mmol HF (1 mmol HF for 78421), in an

Beryllium-10 from the Sun

K. Nishiizumi^{1*} and M. W. Caffee²[†]

Beryllium-10 (¹⁰Be) in excess of that expected from in situ cosmic ray spallation reactions is present in lunar surface soil 78481; its presence was revealed with a sequential leaching technique. This excess ¹⁰Be, representing only 0.7 to 1.1% of the total ¹⁰Be inventory, is associated with surface layers (<1 micrometer) of the mineral grains composing 78481. This excess ¹⁰Be and its association with surficial layers corresponds to $(1.9 \pm 0.8) \times 10^8$ atoms per square centimeter, requiring a ¹⁰Be implantation rate of $(2.9 \pm 1.2) \times 10^{-6}$ atoms per square centimeter per second on the surface of the Moon. The most likely site for the production of this excess ¹⁰Be is the Sun's atmosphere. The ¹⁰Be is entrained into the solar wind and transported to the lunar surface.

Lunar soils record exposure to the energetic particle environment of the inner solar system. This exposure includes the constant bombardment of the lunar surface by lowenergy solar wind (SW) ions and high-energy galactic cosmic rays (GCR). Near the surface of the Sun, energetic protons are accelerated and trapped in closed magnetic loops (1). These trapped energetic protons bombarding the ambient solar atmosphere produce ¹⁰Be from target nuclei such as C, N, and O. A fraction of this ¹⁰Be escapes from the Sun and is probably entrained into the SW or into solar energetic particles (SEP). A small fraction of the ¹⁰Be that escapes from the Sun can be intercepted by the Moon and implanted in lunar surface materials.

Because the range of GCR and solar cosmic ray (SCR) protons is larger than the average grain size of 100 to 200 μ m, the distribution of both GCR- and SCR-produced ¹⁰Be is correlated with grain volume. Implanted species, on the other hand, may be associated with grain surface area. Accordingly, we performed step-wise acid leaching experiments designed to delineate volumeand surface-correlated components.

Two Apollo 17 trench soils, 78481,16 (0to 1-cm depth) and 78421,16 (bottom layer, 10 cm thick, of a 25-cm-deep trench), were selected for this experiment. The soil samples were collected from Station 8, the base of the Sculptured Hills (2). The soil samples were separated into 30- to 65- μ m, 65- to 130- μ m, and >130- μ m-diameter fractions. Each sample (150 to 200 mg) was subsequently leached by 10 ml of different concentrations of acid solution, 0.02 mmol HNO₃, 0.5 mmol HNO₃ + 0.002 mmol HF, 1 mmol HNO₃ + 0.20 mmol HF, and 1 mmol HNO₃ + 0.2 ultrasonic bath for 30 min each. Each leachant contained 0.4 mg of Be carrier. After each leaching step, the supernatant and the undissolved sample were isolated by centrifugation. The supernatant was filtrated by a 0.45-µm Millipore filter to remove micrometer-sized grains in the solution. After taking an aliquot for chemical analysis, Al and Cl carriers were added to the supernatant. In aggregate, the sequential leaching steps dissolved 19 to 34% of the original mass. After leaching, we dissolved a fraction of the undissolved residue. Between 70 and 100 mg was totally dissolved by an HF + HNO_3 mixture into which Be and Cl carriers had been added. Be, Al, and Cl were chemically separated and purified from each fraction with a combination of anion exchange ion chromatography, acetylacetone solvent extraction, and cation exchange ionchromatography techniques. The concentrations of ¹⁰Be, ²⁶Al, and ³⁶Cl were measured with accelerator mass spectrometry (AMS) (3). The concentration of each nuclide was normalized to ICN-10Be, NBS-26Al, and NBS-³⁶Cl standards prepared by one of the authors (K.N.). Concentrations of Mg, Al, Si, Ca, and Fe in aliquots of the leachant and the total dissolution were measured by atomic absorption spectroscopy (AA). The uncertainty of the AA measurements is ± 2 to 3%, except for the Al measurements in leaching steps, which have uncertainties of ± 10 to 20%. The compositions of the leaching steps were normalized to the amount of starting material; these are shown in Table 1. The ¹⁰Be, ²⁶Al, and ³⁶Cl concentrations in each fraction varied by three to four orders of magnitude because different amounts of surface layers were dissolved. The amounts of dissolved materials are calculated from the concentrations of Mg, Al, Si, Ca, and Fe combined with the stochiometric concentration of O.

