

Community comment on ‘Broken $^{206}\text{Pb}/^{238}\text{U}$ carbonate chronometers and $^{207}\text{Pb}/^{235}\text{U}$ fixes’ by Vermeesch et al.

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This manuscript presents a new methodology for calculating disequilibrium-corrected U–Pb ages and age uncertainties for young carbonate samples using a Bayesian statistics approach. It also offers a number of criticisms of existing $^{206}\text{Pb}/^{238}\text{U}$ -based methods for dating young carbonates, going as far as to suggest that ‘...one could argue that the $^{206}\text{Pb}/^{238}\text{U}$ method is of limited use to carbonate U–Pb geochronology’. Following this, the manuscript restates arguments presented in previous publications recommending use of $^{207}\text{Pb}/^{235}\text{U}$ -based methods where accurate correction for initial $[34/38]$ ¹ disequilibrium via a measured $[34/38]$ value is not possible.

While their Bayesian approach to calculating U–Pb ages for carbonate samples lying just outside the bounds of analytically resolvable disequilibrium is a welcome contribution, there are some inaccuracies in the arguments provided against use of $^{206}\text{Pb}/^{238}\text{U}$ -based methods more generally. Furthermore, the authors fail to properly acknowledge and cite previous work undertaken on some key issues discussed in the manuscript, including work which advocates very similar ideas. This community comment addresses some of these issues.

Monte Carlo approach

Line 90 states incorrectly that `Isoplot` (Ludwig, 2003b) implements a Monte Carlo approach for estimating uncertainties in disequilibrium-corrected U–Pb ages that incorporate a measured $[34/38]$ value, before going on to criticise this approach. This is both factually incorrect and somewhat unfair to Ludwig. `Isoplot` includes functions for calculating $^{206}\text{Pb}^*/^{238}\text{U}$ and $^{207}\text{Pb}^*/^{235}\text{U}$ ratios (where the * superscript denotes radiogenic Pb) as a function of age and initial intermediate ‘daughter’ activity ratios. These functions, in combination with the ^{234}U -age equation, can be used to calculate disequilibrium-corrected U–Pb ages using a measured $[34/38]$ value and estimate uncertainties via Monte Carlo simulation as part of a spreadsheet-based approach (Woodhead et al., 2006). However, this functionality is not part of `Isoplot`, nor is it advocated or discussed in the `Isoplot` manual, which is very clear in outlining the program’s functionality.

Section 3 also provides a misleading description of the Monte Carlo approach implemented in `DQPB`. Prior to beginning the Monte Carlo simulation, `DQPB` checks that the measured $[34/38]$ value provided by the user is statistically distinguishable from secular equilibrium. By default, the $[34/38]$ value must be 2- σ removed from secular equilibrium, although this cut-off value can be adjusted. Where the measured $[34/38]$ value fails this test, a warning is provided stating that the results of the Monte Carlo simulation may be inaccurate and should not proceed. The user is then encouraged to cancel the simulation. This is outlined in section 5.1 of Pollard et al. (2023).

Line 105 states that the quasi Monte Carlo approach of Vermeesch et al. is ‘...faster and produces deterministic results that do not depend on the seed of a random number generator.’ In reality the difference in speed is of no practical significance on modern computers and the indeterminate nature of Monte Carlo simulation is trivial provided a sufficient number of trials are performed. There is also no reason to assume that deterministic uncertainty estimates are inherently better than non-deterministic ones: at the end of the day they are both just estimates, and neither is perfectly accurate. A more

¹The notation of Vermeesch et al. is used throughout.

substantial problem, however, is that the quasi Monte Carlo approach appears to introduce spurious noise in some cases. For example, the arbitrary fluctuations in the probability density curves in Figure 2c and 5d are hardly realistic. This potential drawback should be discussed along with the apparent advantages of this approach.

The matrix exponential approach

There are numerous methods that can be used to solve the system of linear first-order differential equations describing the U–Pb radioactive decay series (or part thereof). These include:

- The original solution of Bateman (1910), which was applied directly to the U–Pb decay series by Ludwig (1977)².
- Methods that employ the Laplace transform explicitly (e.g. Catchen, 1984; Pressyanov, 2002; Bourdon et al., 2003).
- The matrix exponential approach, which has been used in nuclear science and engineering for over 50 years (e.g. Bell, 1973), and has been applied previously to the U–Pb decay series by Albarède (1995)³.

