26Al–10Be–21Ne burial dating

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1. Cosmogenic-nuclide burial dating

1.1. Background: 26Al–10Be burial dating

Cosmogenic-nuclide burial dating relies on a pair of cosmic-ray-produced nuclides that are produced in the same rock or mineral target at a fixed ratio, but have different half-lives. For example, 26Al and 10Be are produced in quartz at 26Al/10Be = 6.75:1. If a sample of quartz is exposed at the surface for a time, 26Al and 10Be concentrations reflect this ratio; if it is then buried below the penetration depth of cosmic rays, production stops and both nuclides decay. The half-life of 26Al is half that of 10Be, so the 26Al/10Be ratio decreases over time and can be used to date the burial event. Because quartz derived from surface erosion and then buried by sediment accumulation is common, the method is widely applicable for dating Plio-Pleistocene clastic sediments. All (terrestrial) applications of burial dating so far have used the 26Al–10Be pair. Here we show that coupling cosmogenic 21Ne, which is also produced in quartz, with 26Al or 10Be should improve upon both the age range and accuracy of 26Al–10Be burial dating. We establish the feasibility of this approach by 21Ne measurements at two sites that have already been dated using 26Al–10Be burial dating. Burial ages from all three nuclide pairs agree at both sites, which shows that currently accepted values for decay constants and production ratios are internally consistent. Thus, it is possible at present to increase the useful range of cosmogenic-nuclide burial dating by incorporating 21Ne. Fully realizing the potential improvements in accuracy would benefit from additional estimates of 21Ne/26Al and 21Ne/10Be production ratios that are independent of the 26Al and 10Be decay constants.

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1.1. Background: 26Al–10Be burial dating

Cosmogenic-nuclide burial dating is a means of dating geological deposits by measuring pairs of rare nuclides that are produced at a fixed ratio during cosmic-ray bombardment of a rock or mineral target, but have different half-lives (see Granger, 2006 for a complete overview). The nuclides most commonly used for this purpose, 26Al and 10Be, are produced in quartz at a ratio 26Al/10Be = 6.75:1. A sample of quartz that experiences a single period of exposure at the Earth’s surface has 26Al and 10Be concentrations governed by this ratio. If this sample is then buried deeply enough to be shielded from the cosmic-ray flux, nuclide production stops and inventories of both nuclides decrease by radioactive decay. The half-life of 26Al (0.705 Ma) is shorter than that of 10Be (1.39 Ma), so the 26Al/10Be ratio decreases exponentially with the duration of burial.

Burial dating only requires quartz that has been exposed at the surface for a time and then buried. Neither the formation of new minerals nor the preservation of age-diagnostic fossils is required. Quartz is ubiquitous, and the formation of nearly all sedimentary deposits naturally involves surface exposure of the sediment followed by burial after deposition; thus, the method is potentially attractive for dating Plio-Pleistocene clastic sediments that cannot be dated by other means. Applications of burial dating have, in fact, focused on problems in geology and anthropology that could not be solved by existing dating methods, including i) determining valley incision rates from the age of stranded cave and terrace sediments (Granger et al., 2001; Stock et al., 2005a; Haeuselmann et al., 2007); ii) dating early and middle Pleistocene ice sheet expansions (Balco et al., 2005a; Balco and Rovey, 2008); and iii) dating hominin fossils and stone tool assemblages in regions that lack a volcanic ash chronology (Gibson et al., 2009; Shen et al., 2009).

Although there are a number of cosmogenic-nuclide pairs that could be used for burial dating, all terrestrial applications of the technique so far have used 26Al and 10Be in quartz. For the most part this is because these nuclides are relatively easy to measure and the 26Al/10Be production ratio in quartz is well established. However, using other nuclide pairs would in principle significantly improve both the useful age range and the accuracy of the method. One possibility is to use the stable cosmogenic nuclide 22Ne, which is also produced in quartz, with 26Al or 10Be. Several studies have combined 26Al, 10Be, and 21Ne measurements on surface quartz samples, and some of them have used 21Ne/26Al or 21Ne/10Be ratios to show that these surface samples must have been buried at one time (e.g. Fujioka et al., 2005; Kober et al., 2008), but there have been no attempts to use 22Ne for burial dating. In this paper we show that 22Ne/21Ne and 10Be–21Ne burial dating can yield improvements in both age range and accuracy over 26Al–10Be burial dating, and we establish its feasibility.

