

## *Interactive comment on* "Extreme isotopologue disequilibrium in molecular SIMS species during SHRIMP geochronology" *by* C. W. Magee Jr. et al.

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## by C. W. Magee, on behalf of myself, T. Mernagh, and M. Danisik

The authors would like to thank the referees for their time and thoughtful reflections on our manuscript gi-2016-07. As a general rule, we find their comments useful, and on the whole we will accept them and incorporate them into the manuscript. Where there are exceptions to this, or where further detail is needed, we will expound below:

Reviewer 2 comment P1, L10: "It's not clear what this paper is about as you start to read the abstract. It needs a couple of sentences of introductory material concerning the issues to be covered." An expansion to the abstract and introduction will be provided, explaining how the U-Pb calibration is the limiting factor on SIMS geochronology,

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and how this study helps explain the mechanism of this calibration with an aim towards finding improvements.

L18, "You could add xenotime in here for alarming matrix effects.": While xenotime does exhibit matrix effects, we did not look at it in this study, so we will not include it.

P3 L18. "Not sure this is quite right but it doesn't make a lot of difference. I believe the extraction voltage power supply (2.5 kV) floats on the 10 kV column grnd/sample potential. The offset potential is around say 700 V, so the intermediate extraction sits at 9.3 kV relative to real ground in this example. So there's a 700 V bias from the sample to the IE plate.": The text is correct, but poorly phrased. We will rewrite for clarity.

P6L6 "Ratios are then typically multiplied by 100. Why "typically"? Should be consistent. In abstract it is permil. In figures it is left as the absolute ratio." We will edit the abstract, main text, and figures so that they are all consistent.

L19, L21 "Scan uraninite over the region of interest to sort out the interferences. If you suspect UO2H2 then it should be supported by a higher UO2H. 235U species, 17O species?" and "Suggest not mentioning the 18O experiments here, I got confused. Put it in as part of Expt B description.":

The point of this section is that the interferences are minor enough that we don't need to identify them or take them into account, as they will be negligible once the isotopically labeled primary beam is used.. We will rewrite the paragraph to make that clear.

P7L22. "Did you retune down hole? Some of this variability can be recovered by secondary ion retuning. Also there may be some issue concerning different angular emission between the different species. Also, different energy distributions if there is any target charging. Was an energy slit included?": We will add details of analytical procedure (no retuning, no energy slit)

L30 "Monazite anomalies - okay so I suspect there are some molecular interferences in the monazite spectrum. Some long range scans might have helped identify them.

"Unlabeled monazite analyses"; what does this mean?": We will clarify that "labeled" refers to the isotopic composition of the primary beam.

P8P25: "Abrupt start to Discussion. What is expected from the calibration and the distribution of the isotopologues?": We will make the discussion less abrupt by leading in with What we know and which experiments seem to make sense. Once we explain the isotopologue behaviour of the well behaved calibration, we can then discuss what may be making the poorly behaved calibration fail.

P9L5 "Unclear what this is about. Where did this "Molecular ion escape model" come from? Wouldn't speciation be a bigger issue (neutral vs ion) or angular/energy distributions? What does it mean "escape without reacting"?": We will introduce this model as a testable hypothesis which can be examined with the data at hand (as opposed to hard-to-constrain, but more likely models). We will explain it enough to include comment L9.

L31. "the tails on UO2 are not documented. I've generally seen the poorest peak shape on U+, which appears to be energy related. The form of these tails could be indicative as to the source.": We are referring to Energy tails, not peak shape tails here. We can include a figure to demonstrate.

P10L1: ""agree" with what? each other? "higher potential valence states of uranium" - where did this come from?": We will rephrase this to state that we mean "behave the same way."

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