

Ma, Y. and Stuart, F. M. (2018) The use of in-situ cosmogenic 21Ne in studies on long-term landscape development. *Acta Geochimica*, 37(2), pp. 310-322. (doi:10.1007/s11631-017-0216-9)

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The use of *in situ* cosmogenic ²¹Ne in studies on long-term landscape

development

Abstract	
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Cosmogenic Ne isotopes are stable and are routinely used for constraining the timing of events and the rate of surface change beyond the limit that can be studied with the radionuclides ¹⁰Be, ²⁶Al and ³⁶Cl. In addition to quartz it can be used in a range of other minerals. Analysis typically requires significantly less material than for cosmogenic ¹⁰Be and ²⁶Al opening up the technique for small samples, for example, individual pebbles in river sediments. Analysis is easier and faster than for the radionuclides, not least because Ne measurements do not require significant chemical procedures. However, the presence of other sources of Ne in minerals tends to restrict the use of cosmogenic ²¹Ne to old landscapes and long exposure durations. In this review we briefly outline the background to cosmogenic Ne production in rocks and minerals at the Earth's surface, then documents the key uses of the technique by highlighting some earlier studies, and finishes with a short perspective on the future of the technique.

Keywords: cosmogenic nuclides, ²¹Ne, long timescale, landscape evolution.

1. Introduction

- The last two decades has seen the development of routine techniques for determining the concentration of cosmogenic nuclides in terrestrial rocks and minerals. The widespread application of, in particular, the cosmogenic radionuclides has led to a step change in knowledge of the timing and rates of Earth surface processes (Gosse and Phillips 2001). The headline applications of the technique has included the precise determination of deglaciation timescales (e.g., Ivy-Ochs et al. 2006), the slip history of faults (e.g., Benedetti et al. 2002), the erosion rate of river catchments (e.g., Portenga and Bierman 2011) and the time of sediment burial in order to determine rates of landscape evolution (e.g., Granger and Riebe 2007).
- The vast majority of terrestrial cosmogenic nuclide studies have exploited the radioactive nuclide

¹⁰Be in quartz. The stable cosmogenic nuclides ³He and ²¹Ne have higher production rates than the radionuclides, and can be measured on conventional gas source mass spectrometers that are significantly less expensive and require less manpower to operate than accelerator mass spectrometers (AMS) required for radionuclides. Despite this they have been exploited far less routinely. This is in large part due to the presence of other sources of ³He and ²¹Ne in minerals that must be quantified precisely prior to successful application of the technique. Diffusive loss of cosmogenic ³He from quartz limits the technique to mafic minerals such as olivine, pyroxene and FeO (e.g., Foeken et al. 2009; Margerison et al. 2005). Despite this restriction it has had a broad range of applications; from constraining the evolution of Cenozoic ice history of Antarctica (Kong et al. 2010; Margerison et al. 2005) and the topographic evolution of mountain belts (Evenstar et al. 2015), to determining the timing of the collapse of volcanic edifices in the Holocene (Foeken et al. 2009). Cosmogenic ²¹Ne is retained in quartz. As well as providing a useful compliment to cosmogenic radionuclides, ²¹Ne has proved to be a powerful tracer of long-term landscape change of, in particular, slowly eroding landscapes. In this paper we first review all aspects of the production of cosmogenic ²¹Ne in minerals at the Earth's surface, then discuss the techniques that are commonly used for Ne isotope measurement. This is followed by a short explanation of the complications generated by the production of Ne by nuclear reactions in minerals, and trapped air-derived Ne. It finishes with an overview on the recent advances in the application of cosmogenic 21Ne to long-term landscape development. This is separated into two sections; studies that concentrate on exposure age determinations and those that aim to quantify erosion rates.

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2. Background

2.1 Production of terrestrial in-situ cosmogenic isotopes

The terrestrial in-situ produced cosmogenic nuclides are generated by interactions of secondary cosmic-ray particles with the target nuclei in the minerals near the Earth's surface. There are three main nuclide production pathways: (i) spallation reaction by fast- and high-energy secondary cosmic ray particles, where neutrons are the dominant particle. The intensity of neutrons is attenuated

exponentially in the rock due to nuclear interactions and ionization losses (Lal and Peters 1967). The attenuation of energy and flux of the cosmic-ray particles provides the possibility for the studies of surficial processes with *in situ*-produced cosmogenic nuclides, whose production rates beneath rock surface are dependent on the intensity of neutrons and can be modeled by an exponential function with depth in the surface material (Lal 1991). (ii) negative muon capture and (iii) fast muon-induced reactions. The negative muon-induced production is only significant at the depth of 2.5-3 m in most rocks, where the high-energy neutron flux is less than 1% of the surface. Fast muons have sufficiently high energy to cause neutron reactions, but due to their very low abundance they are only become important at great depth when no other secondary cosmic-ray particles survive. Muon-induced reactions contribute only a few percent to the total cosmogenic nuclide production in the surface rocks of the Earth (e.g., Borchers et al. 2016; Braucher et al. 2011; Marrero et al. 2016).

Due to atmospheric and geomagnetic shielding the cosmic ray particle flux, and therefore cosmogenic nuclide production rates at the Earth's surface, vary with altitude and geomagnetic latitude. Consequently the rate of nuclide production has to be scaled with the variation of latitude and elevation. Scaling is mainly based on measurements of the present day cosmic ray flux. The first model was based on photographic records of spallation events in photo-emulsions (Lal 1991). This model has been modified subsequently to take account of the effect of pressure (Balco et al. 2008; Stone and Vasconcelos 2000). The majority of scaling models are based on the global neutron monitor network that is maintained by the cosmic-ray physics community (Desilets and Zreda 2003; Desilets et al. 2006; Dunai 2001; Lifton et al. 2005). These models scale production with altitude in regions of high magnetic strength. Most recently, Lifton et al. (2014) have used numerical simulations of cosmic-ray modulation by the geomagnetic field and the atmosphere calibrated using atmospheric differential flux measurements for neutrons and protons. Collectively these models produced small differences in cosmogenic nuclide production rates. Production rates are typically normalized to sea level high latitude (SLHL).

