



## Cosmogenic noble gas paleothermometry



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### ABSTRACT

We present a theoretical basis for reconstructing paleotemperatures from the open-system behavior of cosmogenic noble gases produced in minerals at Earth's surface. Experimentally-determined diffusion kinetics predicts diffusive loss of cosmogenic  $^3\text{He}$  and  $^{21}\text{Ne}$  from common minerals like quartz and feldspars at ambient temperatures; incomplete retention has also been observed empirically in field studies. We show that the theory of simultaneous production and diffusion that applies to radiogenic noble gases in minerals—the basis of thermochronology—can also be applied to cosmogenic noble gases to reconstruct past surface temperatures on Earth. We use published diffusion kinetics and production rates for  $^3\text{He}$  in quartz and  $^{21}\text{Ne}$  in orthoclase to demonstrate the resolving power of cosmogenic noble gas paleothermometry with respect to exposure duration, temperature, and diffusion domain size. Calculations indicate that, when paired with a quantitatively retained cosmogenic nuclide such as  $^{21}\text{Ne}$  or  $^{10}\text{Be}$ , observations of cosmogenic  $^3\text{He}$  in quartz can constrain temperatures during surface exposure in polar and high altitude environments. Likewise,  $^{21}\text{Ne}$  retention in feldspars is sensitive to temperatures at lower latitudes and elevations, expanding the potential geographic applicability of this technique to most latitudes. As an example, we present paired measurements of  $^3\text{He}$  and  $^{10}\text{Be}$  in quartz from a suite of Antarctic sandstone erratics to test whether the abundances of cosmogenic  $^3\text{He}$  agree with what is predicted from first principles and laboratory-determined diffusion kinetics. We find that the amounts of cosmogenic  $^3\text{He}$  present in these samples are consistent with the known mean annual temperature (MAT) for this region of Antarctica between  $-25$  and  $-30^\circ\text{C}$ . These results demonstrate the method's ability to record paleotemperatures through geologic time.

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### 1. Introduction

Reconstructing past surface temperatures from continental settings is important for understanding how climatic processes, such as glacial cycles, and tectonic processes, such as orogenic-driven elevation change, have shaped terrestrial environments and landforms. The most widely applied methods for reconstructing past surface temperatures on continents involve measuring stable isotopes of oxygen in carbonates or ice. However, in the case of carbonates, traditional stable isotope methods require that the oxygen isotopic composition of water from which carbonate formed is known, which is rarely the case (e.g. Eiler, 2011; Kim and O'Neil, 1997; Rowley and Garzzone, 2007). In the past decade, measurements of multiply substituted isotopologues (“clumped isotopes”)

in carbonates have circumvented this limitation (Eiler, 2007; Ghosh et al., 2006). Nevertheless, temperatures calculated from carbonate clumped isotope measurements can be anomalously high (Quade et al., 2011) and vary as a function of seasonal parameters like precipitation (Peters et al., 2012) precluding simple interpretation in some cases. Leaf physiognomy (e.g. Forest et al., 1999; Gregory and McIntosh, 1996) and pollen distribution (e.g. Bartlein et al., 2011) have also been used to estimate terrestrial paleotemperatures. However, these bioclimatic proxies depend on numerous additional climate parameters, including precipitation, plant-available moisture, seasonality, and length of the growing season. All existing terrestrial paleotemperature proxies are limited by the abundance of specific minerals/fossils in the rock record and can suffer from poor preservation and diagenetic alteration (Eiler, 2011). Thus quantitative reconstruction of past terrestrial temperatures remains a major challenge despite the development of several paleotemperature proxies over the past few decades.

