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Cosmogenic ^{3}He and ^{21}Ne measured in quartz targets after one year of exposure in the Swiss Alps

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ABSTRACT

All currently used scaling models for Terrestrial Cosmogenic Nuclide (TCN) production rates are based on neutron monitor surveys. Therefore, an assumption underlying all TCN studies is that production rates are directly proportional to secondary cosmic ray intensities for all cosmogenic nuclides. To test this crucial assumption, we measured cosmogenic ^{3}He and ^{21}Ne in artificial quartz targets after one year of exposure at mountain altitudes in the Swiss Alps. The targets were inconel steel tubes containing 1 kg of artificial quartz sand (250–500 μm), degassed for one week at 700 °C in vacuum prior to exposure. From August 2006 until August 2007, ten of these targets were exposed at five locations in Switzerland and Italy: Zürich (556 m), Davos (1560 m), Säntis (2502 m), Jungfraujoch (3571 m), and Monte Rosa (4554 m). Additionally, a sixth set of two blank targets was kept in storage and effectively shielded from cosmic ray exposure. Cosmogenic noble gases were measured at room temperature and at 700 °C. Up to 9% of the cosmogenic ^{3}He was measured in the cold step, indicating that ^{3}He diffuses out of quartz at room temperature on short time scales. The remaining ^{3}He and all ^{21}Ne were released at 700 °C, as shown by a repeat measurement at 800 °C for the Monte Rosa target, which yielded no additional cosmogenic helium and neon. As expected, the Monte Rosa target contained the highest cosmogenic nuclide content, with $1.56 \pm 0.07 \times 10^6$ atoms of excess ^{3}He and $4.5 \pm 1.2 \times 10^5$ atoms of excess ^{21}Ne (all errors are 2σ). The raw measurements were corrected for non-atmospheric blanks, shielding (roof + container wall), tritiogenic helium and solar modulation (normalised to the average neutron flux over the past five solar cycles). The ^{3}He/^{21}Ne production rate ratio of 6.8 ± 0.9 indicates that cosmogenic ^{3}He production by the container walls is negligible. The main goal of the artificial target experiment was to determine the production rate attenuation length. Because all our targets had an identical design and were exposed under identical conditions, all systematic errors cancel out in the calculation of an attenuation length. Our best estimates for the ^{3}He and ^{21}Ne attenuation lengths are 134.8 ± 5.9 g/cm^2 and 135 ± 25 g/cm^2, respectively, agreeing very well with currently used scaling models. We conclude that TCN production rates are indeed proportional to neutron monitor count rates, and that ^{3}He and ^{21}Ne production rates follow the same altitudinal scaling relationships as the cosmogenic radionuclides. Finally, the measurements were scaled to sea level and high latitude using the empirical attenuation length, yielding weighted mean production rates of 107.6 ± 6.6 at/g/yr for ^{3}He and 15.4 ± 2.1 at/g/yr for ^{21}Ne. Despite the significant uncertainties associated with the corrections for shielding, solar modulation and especially the ^{3}He/^{3}H branching ratio, these estimates are in good agreement with production rates derived from long-term exposure experiments at natural calibration sites and physics-based simulations.

1. Introduction

All currently used scaling models for Terrestrial Cosmogenic Nuclide (TCN) production rates are based on neutron monitor surveys (Dunai, 2001; Lal, 1991; Stone, 2000; Dunai, 2000; Desilets and Zreda, 2003; Pigati and Lifton, 2004; Lifton et al., 2008). Therefore, an assumption underlying all cosmogenic nuclide studies is that production rates are directly proportional to secondary cosmic ray intensities for all cosmogenic nuclides. Several efforts are underway to test this crucial assumption by TCN production rate calibrations in the

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framework of the CRONUS-EU and CRONUS-Earth initiatives. The bulk of this work is done on landforms of known age (Desilets and Zreda, 2006). These so-called 'natural calibration targets' are the method of choice for the calculation of accurate TCN production rates integrated over millennial time scales but are, unfortunately, often affected by poorly constrained factors such as shielding and erosion. It is notoriously hard to find vertical transects of natural calibration sites that allow the calculation of production rate attenuation lengths. Herein lies the complementary strength of artificial calibration targets. Because the exposure conditions of the latter are either known or constant, all systematic errors cancel out in the calculation of a production rate attenuation length. We here present the first results of an artificial target experiment measuring, for the first time, cosmogenic ^{3}He and ^{21}Ne in quartz after one year of exposure at mountain altitudes in the Swiss Alps. Previous artificial target experiments have mainly focused on water (Lal et al., 1960; Nishiizumi et al., 1996; Brown et al., 2000; Graham et al., 2000), although one pilot experiment used a silicate glass (Graf et al., 1996). We used quartz as the target material, because it is the most commonly used mineral for exposure dating and both cosmogenic helium and neon are produced and retained in the target container.

