing the magmatic history of fine grained extrusive volcanic rocks using single grain detrital ^{40}Ar/^{39}Ar incremental heating geochronology. The DARL (or Detrital Argon Lithics) **method thus far** has consisted of ^{40}Ar/^{39}Ar total fusion analyses, which pose a problem in the case of Iceland, due to the nature of its young glassy lava flows commonly displaying subatmospheric ^{40}Ar/^{36}Ar isochron intercepts and low ^{40}Ar*.

Around line 25 Benowitz et al., 2014**;** Trop et al., 2022 did both a combination of informed single grain fusions based on incremental heating results; n = ~2600 grains are what was eventually analyzed and reported in Trop et al (2022)

For this reason, **we propose combining the aspects of the total fusion and incremental heating** DARL methodologies to acquire age data for the large N values needed for detrital studies while improving the accuracy of total fusion DARL analysis.

Around line 40 (DARL has been applied to sand and pebble grains and cobbles, and as a combination of modified fusion and incremental step-heating…. Benowitz et al., 2014; VanderLeest et al 2020; Kenny et al., 2022; Trop et al., 2022)

The detrital ^{40}Ar/^{39}Ar lithic (DARL) method is a relatively new detrital geochronological tool that **thus far** employed ^{40}Ar/^{39}Ar total fusion analyses on single grains or multi-grain aliquots recovered from cobble sized (>10 cm) volcanic sediments (Trop et al., 2022; Brueseke et al., 2023).

Around line 50 (this has already been done…Benowitz et al., 2014; Kenny et al., 2022; Trop et al., 2022**)**

Here **we expand** upon the method through incremental heating experiments on single coarse sand grains of volcanic lithic fragments from Icelandic rivers.

Line 220 (This seems a little overstated given ~10 years of DARL step-heating work and the orders of magnitude larger number of individual DARL analyses from Trop et al., 2022 and the combination of geochemistry and DARL dating in VanderLeest et al 2020 and Brueseke et al., 2023).

Provided the level of difficulty, the incremental heating DARL experiments worked well and **represent an advancement in the field of detrital geochronology.**

Around line 250 (this was sort of done -Trop et al., 2022- to evaluate alteration and excess ^{40}Ar and for sure the DARL method has been applied to dominantly mafic bedrock sources.)

Although the internal concordance test afforded by the incremental heating method has many advantages, the long analyses time hinders the method's use for detrital geochronology studies, which rely on high= N values. Therefore, **we propose that a subset of grains from a sampling site be analyzed with the incremental heating method** in order to define the best partial fusion temperature ranges and appropriate assumed $^{40}Ar/^{36}Ar0$. More work is required to assess the validity of the method in different geologic settings, but the primary data from this study indicates the method is valid and **allows for detrital geochronology studies of dominantly mafic bedrock sources.**

Around Line 260 (at the time we used 295.5 for atmospheric $^{40}Ar/^{36}Ar0$)…which now is not standard…but does make the reference to our work a bit confusing…perhaps remove?).

An atmospheric $^{40}Ar/^{36}Ar0$ was assumed with the age calculations and the results were ~equivalent to K/Ar ages collected from the region.

Around line 295

The DARL method provides a novel means of constraining the volcanic history of a region through detrital geochronology of lithic grain sand samples.

Please Add the reference to Trop et al. (2022), given that is where the DARL technique originated and was first published:

The DARL method (***Trop et al., 2022***) provides a novel means of constraining the volcanic history of a region through detrital geochronology of lithic grain sand samples.

Other manuscript notes that need to be addressed:

Please define what you mean by discordant: We think we know what you are referring, but it is never defined/explained how you are applying this broad term.

Table 1: Please add the known age range for magmatism for each sample/drainage.

How often did you measure mass discrimination? Did it drift? Could applying the "incorrect" mass discrimination explain your excess ^{36}Ar (and excess ^{40}Ar) measurements?

^{36}Ar was measured on a more sensitive electron multiplier? Where ^{40}Ar was measured on a sensitive (but less so?) faraday? Is this a factor in the excess ^{36}Ar measurements?

We doubt these are controlling factors on the excess ^{36}Ar measurements…but these factors should be at least documented and mentioned-dismissed in the text/methods.

Perhaps more discussion on how modern mass spectrometer instrumentation allows for the clearer identification of excess ^{36}Ar could be added?

Around Line 275

Therefore, we can calculate the partial-fusion age between those temperature steps, using an $^{40}Ar/^{36}Ar_0$ that is representative of our dataset (296 ± 4; Figure 8).

What was the range of determined $^{40}Ar/^{36}Ar0$ for all the grains analyzed?

289.7 to 300.3….Is it really sensible to assume a single subatmospheric $^{40}Ar/^{36}Ar_0$ for all samples? Given most results approximated or were greater than 298.56 ± 0.62 (Lee et al., 2006)?

The 296 ± 4: Is that a weighted average? The uncertainty is propagated during the age calculations?

