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Research paper

The effects of a hydrogen-rich ground cover on cosmogenic thermal neutrons: Implications for exposure dating☆

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ABSTRACT

We present results of thermal neutron flux measurements in experimental granite piles that were tailored to study the effect of hydrogen-rich covers on that flux. We find that hydrogen-rich covers (polyethylene, water), used as proxies for snow, dead and/or live plant matter, increase the thermal neutron flux in an underlying rock surface significantly, as compared to the state without cover. The rock serves as the main source for thermal neutrons, the hydrogen-rich cover as a neutron reflector. In situations where the thickness of such a cover would be negligible in terms of high-energy neutron (>10 MeV) attenuation, e.g. 2–3 cm water equivalent cover, a significant enhancement of the thermal neutron flux (factor >2.5 ± 0.5) can be achieved. This increase is made up of three components (Masarik et al., 2007): (1) reflected thermal neutrons (albedo neutrons), (2) moderated fast neutrons from the ground, and (3) moderated fast neutrons from the atmospheric cascade (Masarik et al., 2007). The higher thermal neutron flux increases the production rates of those cosmogenic nuclides that have a significant thermal neutron production pathway (^{3}He, ^{36}Cl, ^{41}Ca). Ignoring this effect in situations where target nuclei (^{6}Li, ^{35}Cl, ^{40}Ca) are abundant will severely underestimate production rates. The effect of hydrogen-rich ground cover on the thermal neutron flux has the potential to be used for studies that are aimed at reconstructing the persistence of past plant/snow cover. Isotopic ratios of spallogenic versus predominantly thermal neutron produced nuclides, would reveal the presence or absence of hydrogen-rich cover in the past as compared to the present-day situation.

1. Introduction

The energy spectrum of the secondary neutrons of the cosmic ray cascade is in equilibrium from a few hundred metres above the Earth's surface to several kilometres in the atmosphere, i.e. the neutron energy spectrum remains essentially unchanged (Goldhagen et al., 2003, 2002; Kowatari et al., 2005; Lal and Peters, 1967). However, near the Earth's surface both neutron production and scattering properties change profoundly, leading to a non-equilibrium situation (Hendrick and Edge, 1966; Kastner et al., 1970; Kodama, 1983; Masarik et al., 2007; O'Brien et al., 1978). In particular, the flux of thermal neutrons increases dramatically, by approximately one order of magnitude, due to increased production in the solid Earth and effective moderation by water/moisture at the ground level (Hendrick and Edge, 1966; Kastner et al., 1970; Kodama, 1983; Masarik et al., 2007; O'Brien et al., 1978). This perturbation of the thermal neutron flux is measurable up to 100 m above ground (Hendrick and Edge, 1966); the attenuation of this perturbation occurs due to the large reaction cross section of ^{14}N for thermal neutrons and the high abundance of nitrogen in air (Hendrick and Edge, 1966). Over the length-scale of 100 m, the atmosphere (at sea level) can be considered as an effective sink for thermal neutrons emanating from the Earth's surface.

The increased thermal neutron flux above the Earth's surface derives from secondary neutrons that are produced and moderated in the uppermost ~50 g/cm^2 of the ground and then 'leak' back into the atmosphere (Masarik et al., 2007; O'Brien et al., 1978; Phillips et al., 2001). The scattering of neutrons by protons (hydrogen nuclei) is a particularly effective way by which higher energy level neutrons are moderated to thermal neutrons (Reuss, 2008). As a source of hydrogen nuclei the water content of rocks or soils is, therefore, a crucial parameter for determining the thermal neutron flux near the ground/air interface (Phillips et al., 2001).

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Once ‘thermal’, neutrons are in energy equilibrium with their surroundings they are equally likely to become accelerated or decelerated in response to collisions with other nuclei (Reuss, 2008) and the resulting direction of movement is random (Phillips et al., 2001). The attenuation of the thermal neutron flux in rocks/soils occurs due to neutron capture reactions by elements that constitute the rock/soil matrix (Phillips et al., 2001). These neutron capture reactions may produce cosmogenic nuclides used in Earth science applications. Cosmogenic nuclides with significant production by thermal neutrons are ^{3}He, ^{36}Cl and ^{41}Ca; they are produced by neutron capture by ^{6}Li, ^{35}Cl and ^{40}Ca, respectively (Audi et al., 2003; Dunai, 2010; Dunai et al., 2007; Liu et al., 1994; Nishiizumi et al., 2000).

Any hydrogen-rich layer (e.g. snow, ice, plant matter) covering a neutron source (e.g. rock or soil) can effectively reflect thermal neutrons back into the source, i.e. it has a large albedo for emitted thermal neutrons. For example, a thick (>∼6–8 cm) water layer will reflect 80% of incoming thermal neutrons back into their source (Reuss, 2008). The efficiency of natural neutron reflectors is largely governed by their hydrogen concentration and neutron absorption properties (Csikai and Buczkó, 1998; Király and Csikai, 2000; Reuss, 2008). Furthermore, a hydrogen-rich layer on the ground may increase the thermal neutron flux beneath by returning fast neutrons (e.g. neutrons from evaporation reactions in the ground, 1–10 MeV) moderated to thermal energies, which may otherwise leak to the atmosphere (Kodama, 1983; Masarik et al., 2007). Also incoming secondary neutrons from the atmospheric cascade will be moderated by a hydrogen-rich layer and increase the thermal neutron flux beneath it (Masarik et al., 2007). The increase of the thermal neutron flux in the ground near the ground/air interface, by a hydrogen-rich layer at that interface, is therefore made up of three components (Masarik et al., 2007): (1) reflected thermal neutrons, (2) moderated fast neutrons from the ground, and (3) moderated fast neutrons from the atmospheric cascade (Masarik et al., 2007). Conversely, the thermal neutron flux in the ground may be reduced to some extent by hydrogen-rich layers which shield the ground from incoming cosmic rays (Hatton and Carmichael, 1964).