The measurement of absolute production rates of the commonly-used cosmogenic nuclides are generally done using natural calibration sites, where the accumulated cosmogenic nuclide concentration is measured in a rock surface of known age (e.g., Balco and Shuster 2009b). These

calibration sites must be old enough to have accumulated measureable nuclide concentrations, but must be young enough to have avoided significant degradation by weathering and erosion. Phillips et al. (2016) reviews the prevailing radionuclide calibration studies and introduces cosmogenic ¹⁰Be, ²⁶Al and ³⁶Cl data for four new calibration sites that were selected by the CRONUS-Earth project. Goehring et al. (2010) reviewed the prevailing calibration sites for ³He production rate, though work is ongoing (Foeken et al. 2012).

2.2 Cosmogenic neon

Neon has three stable isotopes, ²⁰Ne, ²¹Ne and ²²Ne, of which ²⁰Ne is the most abundant in atmosphere (90.50%). All three neon isotopes are produced at similar rates by cosmic ray secondary neutron spallation reactions in rocks. Due to the lower natural abundance in air, ²¹Ne and ²²Ne concentrations are most sensitive to the cosmogenic production in minerals. Cosmogenic ²¹Ne is generally adopted as the parameter to reflect the surface exposure history as the production rate is high in comparison to the amount present in minerals.

Cosmogenic Ne isotopes are produced largely from the target elements Na, Mg, Al and Si, as these have the largest reaction cross-sections and are closest in mass to Ne. Cosmogenic Ne is quantitatively retained by most silicates (Shuster and Farley 2005). The ubiquity of quartz at the Earth surface means it is the dominant target mineral, but olivine and pyroxene are used in studies of igneous rocks.

Atmosphere-derived Ne is ubiquitous in minerals. At its simplest, the Ne isotope composition of surface exposed minerals reflects the two-component mixture of atmospheric and cosmogenic neon. This mixing line is defined by the "spallation line" in the Ne three-isotope diagram (Fig. 1). The slope of the spallation line depends on the mineral composition, and the cosmogenic 21 Ne/ 22 Ne values determined experimentally for quartz are 1.120 ± 0.020 (Niedermann et al. 1993) and 1.143 ± 0.038 (Schäfer et al. 1999). The corresponding value for pyroxene is similar, 1.055 ± 0.017 (Bruno et al. 1997).

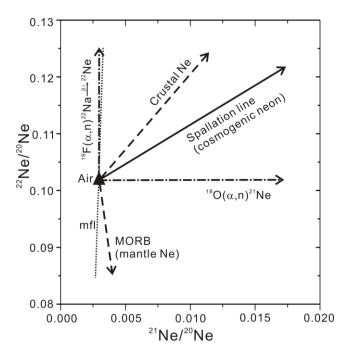


Fig. 1 Neon three-isotope diagram (22 Ne/ 20 Ne vs. 21 Ne/ 20 Ne) showing the compositions and trends of various Ne components (Niedermann 2002) mfl: mass fractionation line

Determining the production rate of cosmogenic ²¹Ne isotope is more difficult than the radionuclides and ³He, due to the relatively low production rate compared to atmosphere concentration in minerals. The first determination of cosmogenic ²¹Ne production rate in quartz was reported by Niedermann et al. (1994). Based on the production rate ratios of ²¹Ne/²⁶Al (0.65 ± 0.11), which was considered to be equal to the nuclide concentration ratio in the short exposed quartz samples (assuming an age of 11 ka based on radiocarbon dates from the work of Nishiizumi et al. (1989)) collected from the glacially polished granite surface at Sierra Nevada, California, the cosmogenic ²¹Ne production rate was determined to be 21 atoms g⁻¹ a⁻¹. These authors managed to distinguish the various neon components containing in the Sierra Nevada rock samples by isotopic signatures and release systematics. They measured all three neon isotopes in stepwise gas extraction to evaluate the cosmogenic neon, trapped components including the atmospheric, as well as inherited components. The nucleogenic neon impacted the precision of the measurement of the extremely low concentration of cosmogenic neon in this short exposed quartz samples, nevertheless, authors have shown a ²¹Ne/²⁶Al ratio with an uncertainty of 17%. Niedermann (2000) revised the rate to 19.0 ± 3.7 atoms g⁻¹ a⁻¹ after adopting a revised exposure age of 13 ka (Clark et al. 1995), and used the geomagnetic latitude of the sampling

site for scaling the production rate to SLHL.

According to the latest report about the cosmogenic nuclide production rate calibration (Phillips et al. 2016), the SLHL production rate of ¹⁰Be, which is converted from the primary calibration site production rate using Lal-Stone scaling method, is determined to be 4.01 atoms g⁻¹ a⁻¹. Based on this updated ¹⁰Be production rate, cosmogenic ²¹Ne production rate (SLHL) is revised to be 15.69 atoms g⁻¹ a⁻¹.

Balco and Shuster (2009b) reported an independent determination of ²¹Ne production rate in quartz. Instead of relying on a single calibration site with short exposure history, they selected sites in the

production and loss by surface erosion. Relatively high cosmogenic nuclide concentrations allow the high accuracy of nuclide concentration ratio determination. Combining the steady-state surface

Antarctic Dry Valleys where the surface ²¹Ne concentration has reached steady state between nuclide

concentrations ratio of ²¹Ne/¹⁰Be with the established ¹⁰Be production rate of 4.61 atoms g⁻¹ a⁻¹, they

determined the total 21 Ne production rate to be 18.3 ± 0.4 atoms g^{-1} a^{-1} (SLHL) in quartz. Using the

new ¹⁰Be production rate (Phillips et al. 2016), the cosmogenic ²¹Ne production rate (SLHL) is 15.9

atoms g⁻¹ a⁻¹ in quartz.