Isochron plots:

Are the same steps used for the plateau age determinations used for the isochron age determinations? They should be. It seems for some of the samples this is not the case? It is hard to tell given the number of steps used in the isochron determinations are not listed in table 2. If always the same number of steps/same steps are used for isochron regressions as were used for the plateau age determinations (as they should be? Unless justified), please mention in text.

RPJSO1-e

Would you consider this stepping up age spectrum indicative of loss? If so, is it appropriate to perform a regression back to initial $^{40}Ar/^{36}Ar$ (Isochron plot) given the documented loss?

Figure 10

This is a key figure…but we don't see the negative original age determinations in Table 2 and there are no supplemental isotopic files. Please add the negative (original) age determinations to Table 2 and add full supplemental files. Schaen et al. (2021) community based (dozens of noble gas lab authors) makes a strong case and sets out examples of how $^{40}Ar/^{39}Ar$ isotopic information should be documented in scientific manuscripts. Regardless if Schaen et al. (2021) is followed to the "T", detailed isotopic tables are required to be included with $^{40}Ar/^{39}Ar$ geochronology publications to be able to evaluate the authors results/interpretations/methods.

We see on table 2 you correct for excess ^{36}Ar, but don't correct for excess ^{40}Ar. Would it be better to use the original isochron age determinations for all analysis instead of plateau ages?

Figure 10….B looks stretched? i.e., Why are uncertainties so big?

Or are uncertainties blown up with the applied 296 ± 4 $^{40}Ar/^{36}Ar_0$, hence MSWD goes down simply because of the larger uncertainties? Compared to 298.56 ± 0.62 $^{40}Ar/^{36}Ar_0$ (Lee et al., 2006).

What would be the MSWD for graph A be if the youngest three ages were parsed? Seems those are biasing everything and for graph B all the ages are being modified (some far away from there "actual ages"!!!).

Can you please add a table of original ages/uncertainties for all samples vs. modified ages/uncertainty with the assumed 296 ± 4 $^{40}Ar/^{36}Ar_0$ determination. We think this is a key aspect…Yes you are shifting the youngest ages, but you are also shifting the other ages, Is that appropriate given the large variations in actual measured/calculated $^{40}Ar/^{36}Ar_0$?

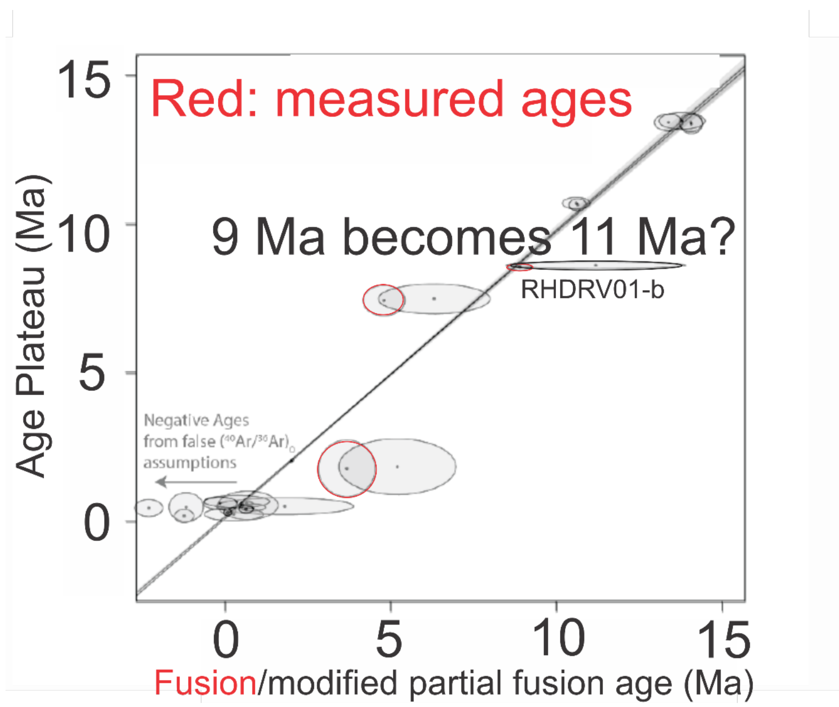
For example, on figure 10….sample RHDRV01-b gets shifted from a total fusion age of 8.9 Ma (± 0.03) with a small uncertainty and becomes >11.0 Ma on 10B with a huge uncertainty (± ~5 Ma?).

Is this an improvement over the original accurate and precise age determinations?

Can you get negative ages simply due to statistics? i.e. An age result of 10 ka ± 20 ka on a lava means given enough analyses you would get a negative age from the same sample.

We are not sure if trying to make "exact" geologic interpretations from modified negative $^{40}Ar/^{39}Ar$ follows best practices. Yes these grains are young and the authors can robustly state that, but we are not sure applying a 296 ± 4 $^{40}Ar/^{36}Ar_0$ to a negative age with a measured 289.67 $^{40}Ar/^{36}Ar_0$ makes for a geologically more meaningful age.

Rough figure showing large shift from measured to modeled ages.



Is 1.79 Ma age on Figure 10? (RJKBR01-h)? might be…. forgive us if it is.

