

Interactive comment on “Percent-level production of ^{40}Ar by an overlooked mode of ^{40}K decay” by Jack Carter et al.

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We would like to thank Anonymous Referee #1 (hereafter referred to as AR1) for their timely and detailed comments.

Overall, we disagree very little with the substance of AR1's review. However, we believe that there is a difference in perspective.

For better or for worse, the geochronological community compiles, calculates, and uses radioisotopic decay constants on its own, independent of the physics community. This is evidenced by the overwhelming use of the K decay constants published in the convention by Steiger and Jager (1977), Min et al. (2000), and to a lesser extent Renne et al. (2010) over any of the compilations favored by the

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physics community. Indeed, the IUPAC and IUGS have commissioned a committee that publishes critical reviews of decay constants specifically for the chemical and geological community, that largely ignore the physics databases (Villa et al., 2015; Villa et al., 2016; Villa et al., in press; <https://doi.org/10.1016/j.gca.2015.05.025>; <http://dx.doi.org/10.1016/j.gca.2015.10.011>; <https://doi.org/10.1016/j.gca.2020.06.022>).

This manuscript is specifically targeted at geochronologists, who follow the conventions of geochronology, not of the physics literature. We agree with AR1 that our results are not *sensu stricto* new – a major point of the manuscript is that the original personal communication in 1962 is essentially correct. However, geochronologists do not typically follow the physics literature, are likely not to be well-trained in the type of nuclear physics that would recognize as obvious that there must be a complementary ground-state decay mode and are not qualified to judge the approximate magnitude. This manuscript makes this clear for geochronologists. That this is important should be clear from the way that decay constants are used by this community, and the literature from which they draw from. Essentially all $^{40}\text{Ar}/^{39}\text{Ar}$ and K-Ar geochronology draws on one of the three publications (Beckinsale and Gale 1969; Min et al., 2000; Renne et al., 2010/2011). Among these three, Min et al. and Renne et al. have the strongest underlying analysis and uncertainty structure (Min et al. effectively repeat the analysis embedded in Beckinsale and Gale, but more critically), and Min et al. is currently more popular than Renne et al. The confusion in the geochronological literature on the ground-state electron capture decay mode can be clearly traced to Min et al. That this is only a single paper does not weaken the argument for our manuscript, it strengthens it.

We strongly believe that there is a good place in the geochronological literature for our analysis; this is also supported by the comments in this regard from Anonymous Referee #2 (hereafter AR2). However, it is clear from the criticisms and comments of AR1 that some of our comments and data presentation may be misleading – we will

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draft a revised manuscript that takes this feedback into account. These are responded to specifically below.

Response to specific points:

1) The “percent-level” change is percent of all decays to ^{40}Ar , but we thank AR1 for pointing out that it may be misinterpreted and are happy to change the title of the manuscript. We disagree that it is not “overlooked”, inasmuch as it is overlooked by the geochronological community, but we can also clarify this in a revised manuscript.

2) We state in the manuscript that the ground state branch is included in the ENSDF and DDEP evaluations. The fact that it is included in decay constant evaluations outside of those used by the geochronological community is important because it strengthens the argument that in favor of the presence of a EC ground state decay mode and should be considered (and is why they are discussed in our manuscript). However, simply because these evaluations exist, does not mean that our manuscript is not useful to the geochronological community: the ENSDF and DDEP evaluations are not used for Ar-Ar or K-Ar dating.

The reference to “many subsequent workers” is somewhat confusingly written, and we thank AR1 for highlighting this. We are referring mainly to the many geochronological works that ignore this decay mode, but we do highlight some physics literature on line 82-83, and are also referring to the controversy over the DAMA experiment backgrounds, discussed on line 280. We will reframe this statement to be more clear and refer to specific literature.

3) We thank the reviewer for bringing this to our attention and the spelling will be corrected.

4) We are pleased that AR1 reads this section as an explanation of textbook knowledge. The audience for our manuscript is geochronologists, who are typically not versed in nuclear physics, so we have tried to provide a straightforward explanation

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of the concepts that underpin this decay mode. The feedback from AR2 suggests that this was in fact useful for the geochronologist audience. We did not intend to represent our calculations as state of the art and will clarify this in the revised text by indicating that Mougeot (2018) provides the most robust calculation. Our purpose in providing additional calculations is to demonstrate to an audience of skeptical geochronologists (who may be skeptical of a decay constant that is derived entirely by calculation) that the derived quantity is relatively robust to differences in the way that it is estimated. For this reason we also include the much older Fireman (1949) calculation, and the extremely crude LogFT extrapolation, neither of which would be considered state of the art.

5) We thank AR1 for pointing out these inconsistencies and will fix them in a revised draft.

