

We thank the reviewers for their time and thoughtful commentary. Their feedback will undoubtedly strengthen the manuscript. Our responses to their suggestions are recorded below in blue.

### **Reviewer #1**

The manuscript “Geochronological and Geochemical Effects of Zircon Chemical Abrasion: Insights from Single- Crystal Stepwise Dissolution Experiments” by McKanna and others is presenting step leaching U-Pb ID-TIMS data on three different zircon reference materials for two different temperatures and giving a recommendation for conditions to use by practitioners of chemical abrasion and an estimated minimum alpha dose at which Pb loss can occur. This study is a follow up from a previous study by the same authors on the same zircon samples that looked at structure of individual zircon grains of in response to chemical abrasion at different temperatures.

The conclusions from this study are largely confirming what has already been published about the chemical abrasion conditions and its consequences (Huyskens et al 2016, Widmann et al. 2019). All three studies concluded that leaching at 180C is not high enough for most samples to effectively remove all Pb loss from a sample and a higher temperature is needed in most cases. This study, however, is the only one that has detailed structural observations of zircons before and after the leaching procedure and it can tie them together with the U-Pb observations. This also leads to the possibility of estimated radiation damage of the leached zones, which could not be done in the two previous studies.

This is the main new angle of the study and these observations and discussions are unfortunately falling short in favour of documenting observations at length that have been made before. Thus, my recommendation is to focus a lot more on the new aspects of the study. To cut back on the repetitions reorganising of the results section would help. For example, for each sample early leachates are enriched in LREE. Thus keeping the standard format of first heading “geologic setting and sample description” with a sub heading of the different samples is advantageous. It also makes it easier to compare and contrast the different zircons analysed in this study.

We thank the reviewer for the suggestion. We will reorganize the results section in revisions to make it more concise and less repetitive.

The authors also looked for a tool to robustly identify zircons that have remaining Pb loss. Currently the identification of such analysis is very subjective and such a tool would make interpretations of a scattered dataset more robust. Unfortunately, neither of the indicators (U concentration,  $Pb^*/Pbc$ , or LREE enrichment) are effective tools for this task.

Yes, the result is unfortunate, but important to demonstrate.

One of the recommendations of this paper is to look for the amount of radiation damage and tailor the leaching conditions this way. However, no information is given on how to determine the radiation damage prior to dissolution. In this study the radiation dose was calculated based on the analysis of U and Th in the leachate and residue of the zircons, which means after already performing the time-consuming analyses. In the prior study Raman was used to estimate the alpha dose for the zircons. Any method to determine the alpha dose would need to have a high spatial resolution for the entire volume of the zircon grain, since it was documented in the previous paper that these zones do exist also in the interior of some zircon grains.

Determining alpha dose prior to dissolution is tricky. The reviewer is correct that the distribution of radiation damage in zircon is also typically heterogeneous. Unfortunately, there isn't a method currently available to derive high spatial resolution alpha dose values for the entire volume of a the zircon grain. Micro X-ray computed tomography ( $\mu$ CT) can distinguish between crystalline and strongly-damaged zircon in three dimensions as demonstrated in our companion paper. However, it is not currently possible to derive alpha dose directly from  $\mu$ CT data. Radiation damage can be estimated using Raman spectroscopy, and Raman mapping can be used to characterize internal variations in radiation damage. Alternatively, one can use Raman spot analyses guided by CL-imagery to bracket minimum and maximum alpha doses (i.e., measure Raman spectra from CL bright areas and CL dark areas). We acknowledge that this can be a time-consuming process and requires access to a Raman system. Another approach would be to measure the U and Th contents of zircon in situ prior to dissolution using laser ablation ICPMS. Paired with an appropriate damage accumulation interval based on the sample's geological history, alpha dose can be calculated. Again, time- and instrument-intensive.

Figure 16 can provide a quick-and-dirty tool for estimating alpha dose in zircon crystals without a lot of information. Given a rough estimate for a sample's damage accumulation interval based on the sample's geology (such as an approximate crystallization age or cooling age from thermochronology data in the literature), one can calculate alpha dose for a range of possible U concentrations. We can add these points to the discussion in revisions. Ultimately, this study and other chemical abrasion studies suggest that leaching at 210 °C is effective for a wide-range of alpha doses.

Please provide all the calculated numbers that are used in the plots like alpha dose at Pb loss or LREE-I. – We can provide this information in a table in the Supplementary Materials.

A description is needed how the amount of dissolved material for the calculation of alpha dose was determined. – The only variables that the alpha dose calculation requires is the concentration of U and Th in the leachate/residue and time – i.e., the damage accumulation interval. The Th concentration was determined by ICPMS, and the U concentration is calculated from the Th concentration and the Th/U ratio determined by TIMS. The mass or volume of dissolved material is not needed.

Fig 12: could you distinguish between the different temperatures maybe using open vs filled symbols? – We can make the suggested change in revisions.

Fig 15: It looks like the samples leached at 210C overall had lower radiation damage compared to the ones used in the 180C experiment in this figure and this would need an explanation. My guess is that it has something to do with the fraction of material in each of the dissolution steps, but this is confusing at first. For me, the figures about the alpha dose are the most important in this study and the groupings in Fig 14 and 15 are so broad that they could be masking interesting details. A plot including alpha dose vs discordance for example on an individual analysis basis could be interesting.

The reviewer is correct that the apparent differences in alpha dose between the samples leached at 180 °C and 210 °C in Figure 15 reflect the fraction of material dissolved in each step. 180 °C leachates reflect the dissolution of small volumes of high-U zones. Whereas, leaching at 210 °C dissolves a larger volume of material including both high-U and medium-U zones, causing the average U concentration (and alpha dose) to be lower for 210 °C L1 leachates than for 180 °C L1 leachates. We will include discussion about this in revisions.

In general, we find considering the data in aggregate to be meaningful for deriving overarching trends. Alpha does vs. discordance is not a useful metric for evaluating the AS3 and the KR18-04 datasets, since the data spread along the concordia line (Fig. 1 and Fig. 9).

Line 31: "However, Ultimately,..." – We will fix this typo.

Line 49: "since the trajectory of Pb-loss follows Concordia" I think it should be the concordia. – We will fix this typo.

Lines 50-51: "the precision of 207Pb/235U dates is also lower than corresponding 238U/206Pb dates due to the shorter radioactive half-life of 235U and lower isotopic abundance (Corfu, 2013; Schoene, 2014)." The precision of the 207Pb/238U dates is lower not because of the shorter half-life. It is only lower due to the lower abundance, which in turn is due to the shorter half-life. – The reviewer is correct. We will add this to the revisions.

Line 59: "annealing zircon samples prior to leaching helps to minimize the unwanted isotopic fractionation effects that plagued earlier leaching attempts" The main improvement is reducing elemental fractionation in the leaching steps. – The reviewer is correct. We will add this to the revisions.

Line 191: "included and altered grains..." Grains with inclusions – We will correct this in revisions.

Line 210: "which had been redone as part of (Schoene et al., 2006)." Brackets are in the wrong place – We will correct this in revisions.

Line 522: frantzing should not be a verb – We will change this in revisions.