Authors' Comments

We would like to thank the editor, reviewers, and community members for their commentary on our manuscript, "Chemical abrasion: The mechanics of zircon dissolution" that will ultimately help to strengthen this contribution. We respond to each of their comments below. If given the opportunity to submit a revised version of this manuscript, some of the major changes we would make in response to feedback include:

1) Focus on the heart of the manuscript more strongly – *textural evidence* for the mechanics of zircon dissolution. Many comments reference the lack of geochemical and geochronological data. We would like to emphasize that a complimentary manuscript that focuses on the geochemical and geochronological evolution of chemically abraded samples is currently in preparation. We would shorten and refocus Sections 4.2 (Implications for ID-TIMS U-Pb Geochronology) and Section 5 (Conclusion) to emphasize that the effectiveness of any chemical abrasion protocol for ID-TIMS U-Pb geochronology will ultimately be sample-dependent and reflect a sample's radiation damage and inclusion content and distribution. We would refrain from prescribing any specific chemical abrasion protocol, since no geochronological and geochemical data are presented in the current work.
2) We would also remove Section 4.3 (Implications for radiation damage annealing models) since it is tangential to the discussion and in need of additional supporting data.
3) Streamline the writing to eliminate wordy text and shorten the manuscript length. We would add two small tables that more succinctly summarize Raman data and basic sample descriptions.

We address each reviewer's specific comments below. Reviewer comments are in black text, and our responses are in blue text.

Reviewer 1 – Fernando Corfu

Overall the paper is well prepared and structured. I find, however, that the present version could be improved by a good weeding of unnecessary words and sentences. Some expressions that the authors like to use frequently should be reconsidered. One example is 'compositional zone'. I have made some suggestions in the file and discuss specific points below.

We can tighten the language in the manuscript including our use of the phrase "compositional zone." We address more specific wording suggestions below.

Although the results are very relevant for zircon U-Pb geochronology, the studied zircons have not themselves been dated, and so the specific data do not have a direct connection with the U-Pb behavior of the samples. Consequently, the discussion of implications for U-Pb dating presented in the paper is very generalized and in part trivial. I suggest to concentrate on the main substance of the study and avoid meandering off in inefficient discussions…. Section '4.2.1 Zircon U-Pb ages' lines 648 – 726: I find the discussion on this topic very generalized and superfluous. The experiments in this paper demonstrate the great variability between zircon of different geneses, compositions and ages. Yet many of the reflections made here focus on some idealized magmatic zircon in young systems. Because of such a simplification the discussion is trivialized and almost meaningless. Clearly, the lessons from the present experiments must be considered separately for each set of zircon used for geochronology. I recommend removing this section, it just detracts from the paper.

We thank this reviewer for these comments. In a revised contribution we would refocus the manuscript on the *textural evidence* for the mechanics of zircon dissolution more strongly since U-Pb data are not included in this piece. We would shorten the U-Pb discussion section significantly to avoid over generalization and more strongly emphasize that the effectiveness of any chemical abrasion protocol will ultimately be sample-dependent. However, since many ID-TIMS zircon studies do target magmatic zircon, we feel that a brief discussion about implications for the dissolution of an idealized concentrically zoned, magmatic grain is worthwhile. Since no other study in the literature to date has systematically documented how effective chemical abrasion is at dissolving inclusions, the section on inclusions is also important to eliminate.

Line 64; '… poorly understood, and several outstanding questions remain. Do most zircon crystals predominantly dissolve from rim to core?' Everyone who has done chemical abrasion will have noted very rapidly that zoned and metamict zircons do not dissolve that way. That can hardly be described as a poorly understood fundamental question.

We can rephrase our study motivation in the introduction and eliminated the phrases that the reviewer cites. The reviewer is correct that many practitioners of chemical abrasion are aware that many zircon crystals dissolve heterogeneously. However, apart from a small number of SE images of chemically abraded zircon crystals that indicate that acid sometimes reaches grain interiors (Mundil et al., 2004; Mattinson, 2005; 2011), the mechanics of zircon dissolution are poorly documented in the literature. In the new

wording, we would emphasize that no study to date has systematically documented zircon dissolution textures given a range of zircon types and leaching conditions, nor leveraged such findings to gain a mechanistic understanding of the microstructural processes that occur during partial dissolution in HF.

Fig.2: The explanations are unclear. Are the grains shown in each of the four panels representing each of the sample? If yes, why does the second panel contain zircon from two different samples (blue and yellow circles). If not, what is the distinction? Overall I find this figure quite useless, not even as a decoration.

We would keep this figure at the suggestion of other reviewers in part to highlight the color change that occurs during chemical abrasion. However, we can update the labeling to better illustrate how samples are distributed in b).

Fig.3: Not sure about using the term 'metamict' once zircon has been annealed. Can be confusing.