In the same year, Goethals et al. (2009a) used quartz collected from the surface of Bishop Tuff in eastern California with 40 Ar/ 39 Ar age of 760 \pm 2 ka (Van den Bogaard and Schirnick 1995) to determine cosmogenic 21 Ne production rate. The precise exposure age of the tuff and its relatively high cosmogenic nuclide concentrations is crucial for the accuracy of nuclide production rate determination. The authors first measured concentrations of cosmogenic 10 Be and 21 Ne; then, based on the conservative estimate of the surface erosion rate using the tuff 40 Ar/ 39 Ar age and their previously published 21 Ne concentrations, they determined the 10 Be/ 21 Ne production rate ratio of assuming steady-state erosion. When combined with the production rate of 10 Be of 4.61 atoms g^1 a^{-1} , the production rate ratio converted to a quartz 21 Ne SLHL production rate of 19.9 atoms g^1 a^{-1} . Again, using the new 10 Be production rate (Phillips et al. 2016), 21 Ne production rate (SLHL) will be 17.3 atoms g^1 a^{-1} .

2.3 Calculation of the exposure ages and surface erosion rates

For stable cosmogenic nuclides, age determinations assume that the concentration of cosmogenic 21 Ne, N(z,t), accumulates in the surface materials with the exposure age, t. At any subsurface depth z, the temporal evolution of N(z,t) in the rock can be expressed as (Lal 1991):

$$\frac{\mathrm{d}N(z,t)}{\mathrm{d}t} = P(z,t) \tag{1}$$

where P(z,t) is the production rate of the cosmogenic nuclide at the depth below the rock surface, and the temporal variation of production rate due to solar activity or geomagnetic field changes is assumed to be negligible, i.e., P(z,t)=P(z). By assuming the rates of nuclide production induced by both spallation reaction and muon reactions decrease exponentially with depth,

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$$P(z) = P_{n}(z) + P_{\mu}(z) = P_{n}(0) \cdot e^{-\rho z/\Lambda_{n}} + P_{\mu}(0) \cdot e^{-\rho z/\Lambda_{\mu}}$$
 (2)

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- where $P_n(0)$ and $P_\mu(0)$ are the rates of nuclide production induced by spallation reaction and muon reaction, respectively, at the rock surface; ρ is the rock density; and Λ_n and Λ_μ are the attenuation length (g cm⁻²) of the particles causing the spallation reaction and muon reaction, respectively. The value of Λ_n is determined by experiment and calculation, with the widely used value ranging from 157 to 167 g cm⁻² (Masarik and Reedy 1995). The value of Λ_μ can be approximated as around 1300 g cm⁻² (Barbouti and Rastin 1983; Brown et al. 1995), or using more accurate mathematical representations, including the interactions with both stopped and fast muons, reported by Granger and Smith (2000). For eroding landforms, z is time dependent, and z(t) can be defined using erosion rate on the surface, $\varepsilon(t)$, according to the following equation (Lal 1991; Niedermann 2002):
- 174 $z(t) = \int_{0}^{t} \varepsilon(t) \cdot dt + const. \tag{3}$
- Defining the initial shielding depth z_0 at time of t=0, and considering a case of constant erosion rate,
- i.e. $\varepsilon(t) = \varepsilon$, then the depth at the time of t is $z(t) = z_0 \varepsilon \cdot t$.
- Then for the differential equation (1), we can obtain the solution for stable nuclide concentration at rock surface, N(t) (Niedermann 2002):

$$179 N(t) = N(0) + \frac{P_{n}(0)}{\rho \varepsilon / \Lambda_{n}} \cdot (1 - e^{-\rho \varepsilon \cdot t / \Lambda_{n}}) + \frac{P_{\mu}(0)}{\rho \varepsilon / \Lambda_{\mu}} \cdot (1 - e^{-\rho \varepsilon \cdot t / \Lambda_{\mu}}) (4)$$

where N(0) is the initial nuclide concentration in the rock at the moment t=0 that inherited from all

previous exposure events.

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Equation (4) illustrates the relationship of cosmogenic ²¹Ne concentration with both exposure duration and erosion rate, which gives only the minimum exposure age and the maximum erosion rate from a measured cosmogenic nuclide concentration in a sample. The measurement of two cosmogenic nuclides with different half-lives in the same sample can be used to constrain exposure age and erosion rate. ²¹Ne is commonly coupled with ¹⁰Be as both can be measured in the same quart sample to investigate the history of old surfaces. The temporal evolution of the nuclide concentration ratio ²¹Ne/¹⁰Be for various erosion rates is plotted in Fig. 2, in which the area between the two curves, steady-state erosion island (Lal 1991), comprises all the possible combinations of steady-state erosion and simple exposure histories (Lal 1991; Niedermann 2002). However, complex exposure, e.g., periods of burial, will also change the ratio of the two nuclides due to ¹⁰Be decay. This is shown in Fig. 2. In a situation that a surface experienced exposure, buried for a period, and re-exposed, the concentrations of the radionuclides ¹⁰Be and ²⁶Al will return to equilibrium with the steady erosion if the re-exposure time is long enough, so the ¹⁰Be/²⁶Al pair will lose the evidence of a complex exposure history. When adding ²¹Ne measurement to ¹⁰Be or ²⁶Al, ²¹Ne concentration which is totally preserved and requires much longer time to reach steady state with the new erosion will retain the information about previous period of exposure and burial, as an evidence for recognizing complex exposure histories.

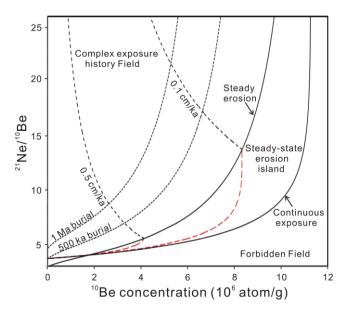


Fig. 2 Plot of the concentration ratio ²¹Ne/¹⁰Be versus the ¹⁰Be concentration, generated using

CosmoCalc version 1.7 (Vermeesch 2007) using the production rate of 5.16 atom/g/a for ¹⁰Be and 19.3 atom/g/a for ²¹Ne. The lower solid line represents the temporal evolution of these two values in the continuous exposed surface without the effect of erosion, the upper solid line in steady-state erosion condition. Dash lines indicate the temporal evolution under erosion rates of 0.5 and 0.1 cm/ka; dotted lines under different burial periods of 1 and 0.5 Ma. The area between the "continuous exposure" curve and the "steady erosion" curve is "steady-state erosion island", which comprise all possible combinations of exposure ages and erosion rates for simple exposure histories

