

SUPPLEMENTARY METHODS: COSMOGENIC NOBLE GAS DEPLETION IN SOILS BY WILDFIRE HEATING

1. Sample preparation and quartz isolation.

We separated quartz from soil samples by disaggregating and wet-sieving to a 0.25-0.5 mm grain size range, followed by cleaning in water in a sonic bath, soaking in dilute HCl to remove iron oxides, and repeated rinsing. This resulted in a sample composed primarily of residual quartz grains remaining after advanced weathering of the soil. We then etched the remaining material in 5% HF at room temperature until the quartz appeared visibly clean.

Although external heat was not applied during sonic cleaning, both sonic cleaning and subsequent oven-drying of samples after rinsing heated samples above room temperature, which could cause loss of helium from quartz. Although of course this could easily be prevented by performing all sample preparation at room temperature, our original intention was to measure only ^{21}Ne (which is not subject to loss at the temperatures involved). ^3He measurements were only carried out after we obtained the ^{21}Ne results. Although it is not possible to calculate ^3He loss exactly without knowledge of the entire thermal history of the sample prior to collection, we performed this calculation with limiting assumptions (complete retention of ^3He produced during the exposure history, followed by 24 hr at 60° C) and found that no more than 6% of ^3He present would be expected to be lost during sonic treatment. As this is significantly less than the uncertainty in the ^3He concentration measurements, we did not make any correction for ^3He loss during sample preparation.

2. ^3He and ^{21}Ne measurements.

We measured concentrations of He and Ne isotopes in quartz samples by noble gas mass spectrometry (NGMS) on the “Ohio” NGMS system at the Berkeley Geochronology Center. 140-170 mg aliquots of clean quartz were encapsulated in Ta packets and heated under vacuum by an 810 nm diode laser. Released gases were then purified by reaction with SAES getters at room temperature, followed by cryogenic separation of remaining noble gases on an activated charcoal trap and introduction of the desired gas into the mass spectrometer. The mass spectrometer is a MAP-215-50 sector field mass spectrometer with updated detectors and electronics. ^3He and all Ne isotopes were measured using a Channeltron-type multiplier operated in pulse-counting mode.

He and Ne were measured on separate aliquots. He was extracted in one heating step of 15 min at 800° C, followed by freezing of He onto the cryotrap at 12.5° K and release at 33° K. As discussed by, e.g., Balco et al. (2016) and Gribenski et al. (2022), this heating schedule has been found to completely degas ^3He from quartz, but does not completely extract ^4He . The most likely reason for this is that some ^4He is contained in more retentive, U/Th-rich, inclusions. As this study is only concerned with cosmogenic ^3He , we did not continue heating to higher temperature to quantitatively measure total ^4He , so ^4He concentrations are not reported. Further details of ^3He measurements at BGC have been recently reported in Gribenski et al. (2022) and Tremblay et al. (2019). Full process blanks measured by heating empty Ta packets interspersed with the Cougar Pass samples were 25000 ± 28000 atoms ^3He , and uncertainties on ^3He concentrations include measurement uncertainty derived from counting statistics, an uncertainty on machine sensitivity derived from replicability of measurements on helium gas standards, and uncertainty in blank subtraction, added in quadrature. ^3He concentrations are reported in Table S1.

Ne was extracted in two heating steps of 15 min at 850° C and 12 min at 1100° C, with separation from He and Ar by freezing of Ne onto the cryotrap at 33° K and release at 70° K. Mass-spectrometric measurement of Ne isotopes involves use of a ^{39}Ar spike to correct for $^{40}\text{Ar}^{++}$ observed on mass 20, as described by Balco and Shuster (2009). After correction of resulting ^{20}Ne , ^{21}Ne , and ^{22}Ne signals for mass spectrometer background, we computed excess ^{21}Ne relative to atmosphere as $N_{21,\text{xs}} = N_{21,\text{total}} - (N_{20,\text{total}} R_{21/20,\text{a}})$, where $N_{21,\text{xs}}$ is excess ^{21}Ne , $N_{20,\text{total}}$ and $N_{21,\text{total}}$ are the total measured amounts of ^{20}Ne and ^{21}Ne , and $R_{21/20,\text{a}}$ is the atmospheric $^{21}\text{Ne}/^{20}\text{Ne}$ ratio (0.002959). As we have found that small quantities of Ne released from heating empty Ta packets have atmospheric composition, correction for any ^{21}Ne blank derived from sample heating is included in the calculation of excess ^{21}Ne and not computed separately. Uncertainties in excess ^{21}Ne concentrations are derived from counting uncertainties on mass 21, uncertainty on the ^{20}Ne signal derived from correction of the mass 20 signal for the $^{40}\text{Ar}^{++}$ contribution, and an uncertainty in machine sensitivity derived from replicability of measurements on atmospheric Ne standards, added in quadrature. Table S1 reports summary excess ^{21}Ne concentrations, and Table S2 shows the full step-degassing results for all Ne isotopes. For this work we have not attempted to separate observed excess ^{21}Ne into separate cosmogenic and nucleogenic inventories, and instead simply refer to ‘inherited ^{21}Ne ,’ which could be either cosmogenic or nucleogenic.