All cosmogenic nuclides in the deeper sample (78421) must be produced in situ by GCR. The factor of five higher concentrations of 26 Al and slightly higher concentrations of 36 Cl in the surface sample (78481) indicate in situ SCR production of these nuclides in addition to GCR production (Table 1). Our ²⁶Al concentration in total dissolution of 78481 (243 to 251 dpm/kg) agrees with the nondestructive gamma-ray measurements of 257 \pm 12 dpm/kg (4). Oxygen is the major target element for the production of ¹⁰Be, Al and Si for ²⁶Al, and Ca and Fe for ³⁶Cl. The ²⁶Al concentrations of all leaching fractions are slightly higher than that of the total dissolution because of preferential dissolution of Al during leaching. Although the ³⁶Cl concentrations of the leaching fractions are slightly higher than that of total dissolution, the ³⁶Cl activities normalized to target elements, in dpm/ kg (8Ca + Fe), are identical to that of total dissolution. GCR- and SCR-produced radionuclides are the background; any additional components may be plausibly ascribed to SW implantation. However, the SW components should occur only in the outermost layer (≤ 0.2 μm), owing to the low energy of implantation. SW-implanted ¹⁰Be should be preferentially released in the first leaching steps.

If each lunar grain was homogeneously etched, the average etched depths in each leaching step, based on the volume of dissolved materials, are 0.3, 0.7, and 1.7 µm for the 30- to 65-µm fraction; 0.9, 2.4, and 5.4 µm for the 65to 130-µm fraction; and 1.5, 3.7, and 10 µm for the >130-µm fraction. Because the total dissolution sample was obtained after sequential leaching steps, it should contain only in situproduced cosmogenic nuclides. The ¹⁰Be activity of each leaching fraction in 78481 was compared with that of the total dissolution sample (Table 2). For the leaching step with the most dilute HF (0.002 mmol), which removed the outermost layer of the grains, 14 to 16% more ¹⁰Be was observed relative to the ¹⁰Be activity from the total dissolution. The 0.02 mmol HF step for 30- to 65-µm fraction also indicates excess ¹⁰Be. However, the release pattern observed for ¹⁰Be does not hold for ²⁶Al and ³⁶Cl, indicating that the ¹⁰Be is produced by a different mechanism than that for the other nuclides. There are ¹⁰Be excesses in most of the leaching fractions of soil 78481; however, for the 0.02 and 0.2 mmol HF steps, except for the 30- to 65-µm fraction, the measured excesses are within the experimental uncertainties. The stated uncertainties include the AMS errors and an uncertainty in the amount of dissolved materials. The calculated amount of excess ¹⁰Be is $(1.37 \pm 0.31) \times 10^{-4}$ dpm/g for the 30- to 65-µm fraction, $(1.19 \pm 0.28) \times 10^{-4}$ dpm/g for the 65- to 130- μ m fraction, and (0.91 ± 0.26) $\times 10^{-4}$ dpm/g for the >130- μ m fraction. The amount of excess ¹⁰Be decreases with increasing grain sizes, as expected. The size distribution of the bulk 78481 soil is 2.8, 61.0, 16.7, and 19.5% in the <30-, 30- to 65-, 65- to 130-, and >130-µm fractions, respectively. Combining the size distribution, assuming 20 μ m as the average size of the grains <30 μ m,

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 Table 1. Dissolved cation composition and cosmogenic nuclide concentrations of each leaching step and those of the total dissolution in lunar soil 78481 and 78421. The compositions of leaching steps were normalized to the amount of starting material. Concentration of Si in total

dissolution was adopted as 20.4% because we did not measure Si in the fraction. ¹⁰Be, ²⁶Al, and ³⁶Cl concentrations in each leached fraction and total dissolution were normalized to the dissolved amount of lunar soil 78481 and 78421.