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3. Measurement of cosmogenic ²¹Ne

Cosmogenic Ne analysis is generally performed on between 5 mg and 1 g of pure mineral. Typically this is quartz, but olivine and pyroxene have been analysed fairly routinely. This compares to 5-30 g of quartz that are usually required for ¹⁰Be and ²⁶Al determinations by AMS. Sample preparation is largely restricted to the physical extraction of a discrete mineral phase from rock. Separation relies on density methods (e.g. shaker table and heavy liquids) and magnetic techniques. The target mineral-rich fraction is usually inspected under a binocular microscope and contaminating grains are removed by hand. Chemical processing of mineral samples is typically restricted variations of the Kohl and Nishiizumi (1992) leaching procedure in order to remove the outer few 10's µm mineral which may have nucleogenic Ne generated by implanted particles and neutrons. Samples are typically ultrasonically cleaned in deionised water to remove traces of HF from mineral surfaces, then acetone to remove contaminating trace hydrocarbons. Typical analysis amounts range from 10 to 250 mg of each sample, depending on the Ne concentration. Samples are typically wrapped in metal foil - Al, Pt or Nb - in order to handle in vacuum systems. Gas extraction is typically undertaken either in double-vacuum resistance furnaces (e.g., Codilean et al. 2008) or using near-visible wavelength lasers to heat mineral samples directly (Balco and Shuster 2009b). Samples are usually pumped to ultra-high vacuum levels for many hours, sometimes heating to 50-70°C, prior to analysis to remove absorbed atmospheric Ne. Cosmogenic Ne is extracted by heating each sample in one step (typically 1300°C) or in several

temperature steps in order to identify different Ne components. Active gases are removed by

exposure to hot metal surfaces, usually ZrAl getter pumps or Ti sublimation pumps. The heavy noble gases and residual active gases are absorbed on liquid nitrogen-cooled traps of either activated charcoal or sintered metal. Getter pumps operated at room temperature are used to absorb hydrogen. Helium is removed by absorbing the residual gases onto cryostatic cold head.

Neon isotope analysis is done in static mode using high-resolution magnetic sector mass spectrometers. These typically employ Nier-type electron impact ion sources tuned to optimise sensitivity for Ne ionisation. Ion beam intensities are measured using electron multiplier, usually in peak jumping mode. The mass spectrometers usually contain room temperature getter pumps and liquid nitrogen-cooled trap in order to minimize the contribution of interfering species during analysis.

On all but the latest high-resolution noble gas mass spectrometers isobaric interferences at mass 20, 21 and 22 require correction. This entails measurement of the abundances of masses 2, 16, 18, 19, 20, 21, 22, 40 and 44 were. Instrumental sensitivity and mass fractionation in the instrument is calculated from repeated analysis of aliquots of Ne in air. In many laboratories the CREU quartz standard (Ma et al. 2015; Vermeesch et al. 2015) is used as an internal check.

243 The abundance of cosmogenic ²¹Ne (²¹Ne_c) in samples is calculated assuming that all the measured ²⁰Ne (²⁰Ne_m) is atmospheric in origin from:

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$${}^{21}\text{Ne}_{c} = {}^{21}\text{Ne}_{m} \times \frac{\left[\left({}^{21}\text{Ne}/{}^{20}\text{Ne}\right)_{m} - \left({}^{21}\text{Ne}/{}^{20}\text{Ne}\right)_{air}\right]}{\left({}^{21}\text{Ne}/{}^{20}\text{Ne}\right)_{m}}$$
(5)

Unresolved contributions to 20 Ne occur as a result of $H_2^{18}O^+$, $^{40}Ar^{2+}$ and HF^+ . Interference at m/e = 20 from $H_2^{18}O^+$ are calculated from the measurement of $H_2^{16}O^+$ at mass 18. The $^{19}F^+$ signal was constant in mass spectrometer background, blank, calibration and sample measurements indicating that the fluorine is dominantly in the mass spectrometer. The absence of an effect of $H^{19}F^+$ on blank measurements implies that it is unimportant for measurement of samples 20 Ne. The dominant interference at m/e = 20 came from $^{40}Ar^{2+}$. The charge state ratio $^{40}Ar^{+/40}Ar^{2+}$ is governed by the partial pressure of H in the mass spectrometer ionization region. A first-order relationship between $^{40}Ar^{+/40}Ar^{2+}$ and H^+ beam size is recorded in many laboratories. The aim is to ensure that partial pressure of H remains constant throughout the analysis periods. Correction for $^{12}C^{16}O_2^{2+}$ at m/e = 22

is calculated from measured mass 44 ($^{12}C^{16}O_2^+$) using CO_2^{2+}/CO_2^+ determined by repeated measurements interspersed with sample measurements. Correction for $^{63}Cu^{3+}$ at ^{21}Ne can be done by measuring $^{65}Cu^{3+}$.

Although all three isotopes of neon need to be measured when calculating the cosmogenic ²¹Ne concentration, the uncertainty of measured cosmogenic ²¹Ne concentration mainly derives from the error of ²¹Ne isotope measurement, usually around 5%. In some cases of high ²¹Ne content, the error can go down to ~3%, which is nearly the lower limiting for cosmogenic ²¹Ne measurement uncertainty. In consequence, the measurement uncertainty is commonly larger for ²¹Ne than for the radionuclide ¹⁰Be or ²⁶Al. However, ²¹Ne is a stable isotope, so it eliminates the uncertainty from the determination of the decay constant of the radionuclide, which is known as ~1% for ¹⁰Be and ~3% for ²⁶Al (Balco and Shuster 2009a).

As to the precision of the final exposure age or erosion rate, it does not only depend on the precision of the parameters involved in data calculation, such as the measured isotope content, the decay constant of the isotope, the nuclide production rate and the scaling factor of production rate, but also depends on how well the model we adopt works in this situation, for example, the age uncertainty depends more on whether the assumption of the simple/complex exposure history is appropriate for what the sample has undergone. As a result, the total uncertainty is high in some geological situations if all error sources are considered, no matter for ²¹Ne or for radionuclides. However, cosmogenic ²¹Ne is produced and retained in a range of minerals and thus has the potential for constraining the timing of the exposure of surfaces that cannot be studied with ¹⁰Be and ²⁶Al. Furthermore, the unique possibility of stable ²¹Ne isotope for quantitative geomorphologic studies in the low-erosion region also endows it with huge potential to become a powerful tool which cannot be replaced by radionuclides or other techniques.