3. ^{10}Be measurements.

Quartz remaining after ^3He and ^{21}Ne measurements was further cleaned by additional HF-etching in a sonic bath, and digested in concentrated HF with 0.2 mg of ^9Be added from carrier obtained from the University of Washington (UW-TRCH-B-1-5; $^{10}\text{Be}/^9\text{Be} = 0.6 \pm 0.1 \times 10^{-15}$). After removal of Si by evaporation of SiF_6 , beryllium was extracted via column chromatography and pH-controlled hydroxide precipitations following standard protocols (Stone et al., 2004; Ditchburn and Whitehead, 1994). Hydroxides were then converted to BeO by heating, mixed with niobium powder, and loaded into targets for $^{10}\text{Be}/^9\text{Be}$ measurement by accelerator mass spectrometry at the Center for Accelerator Mass Spectrometry, Lawrence Livermore National Laboratory. Ratio measurements were normalized to the 07KNSTD3110 standard having $^{10}\text{Be}/^9\text{Be} = 2850 \times 10^{-15}$ (Nishiizumi et al., 2007). A full process blank measured with the samples in this study contained 10900 ± 1900 atoms ^{10}Be ; for all samples, this blank represented <2% of total atoms counted. Reported uncertainties in ^{10}Be concentration include AMS measurement uncertainty derived from counting statistics and target reproducibility, uncertainty in the Be concentration of the carrier, and uncertainties in the blank correction, added in quadrature. ^{10}Be concentrations are shown in Table S1.

4. Random walk model for ^{10}Be and ^{21}Ne production and loss in a mixed soil profile

The forward model used to generate Figure 3 in the main text considers a large number of individual quartz grains that move by a random walk within the soil mixed layer. We considered 5000 individual grains and assumed that the mixed layer extends from the soil surface to 170 cm depth and there is no loss of soil particles at the surface (which is equivalent to specifying zero erosion) or through the bottom boundary of the mixed layer.

Particle depths at the beginning of the simulation are drawn from a uniform random distribution between 0 and 170 cm. Given particles $i = 1 \dots n$ and timesteps $j = 0 \dots m$ with timestep duration δt , $Z_{i,j}$ is the depth of particle i below the surface in timestep j , and $Z_{i,j} = Z_{i,j-1} + k$, where k is drawn from a random normal distribution with mean 0 and standard deviation σk (cm). The impermeable boundary conditions at the top and bottom of the mixed layer are implemented as follows: if $Z_{i,j-1} + k < 0$, then $Z_{i,j} = |Z_{i,j-1} + k|$ and if $Z_{i,j-1} + k > 170$, then $Z_{i,j} = 170 - (Z_{i,j-1} + k - 170)$.

For simplicity, we assume that cosmogenic-nuclide production is exponential such that the ^{10}Be production rate is $P_{10}(Z) = P_{10}(0)\exp(-Z\rho/\Lambda)$, where $P_{10}(Z)$ is the production rate (atoms/g/yr) at depth Z (cm), $P_{10}(0)$ is the surface production rate (atoms/g/yr, estimated using the scaling model of Stone, 2000, as implemented by Balco et al., 2008), ρ is the average soil density in the mixed layer (1.7 g/cm^3 , derived from field measurements), and Λ is an effective attenuation length for spallogenic production (150 g/cm^2). The ^{10}Be concentration in particle i at the end of the model run is:

$$\sum_j P_{10}(Z_{ij})\exp(-\lambda_{10}\delta t(m-j))$$

where λ_{10} is the ^{10}Be decay constant ($4.99 \times 10^{-7} \text{ yr}^{-1}$). We then compute the average ^{10}Be concentration of all particles that lie within each 10-cm depth increment at the end of the model run, and these average concentrations are shown by the black line in Figure 3 in the main text. Given the simplifying assumptions of zero erosion, a simple exponential production rate model, and an impermeable mixed layer boundary, ^{10}Be concentrations below the mixed layer are simply $(P_{10}(Z)/\Lambda)(1-\exp(-\lambda_{10}m\delta t))$. Simplifying the model by disregarding muon production, as we have done, could result in an inaccuracy if we were to try to obtain an exposure age by fitting model to data, but does not have any effect on the basic mixing/heating dynamics that we aim to explain with a simple model. .