These are all mathematically valid approaches, and, as such, should yield identical results for the same inputs (at least, for the simplified form of the U–Pb decay series considered in geochronology where numerical errors are a minor concern). However, this pre-existing literature is not discussed at all in the manuscript and the matrix exponential approach is presented as if it is an entirely novel idea. Given the well-established mathematical credentials of the authors (e.g. Mclean, 2014), it is certainly possible that they derived the matrix exponential equations independently and without knowledge of previous work on this topic. Nevertheless, it is typical practice in academic writing to cite previous work that presents the same or very similar ideas. Some clarification is required here.

This section also contains no substantial discussion of why the matrix exponential approach is chosen here or how it improves on other more widely used pre-existing solutions (e.g., Ludwig, 1977). Perhaps the argument is that the matrix exponential approach is more ‘elegant’ than other approaches. However, this is rather subjective and debatable given that the matrix exponential approach, at least as implemented in `IsoplotR`, requires more computations than simpler approaches to obtain the same result (at least for age calculations of the type discussed in the manuscript). There may be some advantages in terms of flexibility, however, if so, these should be outlined explicitly.

Maximum range of measurable [²³⁴U/²³⁸U] disequilibrium

Section 5 discusses the maximum range at which measured [34/38] is analytically resolvable from equilibrium. Two choices are made here that lead to an overly pessimistic assessment of this maximum range. The first is the adoption of an analytical uncertainty of 0.004 (2σ)⁴, i.e. 4‰, for measured [34/38] values. Analytical uncertainties for [34/38] measurements that are made using modern MC-ICP-MS protocols with the ²³⁴U⁺ beam collected using an ion counter typically range between 1–3 ‰ (2σ) (e.g. Hellstrom, 2003; Shen et al., 2012; Cheng et al., 2013). However, it is possible to obtain analytical

² Note there is a typo in Equation (4) of Ludwig (1977). The $e^{\lambda_{234}t}$ term should read $e^{\lambda_{238}t}$.

³ Albarède does not consider in-growth of stable ²⁰⁶Pb but the same mathematical approach can be adapted for calculations including radiogenic Pb.

⁴ Strictly speaking, Vermeesch et al. adopt a relative analytical uncertainty of 0.002 (1σ). However, given measured values are always close to one in this context, this is essentially the same as adopting an absolute uncertainty of 0.004 (2σ).

uncertainties that are almost an order or magnitude lower than this (i.e. 0.2–0.3 ‰) using an all-Faraday cup analytical protocol (e.g. Andersen et al., 2004; Cheng et al., 2013, 2016; Kerber et al., 2023). Repeating the calculations in Table 1 using a [34/38] analytical uncertainty consistent with these high-precision all-Faraday protocols, which are clearly more appropriate for analysing samples approaching the limit of measurable disequilibrium, leads to significantly different results.

Perhaps the authors would justify use of a 4‰ (2 σ) uncertainty based on arguments put forward in the Vermeesch et al. (2025) preprint—cited multiple times throughout the manuscript—which claims that speleothem [34/38] measurements are, as a general rule, overdispersed with respect to their analytical uncertainties. For example, this line of argument is suggested on line 230 where they write ‘...making the optimistic assumption that the analytical uncertainty of the [34/38]_m-measurements faithfully captures all sources of dispersion’. However, the arguments put forward by Vermeesch et al. (2025) on this matter are deficient in a number of ways:

1. Firstly, they erroneously state that the [34/38]_m analyses from Walker et al. (2006) come from ‘a Sterkfontein speleothem’ (my emphasis), and refer to them as ‘duplicate samples’, implying they are also representative of the same growth interval. However, it is very clear from the text in Walker et al. (2006), as well as from Figure 1, that the [34/38] measurements actually come from at least three⁵ separate flowstones: STA09, STA12, and STA15. These flowstones were not even all collected from the same horizon, and so it is in no way surprising that their combined [34/38]_m data are overdispersed with respect to their analytical uncertainties. It is also not clear how the sampling was performed for each individual flowstone, i.e. were the sub-samples carefully extracted from the same growth layer, or were they sampled across different growth zones? This detail is critical in assessing overdispersion because it is very well known that speleothem [34/38]_i values can vary considerably across growth layers due to factors such as changing hydrological conditions (e.g. Fairchild & Treble, 2009). Therefore, great care needs to be taken in sampling young speleothems for U/Pb and [34/38] analysis to ensure each aliquot is representative of the same growth interval.
2. Beyond their misinterpretation of the [34/38]_m data from Walker et al. (2006), Vermeesch et al. (2025) present a logically flawed argument. They assert that because, according to their assessment, the [34/38]_m data from Walker et al. (2006) are overdispersed relative to analytical uncertainties, then all speleothem [34/38]_m ratios are likely to be similarly overdispersed, rendering U–Pb ages and uncertainties derived from measured [34/38]_m values inherently unreliable. However, this reasoning relies on an extreme extrapolation from a single dataset to the global population of speleothems. This is analogous to claiming that because one isochron dataset of type x is overdispersed, all isochrons of type x must be as well. While some speleothems may indeed show heterogeneous [34/38] ratios along individual growth increments (e.g., due to episodic U-loss), this is far from a universal phenomenon as evidenced by the large body of speleothem U–Th datasets, which show that, for many speleothems, age inversions due to significant U-loss or gain are relatively rare.
3. Finally, Vermeesch et al. (2025) present some new speleothem [34/38]_m data from a South African flowstone in Figure 6ii, which appear to be overdispersed with respect to their analytical uncertainties. However, these data are incomplete and cannot be properly assessed because no details of the sampling or analytical method are provided by the authors.

For these reasons, reference to the Vermeesch et al. (2025) pre-print should not be accepted as justification for use of a 4 ‰ (2 σ) [34/38]_m uncertainty here. It may be helpful if the authors consider

⁵ It is not clear how STA14 fits into the picture, but it is probably a fourth flowstone.

two different scenarios in this section: one assuming that a typical ion counter protocol for $[34/38]$ measurements is adopted and the other assuming a high-precision all-Faraday protocol is used.

The second choice made in section 5 that leads to a particularly pessimistic assessment of the maximum range of measurable $[34/38]_m$ disequilibrium is the adoption of a $3\text{-}\sigma$ cut-off to define what is ‘statistically distinguishable’. This is at odds with common practice in geochronology, where statistical tests and age calculation results are near-ubiquitously reported at the $2\text{-}\sigma$ level (or equivalently using a probability threshold of 0.05). For example, it seems that essentially all results provided by *Isoplot* (Ludwig, 2012) and *IsoplotR* (Vermeesch, 2018) are presented in this way. There may be some justification for using a $3\text{-}\sigma$ cut-off here and not in other geochronological calculations, but if so, this should be provided.

Bayesian approach

The Bayesian methodology introduced in Section 4 seems to be a reasonable approach to handling the situation where a suitable $[34/38]_m$ value is available, but it marginally overlaps secular equilibrium, making standard age uncertainty propagation approaches unreliable. However, the default $[34/48]_i$ prior that is proposed, i.e. a uniform distribution spanning 0 to 20, is not at all consistent with the available global data for speleothems (e.g. Hellstrom, 2013; Markowska et al., 2025). For one thing, there is far too much $[34/48]_i$ probability density assigned to the interval 10–20. Therefore, it is not clear that use of this prior would produce accurate results.

In most settings a more informative local or regional prior could be constructed. It is stated on line 132 that such a prior could easily be accommodated by the Bayesian algorithm, but there is no discussion of how this might influence the calculated age or its age uncertainties relative to use of the uniform prior, which is proposed as the default.

Finally, this age calculation approach yields age uncertainty estimates in the form of Bayesian credible intervals instead of frequentist-type confidence intervals, which are more commonly encountered in geochronology. Perhaps it would be useful to include a discussion of how these Bayesian credible intervals should be interpreted, and what the practical differences are between credible intervals and confidence intervals in this geochronological context?

Limits of U–Th dating method

Line 20 states that the limit of the U–Th disequilibrium dating method is 800 ka, whereas line 204 implies it is ~ 1 Ma. These limits are obviously inconsistent and there are no arguments or citations provided to support either.