4. Neon isotope components in minerals

There are several sources of isotopically-distinct Ne in minerals that must be quantified before the concentration of cosmogenic Ne can be determined.

Air: Neon is present with a volume concentration of 18.2 ppm in air. It has three stable isotopes -

 20 Ne, 21 Ne and 22 Ne - and the atmospheric isotope ratio of 21 Ne/ 20 Ne and 22 Ne/ 20 Ne are 0.002959 \pm 0.000022 and 0.1020 \pm 0.0008, respectively (Eberhardt et al. 1965). Honda et al. (2015) recently redetermined the atmosphere Ne isotope composition, in particular revising the 21 Ne/ 20 Ne to be 0.002905 \pm 0.000003.

Atmospheric neon is ubiquitous in mineral samples, even those that have crystallised deep in Earth with no contact with air. It is likely that much of the air-derived Ne in minerals is absorbed onto mineral surfaces. However, extremely high concentrations are measured in quartz, often probably present in fluid inclusions (Graf et al. 1991; Niedermann et al. 1993). The mixture of atmospheric and cosmogenic neon in samples always lies on the "spallation line" in Fig. 1.

Nucleogenic Ne: Neon isotopes are produced by an array of nuclear reactions in minerals. The dominant source of nucleogenic Ne is produced by (α, n) reactions with 18 O (21 Ne) and 19 F (22 Ne), and (n,α) reactions with 24 Mg (21 Ne) and 25 Mg (22 Ne) (Graf et al. 1991; Niedermann et al. 1993). The α -particles are generated by radioactive decay of U and Th or spontaneous fission of 238 U. Thermal neutrons are generated during (α, n) reactions, and by cosmic ray spallation, termed cosmic ray thermal (CTN) neutrons (Dunai et al. 2007). The effect of these reactions is shown in Fig. 1.

The concentration of target elements F and Mg are negligible in quartz, so the dominant nucleogenic Ne production is ${}^{18}\text{O}(\alpha,n){}^{21}\text{Ne}$. Addition of this mono-isotopic component tends to produce data that plot to the right of the air-spallation line in the neon three-isotope diagram (Fig. 1). U and Th concentrations are typically low in pure quartz, but are often located in accessory mineral inclusions, e.g., biotite, apatite and zircon (Schäfer et al. 2002). α -particles recoil 10-40 μ m during production, and can be implanted into the outer few 10's μ m of grains. As the nucleogenic Ne is located in the crystal lattice of quartz, it is released with cosmogenic neon predominantly below 800°C. The removal of the rims of quartz grains by acid etching can reduce the nucleogenic component (Kohl and Nishiizumi 1992). On the other analysis of shielded samples with the exposed lithology can be used to quantify the contribution of non-cosmogenic nuclides (e.g., Margerison et al. 2005).

Crustal fluids are typically enriched in nucleogenic 21Ne and 22Ne, acquired by water-rock

interaction during fluid circulation in the crust (Kennedy et al. 1990). When these fluids are trapped as inclusions in minerals they impart nucleogenic Ne. The isotopic composition of the fluid depends on O/F ratio of the crust (Ballentine and Burnard 2002; Kennedy et al. 1990). Kennedy et al. (1990) has shown that crustal fluids typically have higher ²²Ne/²¹Ne ratios than that of cosmogenic Ne (Fig. 1) but it may not always be possible to distinguish cosmogenic neon. Usually the inclusion-hosted volatiles require higher release temperatures than cosmogenic Ne during stepwise heating experiments and it may be possible to separate the two components and allow cosmogenic Ne to be quantified (Niedermann et al. 1994).

Mantle Ne: Mantle-derived neon is enriched in ²⁰Ne and ²¹Ne relative to air Ne. It is typically incorporated in magmatic minerals at the time of rock formation, and usually dominantly present as melt or vapour inclusions. It should be clearly identified because of its distinct isotopic composition, whose trend is almost perpendicular to the spallation line (Fig. 1).

Inherited cosmogenic Ne: Cosmogenic Ne that has been produced prior to the exposure event that brought rocks to the surface will not, in contrast to the radionuclides, decay away. It will be isotopically indistinguishable from the modern cosmogenic Ne and, if not identified, will exaggerate exposure durations and lower erosion rates. While it is widely recognized in attempts to date the deposition of modern sediments (e.g., Ma et al. 2016), it is also likely to be a problem in the analysis of bedrock and boulders made of sedimentary or meta-sedimentary rocks which may be composed of detrital quartz that was exposed during erosion and transport of the grains that made up the sediment prior to lithification.

There are several approaches to determining the amount of cosmogenic Ne in a mineral. The commonest is to quantify and remove the atmospheric ²¹Ne:

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$$^{21}\text{Ne}_{\text{ex}} = ^{20}\text{Ne}_{\text{mea}} \times \left[\left(^{21}\text{Ne} / ^{20}\text{Ne} \right)_{\text{mea}} - \left(^{21}\text{Ne} / ^{20}\text{Ne} \right)_{\text{air}} \right]$$
 (6)

where ²¹Ne_{ex} is typically a mixture of *in situ* and crustal nucleogenic Ne, and cosmogenic Ne. The non-cosmogenic (mantle and nucleogenic) component of the excess ²¹Ne can be measured in deeply shielded samples of the same lithology as analysed for cosmogenic Ne. In modern sedimentary deposits the cosmogenic Ne generated during erosion and transport can be determined using the depth

profile method (Anderson et al. 1996; Hetzel et al. 2002a). If no post-depositional displacement of the deposit has occurred, and the transport and depositional processes were invariant in the sedimentary sequence, the concentration of cosmogenic Ne should decrease exponentially with depth, and a concentration versus depth plot should asymptotically approach the mean inheritance at depths of greater than 2 meters (Fig. 3) (e.g., Ma et al. 2016). Other approaches to correcting for nucleogenic Ne concentrations include calculating the *in situ* contribution from measured radioelement concentrations and the algorithm of Yatsevich and Honda (1997), and using the measured radiogenic ⁴⁰Ar and/or fissiogenic Xe isotopes to quantify the amount of trapped crustal ²¹Ne (e.g., Fujioka et al. 2005).