6) We thank AR1 for this observation. We have not considered that the single beta+ experiment might be erroneous, and we have taken the experiments at face value. If this experimental result is incorrect, all the physics literature that we and AR1 cite that includes the ground state decay mode is wrong. We rely on the Engelkemier et al. 1962 experimental observation of the positron decay, in this work the authors include a discussion of the pair production from 1462keV gamma in their experiment, calculating this as 55-60% of the total positron detection rate. The positrons produced from pair production would be mono-energetic at 440keV whereas the positron energy spectrum has an EMAX of 491 keV. We will include amended discussion points clarifying these points more clearly. It is outside of the scope of this manuscript to repeat the experiment of Engelkemier et al. 1962, but we will gladly add the caveat in the revised manuscript that this hinges on a single measurement of a low probability decay mode.

7) We agree with AR1 that this is clumsily worded. The point we were trying to make is that the uncertainty budget for the estimate in Mougeot (2018) is not clearly articulated – it is not clear if the uncertainty presented in that paper is solely propagated from the Q-value as an intermediate precision, or whether it takes into account other sources of

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uncertainty.

8) We take this criticism seriously from AR1. Ultimately, our goal with this manuscript is to provide a simple physics background and argument that will be straightforward to digest by working geochronologists, so that they understand the likely magnitude of a ground state E.C. decay of ^{40}K . In our opinion, it is unlikely that a single estimate – even as cutting edge as possible – is likely to provide a convincing argument to our audience. For this reason, we have provided a range of estimates, using different techniques – yes, some outdated – that all point to a $\text{EC}_{\text{ground}}/\beta^+$ of ~ 200 .

Most of the manuscript is dedicated to harnessing a range of evidence that it is likely that the decay mode exists and has a $\text{EC}_{\text{ground}}/\beta^+$ of approximately 200. Having done this, our next goal was to then communicate the effect this has on the decay rate and branching ratio used in $^{40}\text{Ar}/^{39}\text{Ar}$ geochronology. For this, it's necessary to use an estimate, or several estimates.

We wanted to provide a reader, who for this journal is likely to be a geochronologist with skepticism that any one estimate will be unbiased or correct with a sense of what the possible range of effects is. The most straightforward way to do this is to group several reasonable estimates together and propagate that variance onto the existing decay rates used by the geochronological community (the values from Min et al.). The result is that the additional decay is negligible for most geochronological inference because it ends up being smaller than other uncertainties. Given that any reasonable $\text{EC}_{\text{ground}}/\beta^+$ produces a negligible effect, we are not concerned that “averaging a number of different calculations” yields a result that was metrologically unsound, as AR1 argues.

It appears that the way we attempted to convey the overall magnitude of the effect of the non-zero $\text{EC}_{\text{ground}}/\beta^+$ gives the wrong impression. We intend on revising the manuscript so that the focus is on the effect of range in calculated $\text{EC}_{\text{ground}}/\beta^+$, and we will highlight and emphasize the Mougéot (2018) calculation as the preferred esti-

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mate.

9) We agree that ^{22}Na is not a perfect analog, but it is probably the best choice that has a tractable calculation and enough experimental data that the calculation can be reliably verified. In a revised manuscript we will clarify that this is not strictly analogous.

10) We regret overlooking Nähle et al. (2008), and will include this determination in a revised manuscript, thanks to AR1 for bringing this to our attention. We had not intended to present other calculations for ^{22}Na , but on the suggestion of AR1, we would be happy to include them.

11) In the manuscript, we meant it to be understood as “easier to measure than the ^{40}K ground state decay”, rather than imply that the ^{22}Na decay is a straightforward measurement. We thank AR1 for pointing this out, and we will rephrase this section in a revised manuscript.

12) We will correct this statement in the revised manuscript.

13) We thank AR1 for identifying this confusing statement, and will correct it in a revised manuscript.

14) We thank AR1 for identifying this statement and we agree that by this wording is unclear. We will clarify this in a revised manuscript that will state that with a long enough counting period it will be possible to discern those x-rays that are untagged by the 1.46 MeV gamma and those that are tagged effectively observing the electron-capture to ground state decay mode.

15) We will correct this reference in a revised manuscript

16) ISO compliance is typically associated with certification and recertification of processes or products, and it is not clear how the ISO, or a particular ISO standard, is relevant to this manuscript. Regarding the Guide to Uncertainty in Measurement (GUM; JCGM 100:2008), the estimation of uncertainties follows what that guide refers to as the “law of propagation of uncertainty” described, for example, in section E.3.1

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from the GUM. Our manuscript follows some geochemical and geochronological conventions that depart from GUM recommendations, such as notation and reference to “coverage factors” because this is the style of the journal and the community. These “formal errors” are largely an editorial decision, and we are happy to take direction from the editorial staff of Geochronology on this matter.

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