U–Th age uncertainties tend to increase in an approximately exponential manner with age, and become significantly degraded and skewed as the system approaches secular equilibrium (e.g. Ludwig, 2003a). The nominal limit of the U–Th method depends to some extent on the amount of age uncertainty that can be accommodated in usefully addressing a particular scientific issue. However, there is also a hard limit at the point where the measured $[34/38]$ – $[30/38]$ composition of a sample becomes indistinguishable from infinite age compositions, i.e. where the measured $[34/38]$ – $[30/38]$ composition significantly overlaps the ‘infinite’ age isochron within uncertainty on a $[34/38]$ versus $[30/38]$ evolution diagram (Woodhead et al., 2019). At this point, the method is only useful for providing a reliable lower age bound.

Using the authors’ preferred $[34/38]$ measurement uncertainty of $\pm 4\text{‰}$ (2σ), and assuming a similar analytical precision for $[30/38]_m$, along with an initial $[34/38]$ value of 1.2, U–Th age uncertainties are approximately $-69/+174$ at 550 ka. An age with this level of relative uncertainty is

unlikely to be useful for addressing any key scientific issues in Quaternary science. Not far beyond 550 ka, the upper age bound becomes undefined for this level of analytical precision.

If activity ratio measurement uncertainties more consistent with high-precision U–Th measurement protocols are adopted, then the useful range of the U–Th method may be extended beyond 600 ka (e.g. Cheng et al., 2016). However, there is certainly no argument that this limit is ~1 Ma.

²⁰⁸Pb normalization

Section 6 discusses the normalisation of Pb and U isotopic ratios to ²⁰⁸Pb instead of ²⁰⁴Pb when dating young carbonates. This idea has already been advocated in several previous studies that should be properly cited (e.g. Getty et al., 2001; Walker et al., 2006; Parrish et al., 2018; Engel et al., 2019).

ASH-15 isochron fit

Section 7.2 provides a re-assessment of data previously published for the ASH-15 flowstone, which has been proposed as a suitable reference material for carbonate U–Pb dating by LA-ICPMS (Nuriel et al., 2021). Part of this re-assessment involves fitting an isochron line to the TIMS ²⁰⁶Pb/²⁰⁴Pb–²³⁸U/²⁰⁴Pb data of Nuriel et al. using the model-3 algorithm of Vermeesch (2024). It is not clear whether the ‘3a’ or ‘3b’ version of this algorithm is employed here because this detail is omitted.

The model-3a algorithm attributes all excess dispersion (i.e. dispersion above that which is accounted for by analytical uncertainties) to variability in the inherited Pb component, whereas the model-3b algorithm attributes all excess dispersion to ‘diachronous isotopic closure’ of the sub-samples. In both cases the excess dispersion is attributed to a particular cause and is assumed to follow a strictly Gaussian distribution. These are both strong assumptions, in the sense that they can exert a significant impact on the resulting isochron best-fit line and its uncertainty. Therefore, when implementing these algorithms, there is a clear need to rigorously justify these assumptions.

In this case, it is difficult to envisage how the assumption of ‘diachronous isotopic closure’ could apply, unless the flowstone grew unusually slowly, and the samples were inappropriately extracted from different growth zones. On the other hand, the assumption of a heterogeneous initial Pb isotopic composition, which was suggested as the primary cause of overdispersion in the original publication, seems more reasonable. However, even if this is the only cause of overdispersion, it is not clear why variability in the inherited-Pb isotopic ratio would necessarily follow a strictly Gaussian distribution; it seems as if it could conceivably follow a range of different statistical distributions.

In addition to a heterogeneous isotopic composition of inherited Pb, there are other possible causes of overdispersion that should be considered. These include:

1. Pb contamination: either varying amounts of Pb contamination or contamination with Pb components that have different isotopic compositions. This possibility should be considered here because the sub-samples analysed by Nuriel et al. (2021) were not subjected to leaching in dilute HCl prior to chemistry, which previous studies have shown is crucial to removing Pb contamination introduced during sample handling and preparation, which can easily dominate the overall Pb budget in young carbonates (e.g. Woodhead et al., 2006, 2012).
2. Open-system behavior, and especially U-loss. This may impart a similar effect to ‘diachronous isotopic closure’ but is unlikely to conform to a strict Gaussian distribution.

Whatever assumptions are employed here, it is essential that they are stated clearly and rigorously justified.

Finally, contrary to the what is written in the *Code and data availability* declaration, the ‘ASH15K’ data are not provided in the supplementary information.

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