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Technical Note: Pb-loss-aware Eruption/Deposition Age Estimation

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Abstract. Interpreting overdispersed crystallization and closure age spectra has become a problem of significant import in high-precision geochronology. While Bayesian eruption age estimation appears to provide one promising avenue for the statistical interpretation of such dispersed datasets, existing methods critically hinge on an assumption of fully closed-system behavior after the time of eruption. However, given the presence of two independent decay systems with distinct responses to open-system behavior, the U/Pb system provides in principle the possibility of quantifying and potentially even constraining the timing of Pb-loss. Here we present a method for Pb-loss-aware eruption age estimation, that explicitly models not only the duration of crystallization but also the timing and magnitude of Pb-loss given accurate ^{206}Pb/^{238}U and ^{207}Pb/^{235}U isotopic ratios and covariances, leading to eruption age estimates that are potentially robust to post-eruptive Pb-loss. Further applications to detrital zircon data are considered.

1 Introduction

As a consequence of continually improving analytical precision and accuracy, geologic heterogeneity which was once impossible to discern amongst larger analytical uncertainties has become a primary limitation in high precision geochronology (Schoene, 2014; Samperton et al., 2015; Keller et al., 2018). Over the past decade, such dispersion has become difficult to ignore in single-crystal U/Pb zircon (e.g., Wotzlaw et al., 2013) and more recently Ar/Ar sanidine (e.g., Ellis et al., 2017) age spectra from many felsic volcanics, precluding the use of traditional weighted mean age interpretations. Amongst other potential solutions, a Markov chain Monte Carlo approach for eruption age estimation from dispersed age spectra in volcanic samples (Keller et al., 2018), and analogously saturation/emplacement/solidus age estimation in plutonic samples (Ratschbacher et al., 2018) has recently become relatively widely applied to overdispersed U/Pb zircon (Griffis et al., 2019; Schoene et al., 2019; Liao et al., 2020; MacLennan et al., 2020; Griffis et al., 2021; Al-Suwaidi et al., 2022; Muedi et al., 2022), U/Th zircon (Popa et al., 2020; Cisneros de León et al., 2021, 2023), and Ar/Ar sanidine (Deino et al., 2019; van Zalinge et al., 2022) datasets in volcanic contexts, as well as U/Pb zircon (Kinney et al., 2021, 2022; Pamukcu et al., 2022) and U-Th/Pb monazite (Pye et al., 2022) in plutonic contexts.

Part of the success of this method has likely been due been its flexibility, applicable in principle to a wide range of high-temperature geochronometers, subject only to the choice of an appropriate prior *relative crystallization* (or closure) *distribution*. This distribution, which encodes prior information about the relative shape of the crystallization or closure distribution independent of the absolute ages involved, may be approximated for U/Pb or U-Th zircon data as a uniform or truncated Normal

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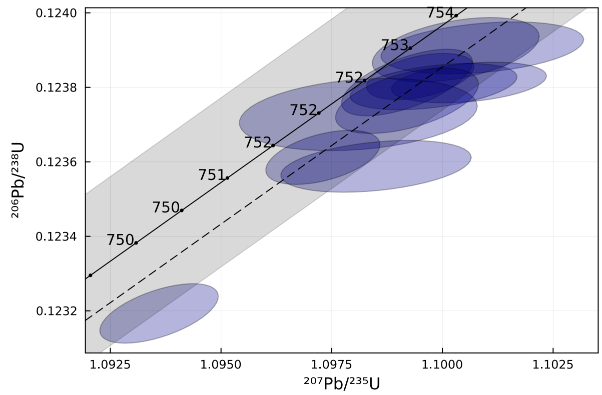

Figure 1. Concordia plot of CA-ID-TIMS zircon analyses from a typical Precambrian sample with signs of mild Pb-loss, from MacLennan et al. (2020), excluding one analysis that did not overlap Concordia within mutual uncertainty. Solid black Concordia line and gray uncertainty envelope show Concordia according to the traditional decay constants and (2-sigma) uncertaities of Jaffey (Jaffey et al., 1971), while the dashed black line shows Concordia using the $\lambda_{^{235}U}$ of Schoene et al. (2006). Error ellipses are plotted at the bivariate 95% coverage level.

distribution, bootstrapped from a set of similar age spectra, or even calculated by first principles given zircon saturation dynamics and magma thermal history (Keller et al., 2018) – and for Ar/Ar sanidine data appears to commonly take the form of an exponential survivorship curve (van Zalinge et al., 2022).