Our project has a history of more than ten years. A first target design was developed back in 1997. These were stainless steel containers with 14 cm radius and 45 cm height, filled with 4 kg of industrial quartz sand of natural origin (Fluka, no. 83340; Schäfer, 2000; Kober, 2004). The targets were heated to >800 °C under vacuum for a week in order to ensure complete degassing prior to exposure, and double sealed with a valve and copper tube clamp to prevent atmospheric leaks during exposure. Fourteen of these targets were exposed at seven different locations for two to four years. Two of them were measured, one unexposed blank target and one target that had been exposed at Jungfraujoch, at an altitude of 3571 m. The pilot experiment was aborted after the neon and helium compositions of these two targets were found to be a mixture of cosmogenic and other components (Kober, 2004). There are two reasons why the first target design failed. The presence of a 'trapped' component (neon plotting above the mixing line between atmospheric and cosmogenic components, the so-called 'spallation line'; Niedermann, 2002) indicates that pre-exposure degassing was insufficient, and that the quartz did not reach the 600 °C degassing temperature of neon (Niedermann, 2002). The presence of a 'nucleogenic' component (neon plotting below the spallation line) indicates that despite the purity of the industrial quartz sand, it still contained sufficient alpha producing U and Th (120 and 172 ppb, respectively) to compromise the helium and neon measurements. These observations led to the development of a second generation target design.

The effectiveness of the revised target design was verified in a custom-built prototype container (Section 1). Ten of these targets were exposed at different elevations in the Swiss Alps, at altitudes ranging from 556 to 4554 m (Section 2). Cosmogenic ^{3}He and ^{21}Ne were measured after one year of exposure, using a custom-built mass spectrometer and an optimised measurement routine (Section 3). Data reduction included corrections for non-atmospheric blanks, shielding, solar modulation, and tritiogenic helium (Section 3). ^{3}He and ^{21}Ne were measured in two steps at room temperature and at 700 °C. Most of the ^{3}He and all of the ^{21}Ne were measured in the hot step (Section 4). The altitude dependency of the TCN production rates was quantified by plotting them against atmospheric depth, yielding attenuation lengths that are in perfect agreement with existing scaling models (Section 5). Production rates were scaled to sea level and high latitude and agree well with previous determinations on natural calibration sites (Section 6). We conclude this paper with an outlook to the future, when duplicate artificial targets will be used to determine the ^{3}He/^{3}H branching ratio and we will monitor cosmogenic noble gas production rates over an entire solar cycle (Section 7).

2. Methods

2.1. Target design

The first generation targets suffered from sub-optimal degassing and impure quartz (Section 1). Both of these problems were addressed in the second generation target design. To eliminate the trapped neon component and ensure optimal degassing, the radius of the seamless stainless steel (grade 1.4301) canisters was reduced from 14 to 6 cm (Fig. 1.c), and in order to eliminate the nucleogenic component, we used artificially grown quartz crystals of optimal purity (supplied by Morion Company, USA), which were crushed to 250–500 µm sand size (Fig. 1.a), and rinsed with water and acetone. Gamma ray spectrometry measurements revealed U and Th concentrations <16 and <49 ppb, respectively, which is below the detection limit of the method and also below the levels measured in the Fluka quartz sand (Strasky, 2008). To verify the effectiveness of the new target design, two thermocouples were installed in a prototype container filled with 800 g quartz sand (Fig. 1.c). After a heating period of ~2.5 h at an external temperature of 900 °C, the temperature reached by the quartz in the innermost part of the container was ~850 °C, well above the degassing temperatures of helium and neon (Niedermann, 2002) (Fig. 1.d). To reduce the blank, the external temperature for the actual target measurements was later reduced to 700 °C, which should yield ~650 °C quartz temperatures. As was the case for the first generation (Section 1), also the second generation targets were double sealed by a 'bellows-sealed' Swagelok® valve connected to a copper tube with a stainless steel pinch-off clamp.

2.2. Pre-treatment and installation

One kg of the artificial quartz sand was degassed inside the targets for one week at 700 °C in vacuum prior to exposure using a custom-built furnace (Fig. 1.b). The targets were rolled in bubble wrap and placed in fibreglass cable trays for protection against the weather, and were installed in a horizontal position to minimise self-shielding. In August of 2006, two targets were exposed at each of five locations: Zürich (556 m), Davos (1560 m), Säntis (2502 m), Jungfraujoch (3571 m), and Monte Rosa (4554 m). All of these locations (except for Monte Rosa) are meteorological observatories of the Swiss Federal Office of Meteorology and Climatology (MeteoSwiss), which were kept snow-free during the winter of 2006–2007. The Zürich, Davos, and Säntis targets were installed outside and secured to the railings of the meteorological equipment. Because of the extremely high wind speeds at the Jungfraujoch and Monte Rosa sites, those targets were kept inside. Additionally, a sixth set of two blank targets was stored in the basement of a 10-storey building housing the ETH noble gas laboratory, ~15 m below street level, and effectively shielded from cosmic ray exposure. Exactly one year later, the targets were retrieved and subsequently measured.

2.3. Measurements

Even in 1 kg of quartz and at mountain altitudes, the expected amounts of cosmogenic gas are extremely low, on the order of tens of thousands of atoms in a volume of more than 4 l. To measure such minute amounts of noble gases, we used a unique kind of mass spectrometer developed at ETH-Zürich, which is equipped with a compressor source (Baur, 1999). The compressor consists of a magnetically levitated rotor, spinning at 1500 Hz, which forces the gas along spiral grooves in the inner wall of the stator. The neutral gas then enters the ionization volume and gets accelerated towards the magnet and ion detectors. The compressor source acts as a pump, consuming a much larger portion of the sample gas than a conventional mass spectrometer, and resulting in a two orders of magnitude gain in sensitivity.