DRAFT VERSION NOVEMBER 3, 2023 Typeset using LATEX manuscript style in AASTeX631

1

	Solar-Energetic-Particle Track-Production Rates at 1 au:
	Comparing In-Situ Particle Fluxes with Lunar Sample-Derived Track Densities
2	A. R. Poppe, $^{1}$ P. S. Szabo, $^{1}$ E. R. Imata, $^{2}$ L. P. Keller, $^{3}$ and R. Christoffersen <sup>4</sup>
3	<sup>1</sup> Space Sciences Laboratory, University of California at Berkeley, Berkeley, CA, 94720
4	<sup>2</sup> Dept. of Astronomy, University of California at Berkeley, Berkeley, CA, 94720
5	<sup>3</sup> NASA Johnson Space Center, Mail Code XI3, Houston, Texas 77058, USA
6	<sup>4</sup> Jacobs, NASA Johnson Space Center, Mail Code X13, Houston, Texas 77058, USA
7	Submitted to Astrophys. J. Lett.
8	ABSTRACT
9	Heavy $(Z > 26)$ solar energetic particles (SEPs) with energies ~1 MeV/nucleon are
10	known to leave visible damage tracks in meteoritic materials. The density of such 'solar
11	flare tracks' in lunar and asteroidal samples has been used as a measure of a sample's ex-
12	posure time to space, yielding critical information on planetary space weathering rates,
13	the dynamics and lifetimes of interplanetary dust grains, and the long-term history of
14	solar particle fluxes. Knowledge of the SEP track accumulation rate in planetary ma-
15	terials at 1 au is critical for properly interpreting observed track densities. Here, we
16	use in-situ particle observations of the $0.50{-}3.0~{\rm MeV/nuc}$ Fe-group SEP flux taken by
17	NASA's Advanced Composition Explorer (ACE) to calculate a flux of track-inducing
18	particles at 1 au of $6.0 \times 10^5$ cm <sup>-2</sup> yr <sup>-1</sup> str <sup>-1</sup> . Using the observed energy spectrum
19	of Fe-group SEPs, we find that the depth distribution of SEP-induced damage tracks

Corresponding author: A. R. Poppe poppe@berkeley.edu

inferred from ACE measurements matches closely to that recently measured in lunar 20 sample 64455; however, the magnitude of the ACE-inferred rate is approximately  $25 \times$ 21 higher than that observed in the lunar sample. We discuss several hypotheses for the 22 nature of this discrepancy, including inefficiencies in track formation, thermal annealing 23 of lunar samples, erosion via space weathering processing, and variations in the SEP flux 24 at the Moon, yet find no satisfactory explanation. We encourage further research on 25 both the nature of SEP track formation in meteoritic materials and the flux of Fe-group 26 SEPs at the lunar surface in recent and geologic times to resolve this discrepancy. 27

# 1. INTRODUCTION

Objects exposed to the harshness of space are subjected to a wide range of charged-particle irra-29 diation that can physically and chemically alter their nature. In particular, fluxes of  $\sim 1 \text{ MeV/nuc}$ , 30 high-Z (Z > 26, typically Fe and heavier) solar energetic particles (SEPs) have been shown to leave 31 observable damage tracks in meteoritic minerals, including interplanetary dust grains (e.g., Bradley 32 et al. 1984; Thiel et al. 1991), meteorites (e.g., Goswami 1981), and returned lunar and asteroidal 33 samples (e.g., Crozaz et al. 1972; Blanford et al. 1974; Keller & Berger 2014). The characterization 34 of these tracks, including their overall density as well as their depth profiles, informs us about both 35 the exposure age of planetary materials to space (e.g., Bradley et al. 1984; Sandford 1986; Keller 36 et al. 2021; Keller & Flynn 2022) and the solar energetic particle flux over solar system timescales 37 (e.g., Price & O'Sullivan 1970; Zinner 1980). 38