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2.1. Advantages of cosmogenic $^{21}$Ne

$^{26}$Al–$^{10}$Be and $^{10}$Be–$^{21}$Ne burial dating offer two potential improvements on $^{26}$Al–$^{10}$Be burial dating: an increased usable age range and improved accuracy. The useful age range of a burial-dating nuclide pair is set by the decay constants of the two nuclides: measurement precision for each nuclide decreases with its concentration, and eventually the shorter-lived of the pair decays to a level too low to measure accurately. The useful range of $^{26}$Al–$^{10}$Be burial dating is ca. 0.5–6 Ma under ideal conditions (Fig. 1). The $^{26}$Al–$^{21}$Ne pair has a somewhat increased useful range due to the fact that only the $^{26}$Al inventory is reduced by decay. However, because i) the half-life of $^{10}$Be is approximately twice that of $^{26}$Al, and ii) $^{10}$Be can be measured more precisely at low concentrations than $^{26}$Al (e.g. Schaefer et al., 2009), the useful range of the $^{10}$Be–$^{21}$Ne pair is potentially more than double that of the $^{26}$Al–$^{10}$Be pair. $^{10}$Be–$^{21}$Ne burial dating should be effective well into the Miocene.

The total uncertainty in a burial age (that is, the uncertainty that should be used in comparing it to dates obtained with an independent method) mainly comprises i) measurement uncertainties, and ii) uncertainties in the decay constants for the nuclides in question. Other uncertainties, in nuclide production rates and the burial history of the sample, make a minor contribution to the total uncertainty in most cases (Balco et al., 2005b; Balco and Rovey, 2008).

Because of the need to deconvolve cosmogenic $^{21}$Ne from other sources of $^{21}$Ne by measurements of multiple Ne isotopes (Niedermann et al., 1993), measurement uncertainties are commonly larger for $^{21}$Ne than for $^{26}$Al and $^{10}$Be. However, as $^{21}$Ne is stable, the $^{21}$Ne inventory does not decrease after burial, so there is no loss of measurement precision with increasing burial age. In addition, the fact that $^{21}$Ne is stable eliminates one of the decay constant uncertainties. Decay constants for $^{26}$Al and $^{10}$Be are known with ±3% and ±1% precision, respectively, (Nishizumi, 2004; Chmeleff et al., 2009; Korschinek et al., 2009), and propagation of these uncertainties into an $^{26}$Al–$^{10}$Be burial age implies an age uncertainty of 5% that cannot be reduced by improving the precision of the $^{26}$Al and $^{10}$Be measurements. The $^{26}$Al–$^{21}$Ne and $^{10}$Be–$^{21}$Ne pairs are only subject to part of this decay constant uncertainty. Finally, the total uncertainty in a burial age is inversely proportional to the difference between the decay constants of the two nuclides. This difference is similar for the $^{26}$Al–$^{10}$Be and $^{10}$Be–$^{21}$Ne pairs. However, it is twice as large for the $^{26}$Al–$^{21}$Ne pair, which decreases the total uncertainty in an $^{26}$Al–$^{21}$Ne burial age. To summarize, although measurements of $^{21}$Ne at typical surface concentrations are often less precise than $^{26}$Al and $^{10}$Be measurements, the total uncertainty in $^{26}$Al–$^{10}$Be and $^{10}$Be–$^{21}$Ne burial ages should be less than that in $^{26}$Al–$^{10}$Be burial ages. Fig. 1 shows this relationship.

2. This study: examples of $^{21}$Ne–$^{10}$Be–$^{26}$Al burial dating

To investigate the feasibility of burial dating with $^{21}$Ne as well as the internal consistency of currently accepted production rates and decay constants for $^{26}$Al, $^{10}$Be, and $^{21}$Ne, we measured $^{21}$Ne concentrations in quartz samples from two sites where we had already measured $^{26}$Al–$^{10}$Be burial ages. We selected these sites for two reasons. First, the samples were derived from slowly eroding cratonic landscapes, so had relatively high nuclide concentrations at the time of burial. This facilitates precise measurement of nuclide concentrations, making it possible to accurately assess consistency between ages derived from the various nuclide pairs. Second, the geologic context at these sites shows that the samples were buried rapidly and deeply enough that post-burial nuclide production by deeply penetrating muons contributes only a small fraction of the total nuclide inventory. This ensures that the burial ages are only weakly sensitive to uncertainties in production rates due to muons.

Riverbluff Cave in Springfield, Missouri, USA, contains a sequence of fissile gravels and backwater sediments that were derived from a slowly eroding bedrock upland, and deposited in the cave by a nearby river that formerly flowed through it. The cave is now stranded above river level due to river diversion and subsequent valley incision. A series of five quartz sand samples from the cave sediments yielded stratigraphically ordered $^{26}$Al–$^{10}$Be burial ages between 0.65 and 1.1 Ma (Rovey et al., in press). We measured $^{21}$Ne in one sample from this site.