In this paper we describe a new paleotemperature proxy based on the open-system behavior of cosmogenic noble gases in common minerals.  $^3\text{He}$  and  $^{21}\text{Ne}$  were the first *in situ*-produced

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cosmogenic nuclides to be unambiguously observed in terrestrial materials (Craig and Poreda, 1986; Kurz, 1986; Marti and Craig, 1987). Since then, measurements of cosmogenic nuclides in minerals have transformed how Earth surface processes are studied and quantified (Granger et al., 2013).  $^3\text{He}$  and  $^{21}\text{Ne}$  are particularly attractive for studying surface processes. Since  $^3\text{He}$  and  $^{21}\text{Ne}$  are both stable, they can be used to quantify exposure histories that are long relative to the half-lives of cosmogenic radionuclides like  $^{10}\text{Be}$  and  $^{26}\text{Al}$ . Cosmogenic  $^3\text{He}$  and  $^{21}\text{Ne}$  also have high production rates in terrestrial materials, minimizing the amount of material needed for analysis, and noble gas measurements are faster and cheaper than measurements of cosmogenic radionuclides by accelerator mass spectrometry. Cosmogenic  $^{21}\text{Ne}$  in quartz, olivine, pyroxene, and amphibole and cosmogenic  $^3\text{He}$  in the latter three phases have been extensively used to study geomorphic and geologic processes (e.g. review by Dunai, 2010). Empirical observations (Bruno et al., 1997; Cerling, 1990; Ivy-Ochs et al., 2007; Schäfer et al., 1999), and in some cases measured diffusion kinetics (Cherniak et al., 2014; Gourbet et al., 2012; Shuster and Farley, 2005; Trull et al., 1991), indicate that these cosmogenic noble gas–mineral pairs exhibit quantitative retention at Earth surface temperatures.

In contrast, cosmogenic  $^3\text{He}$  and  $^{21}\text{Ne}$  experience significant diffusive loss over geologic time at Earth's surface in quartz and feldspars, respectively. As a result, these cosmogenic noble gas–mineral pairs have been almost entirely avoided in surface process studies. Incomplete retention of cosmogenic  $^3\text{He}$  was first inferred from comparisons of  $^3\text{He}$ -based exposure ages with  $^{10}\text{Be}$  or  $^{26}\text{Al}$  ages from the same quartz samples. In diverse settings, the  $^3\text{He}$  ages were systematically younger (Brook and Kurz, 1993; Brook et al., 1993; Cerling, 1990; Trull et al., 1995). Experimentally-determined diffusion kinetics of  $^3\text{He}$  in quartz broadly agrees with these observations and indicates that significant proportions of  $^3\text{He}$  can be lost from quartz over time, even at subzero temperatures (Shuster and Farley, 2005). Although one study showed that  $^3\text{He}$  could be quantitatively retained in very large quartz grains with <100 ka of exposure in Antarctica (Brook et al., 1995), the only published cosmogenic  $^3\text{He}$ –quartz measurements since the mid-1990s were used to screen for relatively old versus young erratics and sub-select samples for  $^{10}\text{Be}$  analyses (Ackert Jr. et al., 2011). Observations of cosmogenic  $^{21}\text{Ne}$  in feldspars (Bruno et al., 1997; Kober et al., 2005) and experimentally-determined diffusion kinetics of Ne (Gourbet et al., 2012) suggest that feldspars can also experience diffusive loss of cosmogenic  $^{21}\text{Ne}$  at Earth's surface, depending on mineralogical composition. Sanidine, which apparently exhibits quantitative retention of  $^{21}\text{Ne}$  (Gourbet et al., 2012; Kober et al., 2005), is the only feldspar that has been used to study surface processes (Ivy-Ochs et al., 2007; Kober et al., 2007).

Although avoided in cosmogenic nuclide studies, the open-system behavior of radiogenic noble gases has been extensively studied and utilized as a method for reconstructing thermal histories of minerals and rocks in the upper lithosphere (Harrison and Zeitler, 2005). Widely applied methods of noble gas thermochronology, such as the  $^{40}\text{Ar}/^{39}\text{Ar}$  system in feldspars and the (U–Th)/He system in apatite and zircon, record both time, through the production of noble gas atoms via radioactive decay, and temperature, through their thermally-activated diffusive loss. In principle, cosmogenic noble gas–mineral pairs exhibiting open-system behavior also record the thermal histories of rocks that can be quantified using the same theoretical framework as radiogenic noble gases. Since cosmogenic nuclide production occurs almost entirely within the uppermost few meters of the Earth's surface, cosmogenic noble gas–mineral pairs ought to record thermal histories of local surface environments.