However, one key limitation of this method (and, indeed of many other methods including both weighted mean and especially "youngest zircon" age interpretations) is the assumption of strictly closed-system behaviour after the time of eruption, or in other words that no crystallization or closure ages can ever postdate eruption except due to analytical uncertainty. In the U/Pb system, this is a generally reasonable assumption for young, chemically abraded (Mattinson, 2005) zircons, and can be checked by concordance (Wetherill, 1956). However, for older zircons, particularly those of Paleozoic and older age, this assumption becomes progressively less reasonable, and while the check of concordance persists, it is common to find borderline cases – where a zircon still overlaps Concordia within uncertainty, but shows subtle signs of Pb loss, and a subjective call must be made. For example, consider the analyses from sample KR18-04 of MacLennan et al. 2020 (MacLennan et al., 2020) shown in Figure 1. One discordant analysis, which did not overlap within mutual uncertainty of Concordia, has already been excluded. However, another suspect analysis remains which, despite overlapping Concordia within mutual uncertainty, remains an outlier with respect to the main cluster of analyses. Even within the main cluster, some analyses are noticeably more concordant than others – but without any clear outliers, and all overlap Concordia within mutual uncertainty.

2 Pb-loss-aware eruption age estimation

Currently, no statistical method exists within the literature to fully obviate the subjective identification of Pb loss. While a traditional discordia array could be fitted to the entire dataset, potentially eliminating the problem of Pb loss, this would be

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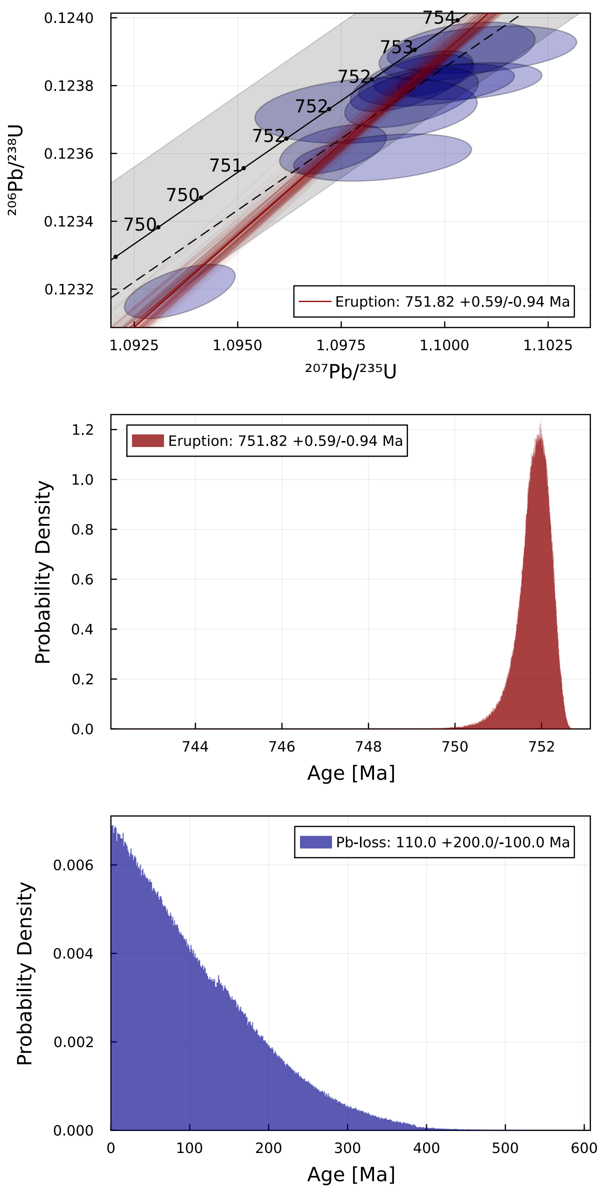

Figure 2. The Pb-loss-aware eruption age estimation approach applied to the dataset of Figure 1. Top: A selection of Concordia arrays between the time of eruption and the time of Pb loss drawn from the stationary distribution of the Markov chain, representing the upper left boundary of the data in Concordia space exclusive of analytical uncertainty. Middle: the posterior distribution of the time of eruption. Bottom: the posterior distribution of the time of Pb loss.

as statistically invalid as a traditional weighted mean given the geologic dispersion of the data. Conversely, while the eruption age estimation approach of Keller et al. (2018) could properly address the geologic dispersion, it can do so only if analyses