At the Pendleton clay pit near Pendleton, Missouri, USA, the Whippoorwill formation, a colluvial deposit derived from weathering of underlying bedrock, was buried by till during an early Pleistocene advance of the Laurentide Ice Sheet. Four $^{26}$Al–$^{10}$Be burial ages from the Whippoorwill at this site (Rovey and Balco, in press), as well as two others from a stratigraphically equivalent site nearby (Balco et al., 2005a), date till emplacement at 2.42±0.14 Ma (average of results from both sites). We measured $^{21}$Ne in all four samples from the Pendleton site.

3. Methods

3.1. Analytical methods

We isolated quartz from sand-sized sediment, extracted Be and Al using standard methods of HF dissolution and column chromatography at the Cosmogenic Nuclide Lab at the University of Washington.
(Stone, 2004), and measured Be and Al isotope ratios by accelerator mass spectrometry at the Center for Accelerator Mass Spectrometry, Lawrence Livermore National Laboratory. ²⁶Al and ¹⁰Be concentrations appear in Table 1. We extracted ⁴¹⁰⁰Ne from aliquots of the same purified quartz samples in the Noble Gas Thermochronometry Lab of the Berkeley Geochronology Center either by heating the sample in a resistance furnace, or by encapsulating it in a Ta packet and heating it with a 75 W diode laser. We analysed the released Ne on a MAP-215 mass spectrometer using an ⁳⁵Ar spike to correct for isobaric interferences on masses 20 and 22. Balco and Shuster (2009) give complete details of the measurement technique. Summary ⁴¹⁰⁰Ne concentrations appear in Table 1 and complete results of the step-degassing analyses in Table S1.

The Riverbluff Cave sample is composed of marine chert derived from local limestone, and contained an unusually large quantity of trapped Ne (Table S1). The isotope composition of Ne in this sample lay significantly above the atmospheric–cosmogenic mixing line (Table S1; Fig. S1), which is consistent with the idea that trapped Ne was fractionated from air either during atmosphere–ocean gas exchange or diagenesis (e.g. Matsubara et al., 1991). Thus, we calculated cosmogenic ⁴¹⁰⁰Ne as excess ⁴¹⁰⁰Ne with respect to a mass fractionation line passing through atmospheric Ne (Niedermann et al., 1994). Even though this sample contains an easily measurable amount of cosmogenic ⁴¹⁰⁰Ne, and the large signal contributed by trapped Ne permits precise Ne isotope ratio measurements, the fact that ~95% of the ⁴¹⁰⁰Ne in this sample is not cosmogenic in origin compounds uncertainties such that the precision of the cosmogenic ⁴¹⁰⁰Ne measurement (12%) is not adequate to improve on the ⁴⁶Al–¹⁰Be burial age. For this reason, we did not pursue the Riverbluff Cave samples any further.

Samples from the Pendleton site contained significantly less trapped Ne and had Ne isotope compositions indistinguishable from the atmospheric–cosmogenic mixing line (Table S1; Fig. S1). Thus, we calculated cosmogenic ⁴¹⁰⁰Ne in these samples on the basis of two-component mixing between atmospheric and cosmogenic Ne. Duplicate analyses of two samples agreed within their respective uncertainties, and total measurement uncertainties were 4–7%.

3.2. Calculation of burial ages

Burial ages for the Riverbluff Cave sample assume steady erosion followed by a single period of burial at their present depth; those for Pendleton pit samples use the multi-stage burial scheme described in Balco et al. (2005a), with overburden ages and thicknesses tabulated in Rovey and Balco (in press). Production rates for ⁴⁶Al and ¹⁰Be reflect the scaling scheme of Stone (2000) and the calibration data set of Balco et al. (2008) renormalized to the Be isotope ratio standards of Nishizumi et al. (2007). This implies an ⁴⁶Al/¹⁰Be production ratio of 6.75. ⁴⁶Al and ¹⁰Be production rates by muons follow Heisinger et al. (2002a,b), as implemented in Balco et al. (2008). The ²¹Ne/⁴⁶Al and ²¹Ne/¹⁰Be production ratios are not as well determined as the ⁴⁶Al/¹⁰Be production ratio; we used ²¹Ne/⁴⁶Al = 0.606 and ²¹Ne/¹⁰Be = 4.08 (Balco and Shuster, 2009). The production rate of ²¹Ne by muons is not well known. Balco and Shuster (2009) found that the proportion of production due to muons was similar for ²¹Ne and ¹⁰Be, so we calculated ²¹Ne production rates due to muons by assuming that the ²¹Ne/¹⁰Be production ratio was constant with depth. Because of the site selection criteria discussed above, this is a minor issue for our purposes. For the ⁴⁶Al and ¹⁰Be decay constants we used values of 9.83 ± 0.25 × 10⁻⁷ (Nishizumi, 2004) and 4.987 ± 0.043 × 10⁻⁷ (Chmielewski et al., 2009; Korschinek et al., 2009) yr⁻¹, respectively.