Here we present calculations to test whether the open-system behavior of cosmogenic  $^3\text{He}$  in quartz and  $^{21}\text{Ne}$  in feldspars can be

used to quantify past temperatures at Earth's surface. We utilize published production rates and diffusion kinetics of these cosmogenic noble gas–mineral pairs, although future paleothermometry applications will warrant tighter constraints on both of these variables. We show that with relatively simple models based on first principles we can predict the observed abundances of cosmogenic  $^3\text{He}$  in an example set of quartz samples from Antarctica with simple Holocene exposure histories. This work focuses on  $^3\text{He}$  and  $^{21}\text{Ne}$  in quartz and feldspars, respectively, but the theoretical framework presented here can be extended to any cosmogenic noble gas–mineral system and to other planetary bodies and higher temperature regimes (e.g. Shea et al., 2012; Shuster et al., 2010; Suavet et al., 2013). Further, this cosmogenic nuclide-based paleothermometer, which is characterized by fundamentally different principles and assumptions from preexisting paleotemperature proxies, may be particularly useful for studying surface processes where pre-existing proxies cannot be applied, or provide important tests for internal consistency in cases where they can.

## 2. Theoretical background

### 2.1. Simultaneous production and diffusion

The theory and mathematics describing the open-system behavior of noble gases in minerals have been extensively developed in the field of noble gas thermochronology; we present a brief review of fundamental concepts here. The concentration of a noble gas  $M$  simultaneously being produced and diffusing from a mineral changes as a function of time  $t$  according to:

$$\frac{\partial M}{\partial t} = \frac{D(T)}{a^2} \nabla^2 M + P_M \quad (1)$$

where  $D$  is diffusivity,  $\nabla^2$  is the second-order spatial derivative or Laplacian,  $a$  is the dimension of the diffusion domain, and  $P_M$  is the mineral-specific production rate of  $M$ . Eq. (1) requires that diffusion of noble gases in the mineral is Fickian (i.e., atoms move from regions of high to low concentration with a flux proportional to the concentration gradient) and isotropic. Published diffusion experiments suggest that these conditions are met for noble gas diffusion in quartz (Shuster and Farley, 2005) and orthoclase (Gourbet et al., 2012). Solutions to Eq. (1) must also satisfy the boundary condition that the concentration of  $M$  at  $a$ , the outer boundary of the diffusion domain, is zero. This condition is effectively valid for minerals when the diffusion domain is equivalent to the grain size, as diffusivities along grain boundaries are typically several orders of magnitude greater than within the mineral grains themselves (e.g. Dohmen and Milke, 2010), and the surrounding environment is nearly an infinite reservoir for rare nuclides. Abundances of cosmogenic  $^3\text{He}$  in different grain sizes sharing a common exposure history indicate that the diffusion domain and grain size are equivalent for quartz (Brook et al., 1993). For thermally-activated diffusion of noble gases in minerals, a large body of research (e.g. review by Baxter, 2010) shows that the diffusivity  $D$  depends exponentially on temperature according to the Arrhenius relationship:

$$\frac{D(T)}{a^2} = \frac{D_0}{a^2} \exp\left(\frac{-E_a}{RT(T)}\right) \quad (2)$$

where  $D_0$  is the diffusivity at infinite temperature,  $E_a$  is the activation energy,  $R$  is the gas constant and  $T$  is temperature. The diffusion parameters  $D_0$  and  $E_a$  are specific to a particular noble gas–mineral pair and can be determined empirically via laboratory experiments. If both temperature (and therefore diffusivity) and the production rate remain constant through time, and if we assume the diffusion domain has a spherical geometry, Eq. (1) has the analytical solution described by Wolf et al. (1998):

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