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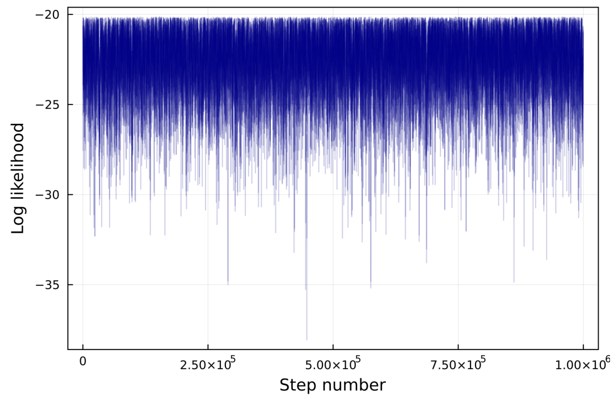

Figure 3. Log likelihood of the first million steps of the Markov chain of the Pb-loss-aware eruption age estimation algorithm. The chain appears to be stationary virtually immediately, likely due to the ease of starting with relatively accurate initial guesses for the times of zircon saturation, eruption, and Pb loss (the oldest zircon, youngest zircon, and 0 Ma, respectively).

influenced by Pb loss have already been eliminated. MacLennan et al. (2020) themselves took a metaanalytical approach to this problem, where the number of zircons to be excluded for Pb loss is merely another another statistical parameter, which can be resampled to propagate this second level of uncertainty. While this method is likely the best applied to date, it does not entirely eliminate the problem. However, given the independence of the chronometers $^{207}Pb/^{235}U$ and $^{206}Pb/^{238}U$, which traditionally allows for the identification of Pb loss, it appears possible that an eruption age estimation algorithm that explicitly considers both chronometers may be able to model, and potentially eliminate, the influence of at least a single Pb loss event.

Consequently, we consider here a procedure which, given a set of dispersed $^{207}Pb/^{235}U$ and $^{206}Pb/^{238}U$ analyses, explicitly models (1) the time of onset of crystallization (saturation), (2) the time of eruption, and (3) the time of Pb loss. This procedure, much like that of Keller et al. (2018) follows a Markov chain Monte Carlo approach, with the modification relative to Keller et al. (2018) that at each step of the chain we also propose a potential time of Pb loss and project back to Concordia before assessing saturation or eruption ages. That is:

1. Propose a time of Pb loss, a time representing the onset of crystallization (and closure) of your chronometer, and a time representing the termination of crystallization (or closure) by eruption

2. Project along a Pb-loss array from your proposed time of Pb loss, through each analysis, back to Concordia.

3. Assess the log likelihood of drawing the projected ages from your relative crystallization age distribution given your proposed times for the onset of crystallization and eruption, just as you would in the method of Keller et al. (2018). This likelihood calculation may include the likelihood terms for any prior distributions (if known) for time of Pb loss (by default, a Uniform distribution from 0 to the youngest $^{206}Pb/^{238}U$ age) and the extent of Pb loss (by default, Uniform from 0 to 100%).

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	Pb-loss-aware eruption age estimate	$^{206}Pb/^{238}U$ eruption age estimate
Excluding no analyses	751.84 +0.58/-0.90	743.28 +0.80/-1.86
Excluding one discordant analysis	751.83 +0.59/-0.95	749.57 +0.67/-0.94
Excluding four $>0.07\%$ discordant analyses	751.79 +0.62/-1.07	751.47 +0.64/-0.75

Table 1. Comparison of Pb-loss-aware and standard eruption age estimates, using a uniform prior relative crystallization distribution in each case, with uncertainties reported at the 95% credible interval. While the traditional $^{206}Pb/^{238}U$ eruption age estimate is strongly sensitive to the number of excluded analyses, the Pb-loss-aware eruption age estimate is virtually constant across all variants – with only a minor increase in uncertainty as more analyses are excluded.

4. Accept or reject your current proposal by comparing the likelihood current proposal to the likelihood of the last accepted proposal, accepting with probability equal to the ratio of likelihoods (i.e., the Metropolis algorithm (Metropolis et al., 1953)), again just as you would in the method of Keller et al. (2018)

5. Propose a new time of Pb loss, onset of crystallization, and eruption by drawing from a symmetric proposal distribution around the most recent accepted proposal. Repeat

A reference implementation of this algorithm is provided in the registered Julia (**?**) package Isoplot.jl **?** and, like the original approach of Keller et al. (2018), optionally integrated with the age-depth modelling programs Chron.jl (Keller, 2018) and SubsidenceChron.jl (Zhang et al., 2023).