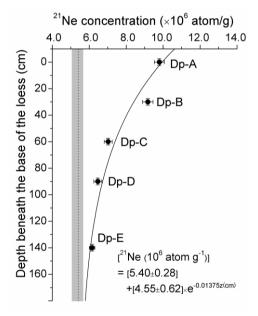


Fig. 3 The cosmogenic ²¹Ne concentration profile, showing the amount of ²¹Ne inheritance in terrace deposits (Ma et al. 2016). Error bars represent 1σ uncertainties. The black curve is the fitted exponential function. The dashed line and the margins of the light gray band correspond to the value of ²¹Ne inheritance and its error boundaries

5. Applications of cosmogenic Ne

5.1 Exposure age determinations under independent constraint on erosion rate

The presence of non-cosmogenic neon components in minerals adds uncertainty to the accuracy of exposure age determined by cosmogenic ²¹Ne. Nevertheless, cosmogenic ²¹Ne is produced and

retained in a range of minerals and thus has the potential for constraining the timing of the exposure of surfaces that cannot be studied with ¹⁰Be, ²⁶Al and ³⁶Cl.

Cosmogenic ³He, in olivine and pyroxene phenocrysts, has been widely applied for dating the eruption of Pleistocene and Holocene basalt lavas where ¹⁴C-datable material is not present (e.g., Foeken et al. 2009; Kurz 1986). Helium isotope determinations are easier than neon isotopes, and uncertainties associated with ³He-derived exposure ages are typically lower than those from cosmogenic ²¹Ne. In several studies cosmogenic ²¹Ne has also been measured on the same samples in order to confirm the ³He-derived exposure ages (Espanon et al. 2014; Fenton and Niedermann 2014; Gillen et al. 2010; Sarda et al. 1993; Staudacher and Allègre 1993). However, helium has only two isotopes, and thus the correction for non-cosmogenic components is complex. The three Ne isotopes allows a more effective resolution of the contribution from cosmogenic component (e.g., Staudacher and Allègre 1993), but it is still difficult to detect small amounts of cosmogenic ²¹Ne in the presence of relatively large amount of non-cosmogenic ²¹Ne.

As well as dating the eruption age of basalts, the cosmogenic noble gas isotopes can be used for determining the time that basalts were eroded. Cerling et al. (1994) determined the timing of flood events by measuring the cosmogenic ³He and ²¹Ne exposure ages of eroded basalt boulders and scoured bedrock of the Big Lost River on the Snake Piver Plain, Colorado. However, they noted that flood surface samples had a range of cosmogenic ³He and ²¹Ne concentrations. A small number of the surface samples appear to have slightly higher concentrations, consistent with the inheritance of cosmogenic nuclides that were produced prior the exposure of new surface during the flooding.

Cosmogenic radionuclides are commonly used for study of glacial histories (e.g., Ivy-Ochs et al. 1995). Cosmogenic ²¹Ne has been less commonly exploited in the study of glacial landforms yet holds huge potential. Graf et al. (2007) used cosmogenic ¹⁰Be and ²¹Ne to date erratic boulders from the top of the Montoz anticline in the Jura Mountains, Switzerland, to establish the pre-LGM glacial history. Strasky et al. (2009b) used ¹⁰Be and ²¹Ne to show the continuous long-term exposure history of the erratic boulders in southern Tibet, and investigated how sensitive the glacial system is to the North Atlantic climate forcing.

Cosmogenic ²¹Ne has also been used to determine the time of sediment deposition, such as river

terraces and alluvial fans. It is used in particular to confirm the simple exposure history of the sediment surface, ruling out periods of burial, and to correct for the cosmogenic ¹⁰Be that is generated during erosion of bedrock and transport of the clasts to the site of sedimentation. Phillips et al. (1998) reported cosmogenic ²¹Ne analysis of quartz sand derived from the Quaternary Bandelier Tuff to estimate the depositional ages of alluvium and soils on the alluvial plateau in New Mexico. They employed a depth profile to constrain the concentration of inherited ²¹Ne and determine the inheritance- and burial-corrected ²¹Ne exposure ages for individual surfaces of two terraces. Hetzel et al. (2002a) used ²¹Ne to determine the exposure age of fluvial terraces, and demonstrated the importance of determining the trapped (non-cosmogenic) Ne components in samples. Cosmogenic Ne measured in boulders on ancient marine terraces from northern Spain has been used to determine the tectonic history (Alvarez-Marrón et al. 2008). The long exposure history, 1-2 million years, includes periods of burial and suggested that the wave-cut platform was generated in the Pliocene, and then the uplift in response to crustal deformation.

Codilean et al. (2008) tested the hypothesis that there is spatial variation in erosion in a river catchment by measuring cosmogenic Ne in 32 quartz pebbles in the Gaub River catchment, Namibia. This data was compared to distribution predicted using measured catchment-averaged erosion rates and the mean slope values. The ²¹Ne concentrations span nearly two orders of magnitude and are skewed to low values consistent with predicted patterns, which imply that the measured distribution is a signature of the spatial variation in erosion rates.

Later, Kober et al. (2009) employed ¹⁰Be, ²⁶Al, and ²¹Ne in river sand quartz from Rio Lluta catchment in the arid area of northern Chile to determine catchment-wide denudation rates. Denudation rates derived from ²¹Ne gave lowest rates (12-17 m Ma⁻¹). By comparing the catchment-wide ²¹Ne-derived erosion rate with the headwater region erosion rates derived from ²¹Ne/¹⁰Be and ²⁶Al/¹⁰Be nuclide pairs, the authors highlighted ²¹Ne can be used to determine the temporal variation in sediment transport processes.