The effect of hydrogen-rich layers on the cosmogenic thermal neutron flux in solids has been experimentally determined (neutron source: lead; moderator/reflector: polyethylene; Hatton and Carmichael, 1964) and numerically modelled (neutron source: average terrestrial body; moderator/reflector: water; Masarik et al., 2007). These results indicate that there is a potentially large (1.3–4-fold) increase in the thermal neutron flux (Hatton and Carmichael, 1964; Masarik et al., 2007) with only thin covers of neutron reflectors (Hatton and Carmichael, 1964). Such an enhanced thermal neutron flux will necessarily increase cosmogenic production of nuclides with a significant thermal neutron capture pathway. These studies (Hatton and Carmichael, 1964; Masarik et al., 2007) demonstrate that the regular approach of considering any mass-cover (of hydrogen-rich or not) as an attenuator of the cosmic ray flux, as commonly undertaken for exposure dating (cf. Gosse and Phillips, 2001) is probably incorrect for nuclides with a significant thermal neutron capture production pathway (^{3}He, ^{36}Cl and ^{41}Ca) and a hydrogen-rich ground cover (snow, ice, plant matter, moist soil). While the previous studies (Hatton and Carmichael, 1964; Masarik et al., 2007) clearly show the importance of hydrogen-rich cover, the translation for the use in applications of in situ produced cosmogenic nuclides remains difficult. The extent to which lead (Hatton and Carmichael, 1964) can be used as proxy for rock is currently unclear, while the numerical calculations of Masarik et al. (2007) consider only thick (20 cm water) covers and would benefit from experimental verification.

In this study we investigate the effect of a hydrogen-rich cover for rocks of granitic composition. Granitic rocks contain minerals often used for cosmogenic applications, utilizing ^{3}He and ^{36}Cl, and other minerals which show potential for future development utilizing ^{41}Ca (Dunai, 2010; Farley et al., 2006; Gosse and Phillips, 2001). In addition granite contains abundant concentrations of target nuclides (^{6}Li, ^{35}Cl and ^{40}Ca), with significant thermal neutron capture cross-sections producing these cosmogenic nuclides. Our experimental results confirm the magnitude of effects described by Hatton and Carmichael (1964) and Masarik et al. (2007), and demonstrate their importance for common situations encountered in applications of in situ produced cosmogenic nuclides to Earth sciences.

2. Methods and experimental setting

We measured the thermal neutron flux in two identical granite piles. One pile was used to monitor changes in the cosmic ray flux and the environmental neutron field (Kodama, 1983; Zreda et al., 2008), the other was modified by placing polyethylene, water (of various thickness) and/or further granite on top. Thus, in response to these modifications, the relative changes in thermal neutron flux rates, corrected for external flux variations, were obtained. The basic dimensions of both granite piles were 120 × 130 × 60 cm (Fig. 1), the piles were built from individual

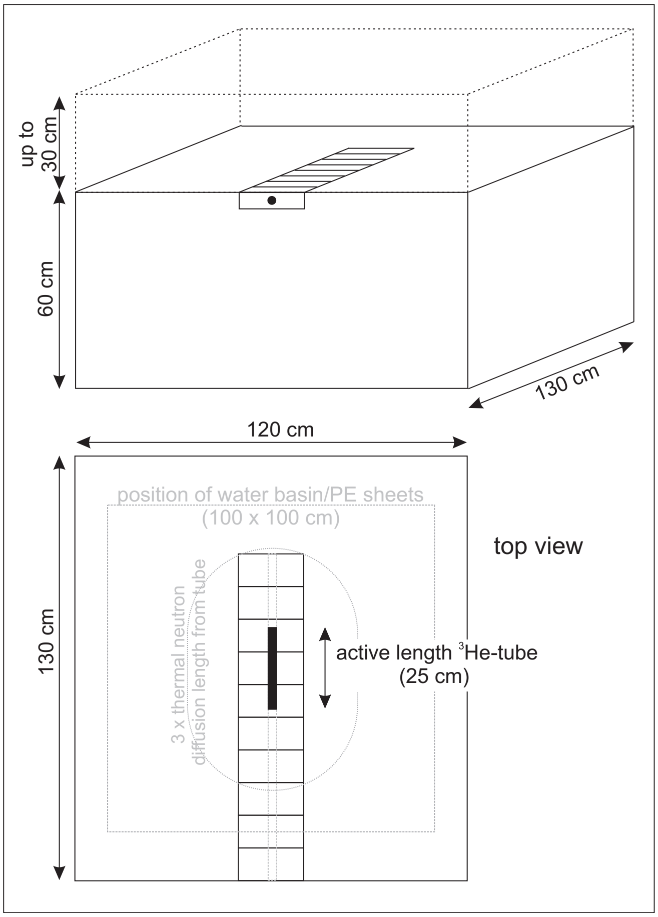

Fig. 1. Sketch of the principal dimensions of the two granite piles in oblique and top view. The core-drilled granite blocks that held the neutron counting tubes are shown; the other blocks are not shown individually but had the same dimensions 5 × 10 × 20 cm, only without holes. In the oblique view the position of the additional granite blocks put on top of one of the piles, to mimic a depth profile, is shown in stippled outline. In the top view the centred positions of the water basin and PE sheets are indicated, as is the limit of thrice the thermal neutron diffusion length around the active length of the tube.