The results of this procedure, applied to the dataset of MacLennan et al. (2020), are shown in Figure 2 and Table 1. After a potential period of burn-in (too rapid to be observed in Figure 3, likely due to the relative ease of generating a good initial guess), the resulting stationary distribution gives us a posterior estimate for the time of eruption (Figure 2b), as well as our posterior estimate of the time of Pb loss (Figure 2c). This posterior distribution for the time of Pb loss might in principle be multimodal or otherwise complicated, but in practice appears to often take the form of a broad triangular or truncated Normal distribution with a mode at or near 0 Ma – consistent with the long-observed phenomenon of "zero-age Pb loss" (Wetherill, 1956; Stern et al., 1966; Black, 1987; Keller et al., 2019), which has been attributed to alteration by meteoric water during modern surficial exposure and incipient weathering (Stern et al., 1966; Black, 1987; Keller et al., 2019; Andersen and Elburg, 2022). Despite the significant uncertainty in the time of Pb loss indicated by such a broad distribution, the concomitant uncertainty in the time of eruption is generally small given data that are close to Concordia.

In contrast to traditional $^{206}Pb/^{238}U$-based eruption age estimation, the Pb-loss-aware eruption age estimate is markedly insensitive to the inclusion or exclusion of individual young, discordant analyses – as indeed might be anticipated if our Pb-loss-aware eruption age estimation approach is operating as designed. While the inversion shown in Figure 2 excludes only one clearly discordant analysis, Figure 4 shows two alternative endmember approaches – ranging from excluding all analyses with $>0.07\%$ discordance (top), to at the other extreme not excluding any data whatsoever (bottom). Despite the significant differences in screening, all yield Bayesian Pb-loss-aware eruption estimates, as seen in Table 1, of approximately 751.9

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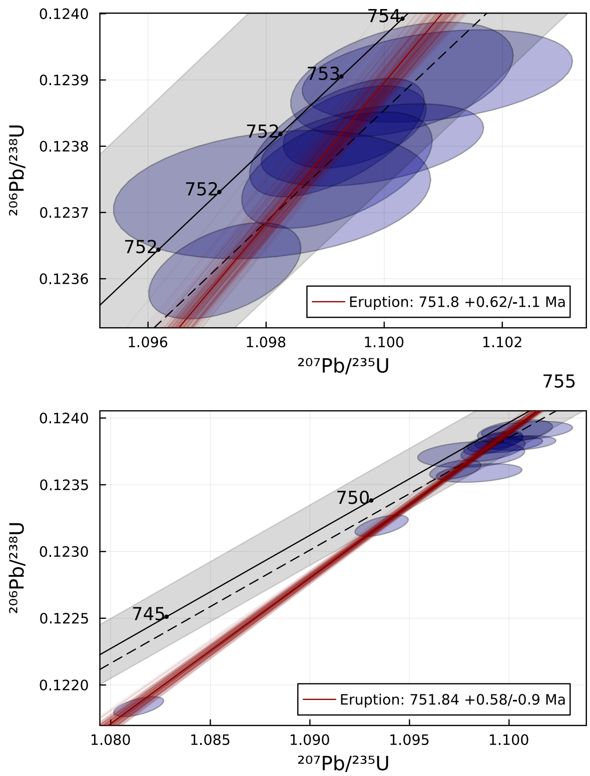

Figure 4. The application of Pb-loss-aware eruption age estimation to both filtered (top; excluding four analyses with $> 0.07\%$ discordance) and unfiltered (bottom; excluding no analyses) datasets. Despite the significant differences between the filtered and unfiltered datasets, effectively equivalent eruption age estimates are obtained in each case. As in Figure 1, solid black Concordia line and gray uncertainty envelope show Concordia according to the traditional decay constants and (2-sigma) uncertaities of Jaffey (Jaffey et al., 1971), while the dashed black line shows Concordia using the $\lambda_{^{235}U}$ of Schoene et al. (2006). Error ellipses are plotted at the bivariate 95% coverage level.

+0.6/-1.0 Ma (95% CI). Traditional $^{206}Pb/^{238}U$ Bayesian eruption estimates, meanwhile, vary by nearly 10 Myr from 743.3 +0.8/-1.9 Ma when excluding no analyses to 751.5 +0.6/-0.8 Ma (within uncertainty of the Pb-loss-aware estimate) with the strictest screening (Table 1).