Antón et al. (2012) used cosmogenic ¹⁰Be and ²¹Ne to understand the Cenozoic history of the Duero River in the Arribes Gorge. Exposure ages for surfaces at various levels in the incised Duero River suggests that the final 2–300 m of fluvial incision in the Arribes Gorge occurred at a rate of 2–3

mm/yr over the last ~100 ka.

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The different half-lives of ¹⁰Be and ²⁶Al have been exploited to determine the duration of sediment burial, which can be used to determine sedimentation age and deposition rates (e.g., Balco and Stone 2005; Granger et al. 1997). Balco and Shuster (2009a) first reported the incorporation of ²¹Ne into the burial dating method to date cave sediments from the Riverbluff Cave in Springfield and the sedimentation age of Whippoorwill formation at the Pendleton clay pit near Pendleton, Missouri, USA. The authors established the feasibility of this method and showed the improved age range and accuracy of ¹⁰Be/²¹Ne nuclide pair comparing to that of ¹⁰Be/²⁶Al. Subsequently Davis et al. (2011) applied ¹⁰Be, ²⁶Al and ²¹Ne to date the burial age of the late Neogene sedimentary formations from central Jordan Valley, Israel. They obtained an age range from 3.5 to 5.3 Ma for Pliocene-early Pleistocene lacustrine sediments. This provided them with independent age constraints that were used in combination with magnetostratigraphy to understand the depositional history. Matmon et al. (2014) employed ²¹Ne, ¹⁰Be and ²⁶Al to date the sedimentary sequences deposited in the Sedom Lagoon to investigate the rift-forming and escarpment evolution along the Dead Sea fault, which reflects the Neogene development of the relative African-Arabian plate motion. Low-relief bedrock surfaces typically occur at high altitude, and are commonly interpreted as remnants of palaeo-surfaces that had originally a much larger extent (e.g., Clark et al. 2005). The bedrock peneplains can be used to infer significant amount of rock uplift after their generation at low elevation. However, the duration of such surfaces was preserved at high elevation and the rate at which they were modified by erosion are difficult to be quantified. Strobl et al. (2012) combined ²¹Ne and ¹⁰Be data measured in quartz clasts to confirm the simple exposure history with low denudation rates (5-11 m Ma⁻¹) of the peneplain in north of Nam Co of the southern Tibetan Plateau, which indicates the stability of exposure history and landscape evolution in this area. The potential of cosmogenic nuclide dating method not only provides the chronology of geological events, but in addition to offer the information about the rates of erosion and landscape evolution. Hetzel et al. (2002b) determined the long-term slip rate of the active faults in Central Asia (~0.35 mm a-1) based on the ²¹Ne, ¹⁰Be and ²⁶Al exposure ages of the offset landforms. They noted that the rate is two orders of magnitude slower than earlier estimates based on the assumption ages of the Last Glacial Maximum, which indicated the long-term preservation of the geomorphic features in Central Asia. Goethals et al. (2009b) investigated the impact of faulting on erosion rates in low-relief landscape in the Bishop Tuff, California, using in situ ²¹Ne in quartz from an ignimbrite with a known eruption age of 760 \pm 2 ka. Under the steady-state erosion assumption, they obtained a series of erosion rates of the sites with various fault activities, which suggested that tectonic activity does not affect erosion rates in a recognizable way in the low-relief landscape. Ma et al. (2016) employed ²¹Ne to measure the long-term average surface erosion rate at the Shapotou area in the northern margin of the Tibetan Plateau. They confirmed the steady-state erosion for cosmogenic ²¹Ne in the sediment of this area by measuring the ²¹Ne concentrations in quartz collected from a series of river terraces. The same ²¹Ne concentrations within measurement uncertainties in the highest two terraces only provided the information about the erosion rate (0.54-0.72 cm ka⁻¹) instead of the exposure duration of terraces. Perhaps the most exciting potential for cosmogenic ²¹Ne exposure dating is its application for understanding the surface history of other planets. The lack of need for performing chemistry can allow nuclide determinations in planet surfaces, ruling out the need for sample return missions. In the first study of this kind, Farley et al. (2014) used the Sample Analysis at Mars (SAM) instrument on the Curiosity rover to determine the concentration of cosmogenic ³He, ²¹Ne and ³⁶Ar in a mudstone from the floor of Gale Crater on Mars. They obtained consistent exposure ages from different isotopes with an average value of 78 ± 30 million years. Comparing with the formation age $(4.21 \pm 0.35$ billion years) of the rocks comprising the crater rim, which is constrained by K-Ar dating, the authors drew the scarp retreat hypothesis which caused the recent exposure of the mudstone after long-term burial from its deposition. The large uncertainty reflects the difficulty of making these measurements, and assumptions regarding the cosmic ray flux, but it shows a novel perspective for applications of cosmogenic noble gas isotopes in many new fields of Earth sciences and beyond in near future.

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5.2 Long-term landscape development in low erosion rate regions

The presence of air-derived Ne and nucleogenic ^{21,22}Ne in quartz, means that cosmogenic Ne requires approximately 10 times longer exposure than required for ¹⁰Be to obtain the equivalent precision and accuracy. Consequently most landscape elements from temperate climates that are

theoretically datable with cosmogenic ²¹Ne are prone to surface erosion. As a result, cosmogenic ²¹Ne concentrations in landscape elements that are older than a few tens of thousands of years rarely reflect the timing of formation. Landscape elements from low erosion rate regions, e.g. where climate is arid and/or dry, can be preserved for millions of years, though on these timescales processes of burial and exhumation may complicate the exposure histories. Cosmogenic ²¹Ne is ideally suited for understanding the evolution history and stability of erosional and depositional landforms. Here we highlight two extensively studied regions: Antarctica and the High Andes of Peru and Chile.

Cosmogenic ²¹Ne has proved powerful tool for reconstructing the Plio-Pleistocene history of Antarctica glaciations, specifically understanding the glacial volume fluctuations on both temporal and spatial scales, and addressing whether the Antarctic climate is decoupled from global climate variations or has been subject to similar fluctuations as took place elsewhere on the planet, i.e. the dynamic and stable glaciations variation hypothesis about the climatic change in Antarctica. The extremely low erosion rates in the Dry Valleys allows the cosmogenic nuclide exposure dating to be accurately applied over a much longer time range, which is up to millions of years. Several studies have focused on dating the glacial advances and establishing the association of the ancient glacial landforms with the Antarctic climate changes (e.g., Bruno et al. 1997; Kong et al. 2010; Schäfer et al. 1999; Van der Wateren et al. 1999). Many of these studies have exploited pyroxene from Ferrar dolerite bedrock and boulders rather than quartz.