Size (µm)		Mg (%)	Al (%)	Si (%)	Ca (%)	Fe (%)	¹⁰ Be (dpm/kg)	²⁶ Al (dpm/kg)	³⁶ Cl (dpm/kg)	³⁶ Cl [dpm/kg (8Ca + Fe)]
					78481	,16				
30-65	0.5 mmol HNO ₃ + 0.002 mmol HF	0.20	0.4	0.53	0.33	0.40	13.8 ± 0.5	289 ± 17	19.2 ± 0.7	19.7 ± 0.5
65–130	0.5 mmol HNO ₃ + 0.002 mmol HF	0.34	0.6	1.0	0.59	0.60	14.2 ± 0.5	290 ± 19	18.3 ± 0.6	18.5 ± 0.5
>130	$0.5 \text{ mmol HNO}_{3} + 0.002 \text{ mmol HF}$	0.28	0.5	0.89	0.49	0.52	13.5 ± 0.5	293 ± 21	17.3 ± 0.6	18.4 ± 0.5
30-65	1 mmol HNO3 + 0.02 mmol HF	0.14	0.4	0.46	0.31	0.25	13.0 ± 0.6	322 ± 23	17.9 ± 0.8	18.3 ± 0.5
65–130	1 mmol HNO3 + 0.02 mmol HF	0.29	0.8	1.2	0.57	0.50	12.2 ± 0.5	302 ± 28	17.3 ± 0.7	19.5 ± 0.5
>130	1 mmol HNO3 + 0.02 mmol HF	0.37	0.9	1.5	0.71	0.66	12.1 ± 0.4	293 ± 26	16.5 ± 0.6	18.9 ± 0.5
30-65	1 mmol HNO ₃ + 0.2 mmol HF	0.26	0.7	1.2	0.64	0.45	12.6 ± 0.4	295 ± 24	17.8 ± 0.7	18.6 ± 0.5
65–130	1 mmol HNO ₃ + 0.2 mmol HF	0.38	1.0	1.7	0.80	0.66	12.0 ± 0.4	307 ± 28	17.3 ± 0.7	20.1 ± 0.6
>130	1 mmol HNO ₃ + 0.2 mmol HF	0.54	1.4	2.5	1.1	0.97	12.0 ± 0.3	303 ± 23	16.3 ± 0.5	19.2 ± 0.5
30-65	Total dissolution	6.5	8.7	=20.4	8.1	9.6	12.0 ± 0.2	243 ± 13	14.9 ± 0.2	20.1 ± 0.5
65–130	Total dissolution	6.7	8.3	=20.4	8.2	9.7	12.3 ± 0.2	251 ± 17	14.9 ± 0.2	19.9 ± 0.5
>130	Total dissolution	6.8	8.6	=20.4	8.3	9.4	11.8 ± 0.3	251 ± 13	14.3 ± 0.3	18.9 ± 0.5
					78421	,16				
30-65	0.5 mmol HNO ₃ + 0.002 mmol HF	0.22	0.5	0.70	0.44	0.43	10.7 ± 0.8	57 ± 5	14.7 ± 0.6	15.1 ± 0.4
65–130	0.5 mmol HNO ₃ + 0.002 mmol HF	0.33	0.7	1.1	0.56	0.61	12.0 ± 0.4	60 ± 4	15.9 ± 0.7	17.8 ± 0.6
>130	$0.5 \text{ mmol HNO}_{3} + 0.002 \text{ mmol HF}$	0.25	0.5	0.85	0.47	0.47	11.2 ± 0.4	61±5	n.d.	n.d.
30-65	1 mmol HNO3 + 0.02 mmol HF	0.21	0.5	0.84	0.46	0.40	11.2 ± 0.4	51 ± 4	14.2 ± 0.6	15.2 ± 0.4
65–130	1 mmol HNO3 + 0.02 mmol HF	0.43	1.0	1.6	0.80	0.77	11.7 ± 0.5	59 ± 5	13.9 ± 0.8	16.1 ± 0.7
>130	1 mmol HNO3 + 0.02 mmol HF	0.30	0.7	1.2	0.59	0.53	12.3 ± 0.5	60 ± 5	14.8 ± 0.7	16.6 ± 0.7
30-65	1 mmol HNO3 + 1 mmol HF	0.43	1.4	2.3	1.1	0.79	11.4 ± 0.4	58 ± 6	14.6 ± 0.6	16.8 ± 0.5
65–130	1 mmol HNO3 + 1 mmol HF	0.66	1.9	3.4	1.5	1.2	11.5 ± 0.4	60 ± 4	13.6 ± 0.7	15.6 ± 0.7
>130	1 mmol HNO ₃ + 1 mmol HF	0.74	2.2	3.7	1.8	1.4	11.7 ± 0.4	60 ± 5	14.6 ± 0.6	16.8 ± 0.6
30-65	Total dissolution	6.4	8.2	=20.4	7.8	9.7	11.9 ± 0.2	49 ± 3	11.7 ± 0.1	16.2 ± 0.3
65–130	Total dissolution	5.9	6.8	=20.4	7.2	9.7	12.5 ± 0.3	45 ± 2	12.0 ± 0.1	17.9 ± 0.4
>130	Total dissolution	6.6	9.0	=20.4	8.7	9.4	12.3 ± 0.2	53 ± 3	13.1 ± 0.2	16.6 ± 0.4