Despite these advantages, the proposed method is not without limitations. Firstly, due to the projection approach, our method implicitly assumes that all crystals share the same distribution for the time of Pb loss – that is, it assumes all grains share the same Pb loss history in time, if not in magnitude. Pb loss that is particularly close in time to the time of crystallization may also be more difficult to correct. Relatedly, while the Markov chain may sample from a broad distribution for the time of Pb loss may be broad, and will propagate all uncertainty in the time of eruption concomitant to this uncertainty in the time of Pb loss, only a single episode of Pb loss may be simulated at once. Consequently, the method may not be best suited to samples that have complicated Pb-loss histories with multiple distinct, competing episodes of Pb loss. In addition, since the proposed

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method relies on both the ^{206}Pb/^{238}U and ^{207}Pb/^{235}U systems to constrain the potential timing and magnitude of potential Pb loss, the precision and accuracy of the results is necessarily dependent on the accuracy and precision of both ^{206}Pb/^{238}U and ^{207}Pb/^{235}U ages – the latter of which may be more challenging in young rocks due to the low present-day abundance of ^{235}U. Likewise, the influence of intermediate daughter product disequilibrium (e.g., ^{231}Pa and ^{230}Th) should be considered and, if significant, corrected for both decay chains. Analyses which are reversely discordant for any reason are not generally suitable for this method. Considering the several aforementioned limitations of the Pb-loss-aware eruption age estimation method in younger rocks due to, as well as the lower likelihood of significant Pb loss in younger zircons, the proposed method is not best suited to young samples (less than ∼100 Ma); single-chronometer $^{206}Pb/^{238}U$ (or U/Th, Ar/Ar, etc.) eruption age estimation may be recommended instead in such cases.

One final noteworthy consideration involves the choice of uranium decay constants. In contrast to standard single-chronometer eruption age interpretation, where decay constant uncertainties are best excluded during eruption age interpretation (and only reapplied to the final eruption age estimate), this approach requires at minimum the assumption of an accurate $\lambda_{^{235}U}/\lambda_{^{238}U}$ decay constant ratio. While the accuracy of the traditional ^{235}U and ^{238}U decay constants of Jaffey et al. (1971) may require reassessment (Schoene et al., 2006; Parsons-Davis et al., 2018), for our purposes it is sufficient to take the Jaffey et al. $\lambda_{^{238}U}$ as a standard and, to obtain the most accurate possible $\lambda_{^{235}U}/\lambda_{^{238}U}$ ratio, use the $\lambda_{^{235}U}$ of Schoene et al. (2006) ($9.8569 \pm 0.0017 \times 10^{-4} Myr^{-1}$) which is in turn calibrated against the $\lambda_{^{238}U}$ of Jaffey et al. (1971).

3 Application to depositional age estimation

While the initial proposed application of this new method is to eruption age estimation from dispersed CA-ID-TIMS dates with potentially borderline signs of Pb loss, it is worth noting that the same method may be of potential interest to the problem of depositional age estimation from detrital mineral age spectra. Detrital mineral geochronology, particularly including detrital zircon geochronology has exploded in popularity and accessibility over the past decades (Fedo et al., 2003; Gehrels, 2011), providing significant motivation to design a method, if possible, for the precise and accurate estimation of depositional ages from dispersed detrital mineral age spectra (Galbraith and Laslett, 1993; Ludwig and Mundil, 2002; Coutts et al., 2019; Sharman and Malkowski, 2020; Vermeesch, 2021a).

The possibility of applying the method of Keller et al. (2018) to detrital mineral depositional age estimation has been noted by Deino et al. (2019), who applied this method to the problem of interpreting dispersed sanidine Ar/Ar ages from detritally reworked tephra. On one hand, the problem of eruption age estimation from a dispersed preruptive mineral age distribution is indeed mathematically equivalent to the problem of deposition age estimation from a dispersed predepositional mineral age distribution. On the other hand, the timescales involved are radically different – with preeruptive zircon crystallization typically spanning some tens to hundreds of kyr, while predepositional detrital zircon age spectra may span millions to billions of years – and while in the case of eruption age estimation there is a positive expectation of continued zircon crystallization until near the time of eruption, in the detrital case there is no equivalent *a priori* expectation that significant zircon input (either by airfall or by detrital transport) must continue right up until the time of deposition. Nonetheless, this limitation is by no means unique