Line 80 (>2 mm sized grains are granule sized gravel as opposed to sand sized grains, so the text should state that fine gravel (or granules) and sand was analyzed**).**

The bulk sediment samples were sieved and grains from the **2-3 mm size fraction** were selected for all sites except RJKBR01, where the 1-2 mm size fraction was used. Each selected grain was separated and given a unique identifier (i.e. -A; Figure 2).

Data Availability

Please include a link to all isotopic information (preferably in excel format) and supplemental figures using a file-sharing site like https://zenodo.org/records/802100.

As is, it is impossible to replot the presented data, evaluate the results, etc.

Summary Suggestion:

Perhaps a better DARL method for Iceland would be to: Degass/not measure/pump out lower temperature steps (below 680 °C). And then a apply a 296 ± 4 $^{40}Ar/^{36}Ar_0$ for the negative age determinations: but acknowledge these modeled age determinations are approximations and not indicative of exact geological eruptive events.

Review References:

Benowitz, J.A., Davis, K.N., Brueseke, M.E., Trop, J.M., and Layer, P., 2014, Investigating the lost arc: Geological constraints on ~25 Million years of magmatism along an arc-transform junction, Wrangell Volcanic Belt, Alaska, Geological Society of America Abstracts with Programs, Vol. 46, No.6, p.363.

Benowitz, J.A., Miggins, D.P., Koppers, A.A. and Layer, P.W., 2018, November. Why are some young volcanic rocks undateable: Chemistry, Environment, or instrumentation?. In *GSA Annual Meeting in Indianapolis, Indiana, USA-2018*. GSA.

Kenny GG, Hyde WR, Storey M, Garde AA, Whitehouse MJ, Beck P, Johansson L, Søndergaard AS, Bjørk AA, MacGregor JA, Khan SA. 2022, A Late Paleocene age for Greenland's Hiawatha impact structure. Science Advances.

Schaen, A.J., Jicha, B.R., Hodges, K.V., Vermeesch, P., Stelten, M.E., Mercer, C.M., Phillips, D., Rivera, T.A., Jourdan, F., Matchan, E.L. and Hemming, S.R., 2021. Interpreting and reporting 40Ar/39Ar geochronologic data. *GSA Bulletin*, *133*(3-4), pp.461-487

Trop, J.M., Benowitz, J.A., Kirby, C.S. and Brueseke, M.E., 2022. Geochronology of the Wrangell Arc: Spatial-temporal evolution of slab-edge magmatism along a flat-slab, subduction-transform transition, Alaska-Yukon. *Geosphere*, *18*(1), pp.19-48.

VanderLeest, R.A., Fosdick, J.C., Leonard, J.S. and Morgan, L.E., 2020. Detrital record of the late Oligocene–early Miocene mafic volcanic arc in the southern Patagonian Andes (~ 51° S) from single-clast geochronology and trace element geochemistry. *Journal of Geodynamics*, *138*, p.101751.

See Trop et al 2022 clipped below

This study reports a total of 3940 sand-sized DZ U-Pb, 2640 sand-sized DARL $^{40}Ar/^{39}Ar$, and 131 cobble-sized DARL $^{40}Ar/^{39}Ar$ dates from modern sediment from 22 major rivers and eight tributaries. Figure 4 summarizes the geology with the watersheds that were sampled. Figures 5–9 display relative age probability plots of modern river sediment samples. Figures 10–12 display composite probability plots of all samples. Figures 13–15 show the spatial distribution of <35 Ma detrital dates. The following sections summarize key age results from the overall study region followed by age patterns from five sub-regions.

We developed a procedure to limit the effects of alteration by degassing each sample at 0.5 W for 60 seconds, and the released gas was pumped off for time efficiency and hence increased throughput. The results have a single-grain and/or multi-grain precision of 1%. **A subset of 14 samples was selected for higher-precision ages and step-heated** from relatively low temperatures until reaching fusion temperatures using the 6 W argon-ion laser (Benowitz et al., 2014). For each step, isotopic ratios of Ar were determined, with a range of mean square of weighted deviates (MSWD) values of 0.0 –6.25 (Table S1).

See: Supplemental S6 and S9 from Trop et al., 2022

The majority of samples were analyzed as single-grain or multi-grain fusion analysis approach. We developed a procedure to limit the effects of alteration by degassing each sample at 0.5 watts for 60 seconds, and the released gas was not measured and pumped off for time efficiency and increased throughput. The results have a single-grain and/or multi-grain precision of 1%. Two different batches were dated from the Cheslina River sand sample (and results were combined); 1000 to 1200 micron sized grains yielded better analytical returns than 500 to 1000 micron sized grains owing to the dominance of young (<1 Ma) volcanic bedrock with limited radiogenic 40Ar in the watershed. **Samples selected for further geochronology analysis were step-heated from relatively low temperatures until reaching fusion temperatures** using the 6-watt argon-ion laser (Śliwiński et al., 2012; Benowitz et al., 2019). Refer to Repository Items DR7–11 for full 40Ar/39Ar analytical results.

From Benowitz GSA 2014 Poster: Where we step-heated sand grains before developing a specific DARL technique for the Wrangell Arc.