with the excess ¹⁰Be contents for each grain size, yields an excess ¹⁰Be of the bulk soil 78481 of $(1.27 \pm 0.30) \times 10^{-4}$ dpm/g or $(1.44 \pm 0.34) \times 10^{8}$ atom/g. Although the excess ¹⁰Be represents only 1.0% of the total amount of ¹⁰Be in the soil, the result suggests that the excess ¹⁰Be is released from the outermost 1-µm layer of 78481. The upper few centimeters of lunar soil was gardened by micrometeorites for a million years (5).

The ²⁶Al concentration in 78481 indicates exposure to SCR for a few million years. If we assume an exposure time comparable to the ¹⁰Be mean-life, as seems reasonable given the ²⁶Al results, we obtain a ¹⁰Be concentration of $(1.9 \pm 0.8) \times 10^8$ atoms/cm², assuming an average soil bulk density of 1.8 g/cm³, a density based on four Apollo 17 surface cores. The corresponding ¹⁰Be implantation rate is $(2.9 \pm 1.2) \times 10^{-6}$ atoms/ cm²·s on the surface of the Moon. The result includes the uncertainty $(\pm 33\%)$ of depth of the skim soil 78481. Assuming that the ¹⁰Be flux varies falls with an inverse square law, the ¹⁰Be escape rate from the Sun is 0.13 \pm 0.05 atoms/s·cm². The SW proton flux at 1 AU is 2.5 \times 10⁸/cm²·s, yielding a ¹⁰Be/H ratio of $(1.1 \pm 0.5) \times 10^{-14}$. Preliminary ¹⁰Be and ¹⁴C measurements in lunar soils have also been reported by Jull et al. (6). Excess ¹⁴C in lunar soils and rock has been detected by several investigators whose rates determined ¹⁴C implantation: $\sim 1 \times 10^{-3}$

Table 2. Excess ¹⁰Be in each leaching fraction of lunar soil 78481. The excess ¹⁰Be of each leaching was calculated by comparison of that total dissolution sample.