Exposure ages of ~6.5 Ma for doleritic clasts from Sirius tillite at Mount Fleming constrained the uplift rates of the Transantarctic Mountain less than 170 m Ma⁻¹ (Bruno et al. 1997). Extraordinarily high surface exposure ages of 10 Ma for dolerite boulder from Mount Fleming obtained by Schäfer et al. (1999) and 11.2 Ma for the bedrock sample from Daniels Range obtained by Van der Wateren et al. (1999) support the hypothesis of a stable East Antarctic Ice Sheet since at least Late Miocene time and imply the decoupled climate variations of Antarctica from lower southern latitude regions.

Summerfield et al. (1999) first focused on measuring surface process rates of this extremely old landscape, and they estimated the long-term denudation rate in the Dry Valleys area of the Transantarctic Mountains, southern Victoria Land from the concentrations of cosmogenic ²¹Ne measured in quartz in both sandstones and granitic basement. The denudation rates were proved

sensitive to the landform elements, ranging from 0.26-1.02 m Ma⁻¹ for the rectilinear slopes down to 0.133-0.164 m Ma⁻¹ for the high-elevation surface sites. Oberholzer et al. (2003) combined ²¹Ne with ³He, and ¹⁰Be to limit Plio-Pleistocene glaciations in the Deep Freeze Range in northern Victoria Land. They measured surface exposure ages and erosion rates of both the bedrock and glacial erratic, and taking into account the erosion rate of 20 cm Ma⁻¹, they obtained a mid-Pliocene exposure age of the landscape surface in the McMurdo Dry Valleys, which indicates a constantly cold and hyperarid climate in this area. Surface cobbles on moraines in the Vemier Valley, located at the inland edge of the Dry Valleys region were analyzed to provide age control of the glacier activity by Staiger et al. (2006). They used a conservative erosion rate of 5 cm Ma⁻¹ in age calculations, and obtained a series of cosmogenic ²¹Ne exposure ages of old moraines of Ferrar dolerite. Subsequently, they used one of modern moraines cobbles sample to constrain the inheritance component in old moraines, and the cosmogenic age for the modern moraine suggested a value for nuclide inheritance of about 50 kyr, which was less than 5% of the total exposure time of the oldest boulders.

To address the response of Antarctic ice sheets to global climate changes, Oberholzer et al. (2008) measured ²¹Ne concentrations in the rock surfaces on nunataks in northern Victoria Land to investigate the duration of exposure generated by the glacial activity on the surfaces of nunataks. They gave the first absolute ages of 3.5 Ma of this area, which recorded the ice-free duration in this area. Strasky et al. (2009a) determined the exposure ages of the erratic boulders taking the surface erosion into account, and the dating results implied the low-amplitude Pleistocene variations in East Antarctic Ice Sheet, Victoria Land. Kong et al. (2010) applied the multiple nuclides ²¹Ne-¹⁰Be-²⁶Al to fit the surface exposure age of the bedrock on the Mount Harding to ~6.3 Ma, which recorded the time of the East Antarctic Ice Sheet retreated below the summit of the Mount Harding and suggested the important impact of the moisture transport from the Southern Ocean on the ice sheet expansion in the interior of East Antarctica. Altmaier et al. (2010) reported a series of rock exposure ages for the Queen Maud Land, Antarctica. They used ²¹Ne to date the samples in which radionuclide concentrations had nearly been saturated, and obtained minimum ²¹Ne exposure ages longer than 8 Ma, which excludes the presence of warm and humid climatic conditions during this period. Recently, Hein et al. (2016) present geomorphological evidence and multiple cosmogenic nuclide data from the

southern Ellsworth Mountains to suggest that the divide of the West Antarctic Ice Sheet has fluctuated only modestly in location and thickness for at least the last 1.4 million years.

The western central Andes are tectonically active and in situ cosmogenic nuclides allow the relationship between erosion, tectonics and climate on landscape evolution to be determined. The Atacama Desert is the driest place on Earth, and the onset of the aridification of the Atacama has been reported to be from at least the late Miocene (Dunai et al. 2005), which is indicated by the ²¹Ne ages ranging from 9 Ma to 37 Ma of individual clasts on depositional surfaces. To investigate whether the slow landscape evolution is characteristic of the entire Atacama, or only of specific regions, or landscape elements, Placzek et al. (2010) employed ¹⁰Be, ²⁶Al and ²¹Ne to quantify the exposure history of a wide variety of landscape elements, including bedrock, alluvial fans, stream sediment and boulders, along a transect across the Central Atacama. Their dating results as well as the field observations indicate the Central Atacama is more active than the Northern Atacama, and has remained geomorphically active well into the Pleistocene.

6. Perspective

In the past few decades *in situ* produced cosmogenic ²¹Ne has found a broad range of applications in many fields of Earth surface processes studies. Most of these studies have utilised quartz. However the retention of cosmogenic ²¹Ne in most other silicate and oxide minerals will no doubt open new areas of surface process research.

Developments in the ability to precisely measure smaller amounts of Ne will continue. Newly developed amplifiers with 10^{12} and 10^{13} Ω feedback resistors with fast response times and low noise characteristics offer the possibility of more stable measurement of small ion beam currents, and improved measurement precision. Combined Faraday-electron multiplier detectors will provide flexibility in analysis protocols. Increased resolution of new generation noble gas mass spectrometers is starting to provide separation of isobaric interferences and thus more accurate and precise measurement. This has, so far, extended as far as separation of 20 Ne $^{+}$ and 40 Ar $^{2+}$. Laser heating systems minimize the noble gases blanks and hence will lower detection limits, and allow precise and accurate measurements from smaller samples.

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553	Conflict of Interest:
554	The authors declare that they have no conflict of interest.
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