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To: George Gehrels, University of Arizona

Dear Prof. Gehrels,

First of all, I would to apologise for the late decision on your manuscript. I was only appointed as handling editor after two of my colleagues became unavailable. I have now had a chance to read your paper, as well as the two reviews and your response to them. I have decided that your manuscript is suitable for publication in *Geochronology* after moderate to major revisions. In the following paragraphs, I will first share my thoughts on the two reviews. Then, I will add some further comments of my own, which mostly relate to the statistical aspects of your work.

Review by Dr. Glenn Sharman

1. Dr. Sharman gave a positive review that raises one major point and several minor ones. The major concern was about the lack of evidence to support the inference that near depositional age zircon is air-fall in origin and older zircon is recycled. You responded to this comment by saying that you are:

   > unable to provide reliable information about morphology of the young grains, as most were plucked out from the mounts and dissolved for CA-TIMS geochronology. We tried to do this analysis with BSE images of the grains (before analysis), but the size/shape of the grains in the images has little bearing on the true size/shape of the grains. This because the mounts were polished down just a little so as to retain more material for the CA-TIMS analyses.

   I would have thought that even a two-dimensional cross section through a c-axis parallel zircon grain would reveal whether the grain is prismatic or not. If you have stored the BSE images, then I would encourage you to include them in an online data respository.
2. The reviewer also has a question about the change in scale of your age distributions. An alternative way to bump the height of the pre-240Ma age component would be to plot the age distributions on a logarithmic time scale. Furthermore, if you replace your Probability Density Plots (PDPs) by Kernel Density Estimates (KDEs), then you can tweak their bandwidth to produce the most informative result. As you know, cumulative distributions are also useful for data visualistion. They, too, can be plotted on a logarithmic time axis.

Review by Dr. Jahandar Ramezani

Dr. Ramezani wrote a more critical review that raises a number of issues about the stratigraphy as well as the U-Pb data.

1. Stratigraphy: Dr Ramenazi is concerned that the observed drift between your MDAs and the depositional ages is due to misidentification of the stratigraphic positions in the CPCP core. In your response, you wrote that your paper does not aim to

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present an age model, and does not claim to estimate accurate MDAs either. I am a bit confused, because the paper does seem to me like an attempt to calibrate the depositional history of the CPCP core in absolute time. If your paper has a different objective, then please state more clearly what the purpose of the study actually is. I apologise if I am missing something obvious here.

2. U-Pb geochronology: Dr. Ramenazi is concerned that the LA-ICP-MS results may be affected by Pb-loss, which would invalidate their use as *maximum* depositional ages. In your response, you write that:

> This manuscript goes to great length to document that Pb loss is a significant factor for many of the grains analyzed. We show this internally with the Uconc-age tests described above. We also document this by comparison of our ages with the CA-TIMS data from the same grains (Appendix 2). Indeed, Pb loss is an important factor for many of our analyses! But the assertion that LA-ICPMS max depo ages are younger than the CA-TIMS ages of Ramezani et al. (2011) and Atchley et al. (2013) due to Pb loss is not supported by the fact that most of the reported LA-ICPMS MDA's are older (not younger) than the equivalent MDA's reported by R+2011 and A+ 2013!!

Appendix 2 clearly shows that the LA-ICP-MS data are consistently 5-10 Ma younger than the CA-TIMS ages. To me this confirms the reviewer's concerns. The fact that the ad-hoc MDA estimates for the youngest LA-ICP-MS peak (which are shown as circles in Appendix 2) are consistently older than the CA-TIMS estimates (which are shown as red bars in Appendix 2) is a result of comparing datasets of different size. Your LA-ICP-MS based MDA estimate uses more grains than the CA-TIMS estimate, making the comparison between the two estimates biased. This problem is diagnostic of a fundamental flaw in three of the four MDA estimation algorithms that are proposed in the manuscript. I will discuss this in more detail below.

Further comments

1. The paper uses four different heuristic MDA estimation algorithms. Three of these methods are problematic, because they drift to ever younger ages with increasing sample size.
   (a) Age of the youngest peak on a probability density plot (PDP): PDPs have no statistical basis, and any quantitative information derived from them is of dubious statistical significance. If you were to analyse one million grains of zircon, then the youngest age cluster on a PDP would likely be younger than the actual depositional age.
   (b) Weighted Mean age and uncertainty of the youngest cluster: Same problem. Any heuristic method that is based on p-values is problematic because p-values are a sensitive function of sample size. The larger the sample size, the greater the likelihood that the χ^2-test identifies spurious peaks.
   (c) Maximum Likelihood age and uncertainty. See Figure 6.3 of Vermeesch (2018b) for an example of how multimodal unmixing models suffer from the same problem as methods a. and b.

   The sample size dependency is actually reported in the paper ("Ironically, the more

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grains analyzed, the greater the inaccuracy of [the] youngest age!"). I do not understand why these broken methods are still used in the paper and would advocate that they are removed. In statistics, it is desirable for estimates to asymptotically converge to the truth with increasing sample size. Only the Tuffzirc age model may have this property. An alternative would be the parametric minimum age model of Galbraith and Laslett (1993). But neither of these techniques is immune to the Pb-loss problem.

2. The paper frequently uses two ad-hoc dissimilarity measures called 'Likeness' and 'Cross-correlation Coefficient' (CCC). These quantities are both derived from PDPs and are flawed for reasons that are given in detail by Vermeesch (2018a). Please remove these from the paper and replace them with bona fide statistical dissimilarity measures such as the Kolmogorov-Smirnov statistic. Of course, if you can present a statistically valid argument against my objections to Likeness and CCC, then I would be happy to change my mind.

3. Is Figure 10 a two-dimensional PDP or KDE? I think that this diagram would be more effective as a contour plot, or as a simple scatter plot. The three-dimensional effect adds no useful information.

In summary, I think that your paper is a great illustration of the need to combine LA-ICP-MS and CA-TIMS. LA-ICP-MS is a powerful screening tool that can be used to infer the provenance of siliciclastic sediments, and to identify the youngest grains in a detrital zircon population. LA-ICP-MS can also yield decent first order MDA estimates. But for chronostratigraphy, the precision and accuracy of CA-TIMS is still needed.

*References

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Sincerely yours,

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