Size (µm)		Excess ¹⁰ Be	Excess ¹⁰ Be (10 ⁻⁶ dpm)	Excess ¹⁰ Be (10 ⁻⁵ dpm/g)
30-65	0.5 mmol HNO ₂ + 0.002 mmol HF	1.16 ± 0.04	13.3 ± 3.2	6.8 ± 1.6
65–130	$0.5 \text{ mmol HNO}_{3} + 0.002 \text{ mmol HF}$	1.16 ± 0.04	20.1 ± 4.7	11.9 ± 2.8
>130	$0.5 \text{ mmol HNO}_{3} + 0.002 \text{ mmol HF}$	1.14 ± 0.04	17.9 ± 5.2	9.1 ± 2.6
30-65	1 mmol HNO, + 0.02 mmol HF	1.08 ± 0.04	5.8 ± 2.7	2.9 ± 1.4
65–130	1 mmol HNO3 + 0.02 mmol HF	1.00 ± 0.04	-0.3 ± 4.4	-0.2 ± 2.6
>130	1 mmol HNO3 + 0.02 mmol HF	1.03 ± 0.04	5.0 ± 6.1	2.5 ± 3.1
30-65	1 mmol HNO3 + 0.2 mmol HF	1.05 ± 0.03	7.9 ± 4.4	4.0 ± 2.3
65–130	1 mmol HNO3 + 0.2 mmol HF	0.98 ± 0.03	-3.8 ± 5.1	-2.3 ± 3.0
>130	1 mmol HNO3 + 0.2 mmol HF	1.02 ± 0.04	5.9 ± 10.3	3.0 ± 5.2
30-65	Total dissolution	1.00		
65–130	Total dissolution	1.00		
>130	Total dissolution	1.00		

¹⁴C atoms/cm²·s (7), 2.3×10^{-3} ¹⁴C atoms/ cm²·s (8), and 1.5 to 4.5×10^{-6} ¹⁴C atoms/ cm²·s (6, 9). The disparity between the Jull *et al.* (6, 9) implantation rates and those of the earlier works is the result of an incorrect "effective" surface area determination. If we use the convention for surface area used by the earlier works and that of this work, the Jull *et al.* (6, 9) ¹⁴C measurements yield a ¹⁴C flux of ~(0.2 to 2) $\times 10^{-3}$ ¹⁴C atoms/cm²·s, consistent with previous determinations.

Further evidence that the excess 10 Be is SW implanted comes from SRIM (the stopping and range of ions in matter) and TRIM (the transport of ions in matter) calculations (10). For C ions having velocities of 300 to

800 km/s, representative of the average SW ion velocity, the maximum implantation depth in Si is more than 0.1 µm, but the peak occurs at 0.03 µm. On the basis of the amount of material dissolved in the sequential leaching experiment, we estimate that the range of implanted ¹⁰Be is no more than 0.3 to 1 μ m (0.1 to 0.3 mg/cm²), which corresponds to less than 10 to 50 keV per nucleon, consistent with SW energies rather than the higher energies, i.e., MeV, typical of SEP. These calculations carry the qualification that diffusion of ¹⁰Be in silicates is not well characterized, but analogy with better characterized elements, e.g., C, indicates that the diffusion rate should be slow.

Seismic Observations of Splitting of the Mid–Transition Zone Discontinuity in Earth's Mantle

Arwen Deuss* and John Woodhouse

The transition zone of Earth's mantle is delineated by globally observed discontinuities in seismic properties at depths of about 410 and 660 kilometers. Here, we investigate the detailed structure between 410 and 660 kilometers depth, by making use of regional stacks of precursors to the SS phase. The previously observed discontinuity at about 520 kilometers depth is confirmed in many regions, but is found to be absent in others. There are a number of regions in which we find two discontinuities at about 500 and 560 kilometers depth, an effect which can be interpreted as a "splitting" of the 520 kilometer discontinuity. These observations provide seismic constraints on the sharpness and observability of mineralogical phase transitions in the mantle transition zone.