Table 1 shows the burial ages. The stated total uncertainties in burial age include measurement uncertainties in the nuclide concentrations and uncertainties in the decay constants. Because we are mainly interested in determining whether or not burial ages calculated using different nuclide pairs for the same sample are mutually consistent, we did not include either i) site-specific uncertainties associated with the burial history of the samples, or ii) uncertainties in the production ratios. We computed the uncertainties using linear error propagation with the partial derivatives estimated by a first-order centered difference approximation.

4. Results and discussion

²⁶Al–²¹Ne, ¹⁰Be–²¹Ne, and ²⁶Al–¹⁰Be burial ages for each sample agreed within their respective uncertainties (Table 1; Figs. 2 and 3). Weighted averages for each nuclide pair of four samples from the Pendleton site agreed at 1.5%, as well or better than can be expected given the measurement uncertainties. This is important because it shows that the independently determined parameters used to compute the burial ages—the nuclide production ratios and the ²⁶Al and ¹⁰Be decay constants—are internally consistent. Thus, these parameters are adequately well determined to realize the increase in the useful range of cosmogenic-nuclide burial dating offered by combining ²¹Ne with ²⁶Al or ¹⁰Be measurements.

Agreement between burial ages determined from the three nuclide pairs does not, by itself, prove conclusively that all these parameters have been accurately determined and thus that the burial ages are accurate. However, i) these parameters were for the most part determined independently by a variety of techniques, and ii) for these parameters to be inaccurate and still yield consistent burial ages for all three nuclide pairs would require a systematic offsetting relationship among errors in estimating the various parameters. These observations strongly suggest that the production ratios and decay constants have been accurately determined.

Table 1

<table>
<thead>
<tr>
<th>Sample name</th>
<th>²⁶Al(10⁶ atoms g⁻¹)</th>
<th>²¹⁰⁰Be(10⁶ atoms g⁻¹)</th>
<th>²¹Ne(10⁶ atoms g⁻¹)</th>
<th>No. of ²¹Ne measurements</th>
<th>²⁶Al–²¹Be</th>
<th>²¹⁰⁰Be–²¹Ne</th>
<th>²⁶Al–²¹⁰⁰Be</th>
</tr>
</thead>
<tbody>
<tr>
<td>Whippoorwill formation, Pendleton clay pit, Missouri</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PP-WH-0</td>
<td>0.705 ± 0.013</td>
<td>1.634 ± 0.061</td>
<td>11.31 ± 0.50</td>
<td>2</td>
<td>2.21 ± 0.010 (0.16)</td>
<td>2.41 ± 0.09 (0.09)</td>
<td>2.29 ± 0.06 (0.08)</td>
</tr>
<tr>
<td>PP-WH-0.5</td>
<td>0.69 ± 0.018</td>
<td>1.459 ± 0.104</td>
<td>10.25 ± 0.63</td>
<td>1</td>
<td>2.47 ± 0.018 (0.23)</td>
<td>2.26 ± 0.12 (0.12)</td>
<td>2.37 ± 0.11 (0.12)</td>
</tr>
<tr>
<td>PP-WH-1</td>
<td>0.709 ± 0.018</td>
<td>1.568 ± 0.075</td>
<td>10.46 ± 0.56</td>
<td>2</td>
<td>2.31 ± 0.12 (0.18)</td>
<td>2.26 ± 0.11 (0.11)</td>
<td>2.28 ± 0.08 (0.09)</td>
</tr>
<tr>
<td>PP-WH-1.75</td>
<td>0.700 ± 0.018</td>
<td>1.568 ± 0.077</td>
<td>9.90 ± 0.69</td>
<td>1</td>
<td>2.28 ± 0.13 (0.18)</td>
<td>2.19 ± 0.13 (0.14)</td>
<td>2.24 ± 0.09 (0.10)</td>
</tr>
<tr>
<td>Error-weighted mean of four samples (internal uncertainties)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>2.28 ± 0.06</td>
<td>2.19 ± 0.05</td>
<td>2.25 ± 0.04</td>
</tr>
</tbody>
</table>

| Riverbluff Cave, Springfield, Missouri | | | | | | | |
| RC-15F | 1.909 ± 0.036 | 7.69 ± 0.31 | 12.5 ± 1.4 | 1 | 0.64 ± 0.08 (0.09) | 0.46 ± 0.18 (0.18) | 0.56 ± 0.13 (0.14) |