We can change the wording from a 'metamict' rim to a 'high-damage, CL black' rim.

209: 'typical magmatic growth patterns' It is true that many magmatic zircons have oscillatory zoning, but the same pattern can in part be seen in metamorphic zircon. Better to use descriptive terms.

We can change the wording to more descriptive terms.

287: ' This could imply that increasing the duration of the leaching step results in a more crystalline zircon residue due to the progressive dissolution of higher damage domains.' Why the 'could'? It seems to be the most logical explanation. And then the following sentence; 'We note, however, that only a small number of AS3 crystals survived 12 h of chemical abrasion, and only a small number of Raman analyses were made. We recommend further study to better evaluate this possibility.' Sounds rather trivial.

We can remove the word 'could' and the lines that the reviewer suggests.

316: 'As evidenced by our SE images and discussed further below, μCT does not capture radiation damage zoning that does not result in a strong density contrast such as variations in radiation damage below the ~1×1018 α/g threshold.' Suggest rephrasing to avoid the double negatives.

We can rephrased this sentence to eliminate the double negative.

Fig. 9, caption: 'interior compositional zone'. Compositional zone? As opposed to what?

We eliminated the use of the term 'compositional zone.'

367: ' All observed dumbbells are oriented parallel to the grain's c-axis'. That is a surprising statement. Looking at the figures I would have assumed that they are all normal to the elongation of the crystal (= c-axis). Please elaborate to avoid confusion

The reviewer is correct. Our original description is inaccurate. We can rephrase this line accordingly.

Fig 13 caption: 'The yellow arrow highlights the grain's shell-like appearance because of significant dissolution in the grain's interior.' I see a highly resorbed grain, not much left of the shell.

We feel that our description of the grain's "shell-like" appearance is still appropriate.

'images of dog-chewed zircon residues ' maybe a bit too colloquial?

We can eliminate the colloquial wording from this line.

419: 'In a visual game of connect-the-dots …' ??

We can rephrase this line.

422: 'We see dumbbell-like fracture patterns again in sample Zr36 (Fig. 13b-III) where crosscutting fractures connect different oscillatory zones removed by dissolution to one another and to the grain surface'. Rather convolute sentence, hard to understand. Please rephrase.

We can rephrase this line.

483: '… The long axes of deep, octahedral etch pits on (100) align with the crystal's c-axis…' I wonder why they are called octahedral. Those in Fig 16a look prismatic to me.

This line actually refers to Fig. 17. The figure reference number was inadvertently mislabeled. We can change the "octahedral" description to "prismatic."

Fig. 19, caption: ' Projection on (100) looking down the a-axis. The c-axis is vertical to the page, and the a2-axis is horizontal' Confusing: should it not be: 'Projection on (100)

looking down the c-axis' ? The same for (b): if the pane is 001 then the view must be parallel to a? Or not?

We can simplify the figure caption; however, the description "Projection on (100) looking down the a-axis is correct, since (100) is orthogonal to the a-axis.

640: '… suggest that a crystal's bulk radiation damage also plays an important role.' That seems a rather trivial discovery. What else could one expect?

We feel it is necessary to describe our findings even if the result is the expected outcome.

641: 'Crystal morphology plays a lesser role in that crystals with very high aspect ratios dissolve more slowly than more equant grains' ??? That is not apparent in Fig. 18e, and would seem to contradict the higher solubility along the c-axis than along a.

We can add a trendline to Fig. 18e to highlight the trend to which we refer. While there is higher solubility along the c-axis, the longer the c-axis is, the longer it will take the crystal to dissolve.

778: '…chemically abraded residues are more crystalline than their annealed counter parts…': Isn't that the logical relationships since CA removed the less crystalline domains? And: '… radiation damage is annealed hydrothermally during HF leaching…'. Speculative?

It is the logical relationship that chemically abraded residues are more crystalline than their annealed counterparts. We can more strongly emphasize this point.

In revisions, we would remove the section "Implications for Radiation Damage Annealing Models" which discusses hydrothermal annealing of radiation damage. There are a few studies that report structural recovering during hydrothermal treatment (Rizvanova et al. 2000; Geisler et al. 2001b, 2002, 2003) that we cite, and this remains our preferred interpretation to explain some characteristics of our Raman data (i.e. changes in the relationship between the two Raman bands post-leaching). However, we recognize that this discussion needs additional data to support it and detracts from the main purpose of this paper.

867: 'Increasing the leaching temperature from 180 °C to 210 °C or increasing the leaching duration leads to the development of more extensive dissolution networks in higher damage grains ' This is a rather trivial conclusion. Something CA-users did not observe before? …. 870: 'More crystalline zircon samples lack fracturing related to

radiation damage zoning.' Another trivial statement. The conclusions should focus on the important aspects of the research, not on trivialities.

Again, while these conclusions may be the expected outcome, we feel it is still important describe these findings in the conclusion.