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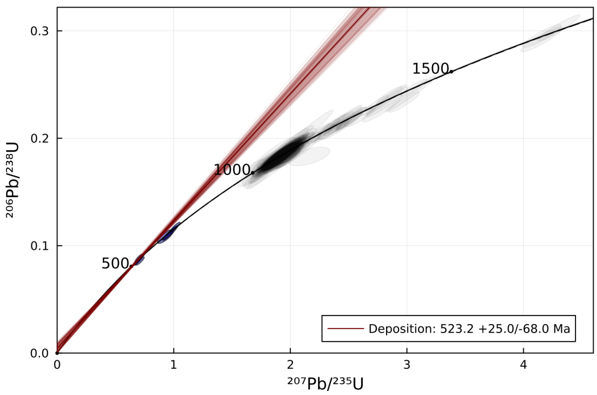

Figure 5. The Pb-loss-aware limiting age method applied to the problem of depositional age estimation from a detrital zircon age spectrum. Here we consider the LA-ICPMS detrital zircon dataset of Karlstrom et al. (2018) for the lowermost Sixtymile formation (unit "A", sample K16-60-19) and apply the Pb-loss-aware MCMC inversion method to the last Gyr of detrital zircon data (blue) using a uniform prior crystallization distribution and a Normal prior of 0 ± 30% (one-sigma) Pb-loss extent.

to Bayesian depositional age estimation methods, but rather is fundamental to the problem of depositional age estimation as a whole, given any type of detrital mineral age spectra.

Pb loss is be a topic of particular importance in detrital samples (Vermeesch, 2021b); given the need to date hundreds to thousands of individual detrital mineral grains in a single analytical session, precision and accuracy are often limited compared to that achievable in low-N CA-ID-TIMS zircon age spectra from individual volcanic samples (e.g. >0.5% relative precision and accuracy versus <0.05% relative (Schoene, 2014)). Since the ability to precisely and accurately recognize discordant analyses is directly dependent on analytical precision and accuracy (Gaynor et al., 2022), the ability to exclude such samples is comparatively limited in most detrital studies. This concern is potentially redoubled if low-temperature aqueous/meteoric alteration and "incipient weathering" is indeed a primary driver of Pb loss in radiation-damaged (e.g. Geisler et al., 2003) zircon as previously suggested (Stern et al., 1966; Black, 1987; Keller et al., 2019; Andersen and Elburg, 2022) – albeit potentially offset somewhat by the competing phenomenon that metamict grains (the same ones that are especially prone to Pb loss) may be preferentially abraded, comminuted, and destroyed during fluvial transport (Balan et al., 2001). In sum, the limitations of the method of Keller et al. (2018) with respect to Pb loss are likely particularly acute with respect to detrital zircon depositional age estimation, in which case the currently proposed method may provide a path forward.

As an illustrative example, applying this method to the LA-ICPMS detrital zircon dataset of Karlstrom et al. (2018) for the lowermost Sixtymile formation (unit "A", sample K16-60-19) yields a depositional age estimate of 523.2 +25/-68 Ma (95% CI), as shown in Figure 5 – less precise than but in good agreement with a youngest CA-ID-TIMS age of 527.4 ± 0.7 Ma interpreted by (Karlstrom et al., 2018) as a potentially near-depositional maximum age. While the full set of potential advantages and disadvantages of this method as applied to detrital zircon data are likely not yet fully understood, this method like any other currently available, is critically dependent on continued juvenile zircon input, and should not be applied to

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settings where such juvenile input is not expected (i.e., sedimentary rocks derived from relatively tectonically passive settings far from active felsic volcanism). More broadly, the accuracy of highly precise depositional age estimates may be suspect. Nonetheless, preliminary results encourage the further application and testing of this method as applied to depositional age estimation from detrital U/Pb geochronology.

Code and data availability. The reference implementation of the Pb-loss-aware eruption age estimation is provided in the Isoplot.jl package **?**, available at https://github.com/JuliaGeochronology/Isoplot.jl under an MIT open-source license and as a registered package on the official General registry for the Julia language. The scripts to produce all figures in this paper are included in the `examples` directory of Isoplot.jl.

Author contributions. C. Brenhin Keller wrote the code, generated the figures, and prepared the manuscript.

Competing interests. The author is a member of the editorial board of Geochronology.

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