Full ²⁶Al and ¹⁰Be process blanks were less than 0.2% of the total number of atoms measured in any sample. Complete results of the step-degassing Ne analyses appear in Table S1. Both internal (including measurement uncertainties only) and external (in parentheses; including measurement and decay constant uncertainties) uncertainties are shown for burial ages of individual samples. Site and sample information and ²⁶Al and ¹⁰Be concentrations are also published in Rovey et al. (in press) and Rovey and Balco (in press).

a Normalized to the isotope ratio standards of Nishizumi et al. (2007).
b Normalized to the isotope ratio standards of Nishizumi (2004).
To further explore this issue, we examined the sensitivity of the burial ages for the Pendleton site derived from the various nuclide pairs to adjusting the values of half-lives and production ratios (Fig. 4). First, as expected given the selection criteria for this site—that nuclide production by muons be relatively unimportant—neither the ages themselves nor their agreement is significantly affected by adjusting the $^{21}$Ne production rate due to muons (Fig. 4a). Thus, these results do not provide any new constraints on $^{21}$Ne production by muons. Of the remaining parameters involved in the calculation, now that the $^{10}$Be decay constant has recently been precisely determined (Chmeleff et al., 2009; Korschinek et al., 2009), the least certain is the...
$^{21}\text{Ne}$ production rate and the $^{21}\text{Ne}/^{26}\text{Al}$ and $^{21}\text{Ne}/^{10}\text{Be}$ production ratios that it implies. Recent estimates of the $^{21}\text{Ne}/^{10}\text{Be}$ production ratio by Balco and Shuster (2009) ($^{21}\text{Ne}/^{10}\text{Be}=4.08\pm0.37$; we used this value to compute the burial ages in Table 1) and Goethals et al. (2009) ($^{21}\text{Ne}/^{10}\text{Be}=4.31\pm0.17$) agree within their measurement uncertainties, and either value would result in acceptable consistency between burial ages computed from all three nuclide pairs (Fig. 4d). However, as one could offset this discrepancy by adjusting the $^{26}\text{Al}$ half-life by an amount smaller than its present measurement uncertainty, it is difficult to choose between the various published values for the $^{21}\text{Ne}$ production rate based on the measurements in the present study alone. To summarize, although accepted values for the $^{26}\text{Al}$ and $^{10}\text{Be}$ half-lives and the $^{21}\text{Ne}/^{10}\text{Be}$ production ratio of Balco and Shuster (2009) yield consistent burial ages for all three nuclide pairs at the Pendleton site, the offsetting relationships among several uncertain parameters make it impossible to conclusively determine the $^{21}\text{Ne}/^{10}\text{Be}$ production ratio from the relationship between burial ages obtained from the three nuclide pairs. Overall this highlights the importance of independent determinations of the production ratios and decay constants in fully realizing the potential improved accuracy of burial dating with $^{21}\text{Ne}$.

5. Conclusions

When cosmogenic-nuclide concentrations are high enough to permit precise measurement of $^{21}\text{Ne}$ in quartz, burial dating with either the $^{26}\text{Al}$–$^{10}\text{Be}$ or $^{21}\text{Ne}$–$^{10}\text{Be}$ nuclide pairs in quartz should have a longer useful age range, and be more accurate, than burial dating with the $^{26}\text{Al}$–$^{10}\text{Be}$ pair. Combined $^{26}\text{Al}$–$^{10}\text{Be}$–$^{21}\text{Ne}$ measurements from two sites show that accepted values for $^{26}\text{Al}$ and $^{10}\text{Be}$ decay constants and a set of production ratios of $^{26}\text{Al}/^{10}\text{Be}=6.75$, $^{21}\text{Ne}/^{26}\text{Al}=0.606$, and $^{21}\text{Ne}/^{10}\text{Be}=4.08$ yield internally consistent burial ages from all three nuclide pairs. Thus, it is now possible to increase

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the useful range of cosmogenic-nuclide burial dating by incorporating $^{21}$Ne. However, fully realizing the potential improvements in accuracy would benefit from additional estimates of the $^{26}$Al decay constant as well as estimates of the $^{21}$Ne/$^{26}$Al or $^{21}$Ne/$^{10}$Be production ratios that are independent of the $^{26}$Al and $^{10}$Be decay constants.

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Appendix A. Supplementary data


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