The characteristics of the mantle transition zone (between 410 and 660 km) are important for our understanding of Earth's dynamics. Constraints on seismic velocity discontinuities in the transition zone can be obtained from reflection observations of seismic waves and high-pressure experiments on mantle rocks. The 410-km discontinuity is generally thought to be caused by the phase transformation of low-pressure olivine (aphase) to wadsleyite (\beta-phase) and the 660km discontinuity by the phase transformation of ringwoodite (y-phase) to perovskite and magnesiowüstite (1, 2). Over the past decades, a number of seismic studies have demonstrated the global existence of these discontinuities and have mapped their topography (3-6).

A number of studies have reported a weaker discontinuity, in the mid-transition zone, at about 520-km depth. The 520-km discontinuity is generally believed to be caused by the transition in the mantle olivine component from wadsleyite (B-phase) to ringwoodite (γ -phase) (1, 7); the Clapevron slope of this transition is 4 MPa K^{-1} . The most consistent seismic evidence for the 520km discontinuity comes from long-period S wave observations using SS precursors (5, 6, (8, 9) or ScS reverberations (10). Some studies claim that the 520-km discontinuity is a global feature (8, 9), whereas others find that it can be observed only in certain regions (5) or that it is not found beneath continental shields (6). It is much more difficult to observe reflections from this discontinuity in shortperiod data (11), and refracted wave studies

*To whom correspondence should be addressed. Email: arwen.deuss@earth.ox.ac.uk (12, 13) do not find evidence for a discontinuity at 520 km, suggesting that it is not a sharp reflector, but a velocity gradient with a wide transition interval (14).

We investigate the characteristics of the 520-km discontinuity in detail by using precursors to the SS phase, which are underside reflections at a discontinuity below the SS bounce point. Long-period SS waves are used to investigate properties of the 520-km discontinuity, because they are sensitive to weak discontinuities. The precursors have small amplitudes and cannot be observed in individual seismograms. Thus, we stack the traces in the slownesstime domain to suppress incoherent noise and to enhance the visibility of the precursors. Such stacks amplify and clarify the peaks in amplitude associated with the major discontinuities at 410 and 660 km and provide a means of investigating hypotheses about the existence and regional variation of the weaker 520-km discontinuity.

We use a global data set of 7018 seismograms collected from the Incorporated Research Institutions for Seismology/International Deployment of Accelerometers (IRIS/ IDA) global network (15); this is the type of data set that has been used in other studies to obtain global maps of topography on transition zone discontinuities. A stack of all seismograms in our data set shows reflections from "410" and "660" and a weaker reflection from a depth of about 520 km (Fig. 1).

We divide Earth's surface into 407 spherical caps with 10° radius (16) and stack all seismograms with bounce points in each cap. North America and the north Pacific provide an excellent region to observe small-scale regional variations in the 520-km discontinuity, because it is well covered by SS wave bounce points. The North American cross

Gamma-ray observations indicate that nuclear interactions between energetic protons and ambient gas occur at various depths in the solar atmosphere, especially those associated with the lower chromosphere or the photosphere (11). The gamma-ray observations also indicate that these reactions occur prodigiously; in number, the interacting protons are generally more numerous than those escaping from the Sun (12). Preliminary theoretical estimates suggest that the time-averaged solar flare production at the surface of the Sun is on the order of 0.1^{-10} Be/cm²·s (13). The SW escape rate from the Sun is $\sim 10^{13}$ H/cm²·s, corresponding to a SW removal rate of 3 \times 10²⁶ H/cm² per million years. This corresponds to a column depth of gas extending down about 2000 km into the photosphere. If the ¹⁰Be produced in the top 10²⁴ H/cm² of the Sun's atmosphere is not mixed any deeper than 2000 km into the photosphere in a million years, we would expect the SW to carry away the entire ¹⁰Be production at the rate of 0.1 atom/cm²·s. If the ¹⁰Be is mixed within that time through the solar convective zone, probably 1 to 2×10^5 km below the photosphere, then the mean ¹⁰Be concentration would be diluted by four to five orders of magnitude. Our result suggests that most of the ¹⁰Be is ejected in the SW and large flares without mixing and loss into the deeper solar convective zone.

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