The adjustable parameters in the mixing model are the exposure duration ($m\delta t$) and the distribution of the random walk distance per time step ($\sigma k\delta t$). Given a ^{10}Be depth profile with an evident mixed layer, one can adjust the exposure duration to match the mean ^{10}Be concentration in the mixed layer and then adjust $\sigma k\delta t$ to match the variation in ^{10}Be concentration with depth. A small value of $\sigma k\delta t$ will predict a poorly mixed profile that resembles the depth dependence of the production rate, whereas large $\sigma k\delta t$ predicts a nearly constant concentration through the mixed layer. For these data, an exposure duration of 425,000 years, $\sigma k = 10 \text{ cm}$, and $\delta t = 250 \text{ yr}$ closely matches the measured ^{10}Be concentrations in the mixed layer (Figure 3 in main text).

Production of ^{21}Ne in each particle in each timestep is computed in the same way as for ^{10}Be , except that $P_{21}(Z) = 4.03 P_{10}(Z)$ (Balco et al., 2019). The ^{21}Ne concentration in particle i at the end of the model run is:

$$N_{21,inh} + \sum_j P_{21}(Z_{ij})L_{ij}$$

where $N_{21,inh}$ is an inherited ^{21}Ne concentration (atoms/g) derived either from prior exposure or decay of naturally occurring U and Th in quartz, and L_{ij} is the fractional loss due to wildfire heating of ^{21}Ne produced in particle i in timestep j .

To compute L_{ij} , we assume that surface and subsurface wildfire heating is characterized by a surface temperature T_0 and exponential attenuation constant Λ_T , such that $T(Z) = T_0\exp(-Z/\Lambda_T)$. This is an approximation to the results of soil heating models such as that in Richter et al. (2022). ^{21}Ne diffusivity in quartz is then assumed to follow an Arrhenius law such that the diffusion constant $D = D_0\exp(-E_a/RT)$, where R is the universal gas constant (8.314 J/K/mol), T is temperature ($^\circ\text{K}$), and the constants D_0 (cm^2/s) and E_a (J/mol) for ^{21}Ne in quartz are those measured by Shuster and Farley (2005). For simplicity, we also assume that quartz grains in the soil have the same diameter as the grain analyzed by Shuster and Farley (0.43 mm). We further assume that a fire of duration t_f (s) takes place in each 250-yr time step. Loss of ^{21}Ne produced in each time step by heating L_{ij} is then proportional to the sum of the quantity Dt_f for all subsequent time steps, which is:

$$\sum_{k=j}^m D_{i,k} t_f$$

L_{ij} is computed using Equation (4) of Fechtig and Kalbitzer (1966). As for ^{10}Be , the resulting ^{21}Ne concentrations at the end of the model run are averaged for all particles in each 10-cm depth increment.

Given assumed values for D_0 and E_a for ^{21}Ne in quartz and a fire frequency defined by the time step length δt , the adjustable parameters for the diffusive loss part of the model are the inherited ^{21}Ne concentration $N_{21,inh}$, the surface temperature during a fire T_0 , the heating attenuation constant Λ_T , and the fire duration t_f . As noted in the main text, some of these parameters are not independent: the controlling parameter for ^{21}Ne loss is the product Dt_f , which can be varied by changing either the fire temperature profile or the fire duration. For the example model runs shown in Figure 3 in the main text we used a time step of 250 yr (which approximates the long-term fire frequency in Long et al., 2018) and heating parameters $T_0 = 450^\circ \text{C}$, $\Lambda_T = 6 \text{ cm}$, and $t_f = 60 \text{ minutes}$ (which approximate a typical wildfire heating scenario in Richter et al., 2022 or similar work). $N_{21,inh}$ is lithology-specific and has not been estimated for the source bedrock at our study site, so is unconstrained. We assumed $N_{21,inh} = 2 \text{ Matoms/g}$, which is a reasonable value for quartz with typical U/Th concentrations and a Cenozoic cooling age, that approximately fits the data. In future work, this parameter could be measured in shielded samples of the source bedrock.

A MATLAB implementation of the model is provided as a supplemental file.

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