<sup>39</sup> A key question in such studies is the rate at which typical meteoritic minerals accumulate SEP tracks <sup>40</sup> at 1 au. Blanford et al. (1974) used acid-etching techniques on Apollo 16 sample 64455 to determine <sup>41</sup> an SEP track accumulation rate of  $\sim 6 \times 10^5$  tracks cm<sup>-2</sup> yr<sup>-1</sup> for an assumed  $2\pi$  exposure; however, <sup>42</sup> this analysis required a series of renormalizations and extrapolations, which leaves uncertainty as <sup>43</sup> to the robustness of the final results. Recently, laboratory measurements of SEP-induced tracks <sup>44</sup> within lunar sample 64455 using more advanced techniques have yielded a re-calibration of the rate <sup>45</sup> of SEP track formation in minerals at 1 au of  $4.4 \pm 0.4 \times 10^4$  tracks cm<sup>-2</sup> yr<sup>-1</sup>, again assuming a  $2\pi$ 

28

exposure (Keller et al. 2021). In turn, the SEP track-formation rate determined in Keller et al. (2021) has led to the conclusion that interplanetary dust grains collected from the terrestrial stratosphere with unusually high track densities ( $\geq 10^{11}$  tracks cm<sup>-2</sup>) may originate from the Edgeworth-Kuiper Belt beyond Neptune (Keller & Flynn 2022). Such a conclusion has significant implications for the distribution and dynamics of interplanetary dust grains throughout the solar system (e.g., Liou & Zook 1999; Kuchner & Stark 2010; Poppe et al. 2019), yet such conclusions rely critically on knowledge of the SEP track accumulation rate.

Here, we use a complementary approach to calculating the track-inducing flux of SEPs at 1 au via 53 in-situ observations from NASA's Advanced Composition Explorer (ACE) (Stone et al. 1998), which 54 has been in a heliocentric orbit at the solar-terrestrial Lagrange-1 point since 1998. We compare this 55 in-situ derived rate to the sample-derived track-formation rate of Keller et al. (2021) and find that 56 while the shape of the track density versus depth profile matches the sample data well, the overall 57 magnitude of the in-situ derived rate is approximately  $25 \times$  higher than the lunar sample-derived 58 rate. We assess several possibilities for the discrepancy between these two measurement approaches, 59 yet find no obvious explanation and therefore urge additional laboratory and in-situ experiments on 60 the nature of SEP track accumulation in meteoritic and lunar minerals. 61

- 62
- 63

## 2. FE-GROUP SOLAR ENERGETIC PARTICLE FLUX AT 1 AU

To calculate the flux of SEP track-producing particles at 1 au, we use observations taken by the 64 Ultra-Low-Energy Isotope Spectrometer (ULEIS) instrument onboard NASA's Advanced Composi-65 tion Explorer. Launched in 1997, the ACE mission was designed to measure the elemental and isotopic 66 composition of space-based particles over a wide range of energies ( $\sim keV/nuc$  to  $\sim GeV/nuc$ ) and 67 masses (atomic numbers,  $1 \le Z \le 28$ ) (Stone et al. 1998). Amongst a broader payload, the ULEIS 68 instrument measures the compositionally resolved energy spectra of elements between He (Z=2) and 69 Ni (Z=28) in the energy range,  $\sim 45 \text{ keV/nuc} < E < \sim \text{few MeV/nuc}$  (Mason et al. 1998). Solar-flare 70 track production within meteoritic materials only occurs for very heavy nuclei with  $Z \ge 26$  (e.g., Ch. 71 1, Fleischer et al. 1975, and refs. therein); thus, we focus our analysis on the Fe-group ( $Z \ge 26$ ) ions 72

#### POPPE ET AL.

4

<sup>73</sup> measured by ULEIS. We acquired the full dataset of Fe-group flux measured by ULEIS between 1998 <sup>74</sup> and mid-2023 in the energy range, 0.035 < E < 3.07 MeV/nuc, via NASA's Coordinated Data Anal-<sup>75</sup> ysis Website (CDAWeb). Note that while the Fe-group flux reported by ULEIS technically includes <sup>76</sup> all species with Z > 26, the elemental abundance of minor species in the solar wind in this range is <sup>77</sup> dominated by Fe (Z=26) (e.g., Meyer 1985; Bochsler 1987). We also note that prior to mid-2001, the <sup>78</sup> ULEIS data occasionally suffered from saturated count rates for the largest SEP events [*G. Mason,* <sup>79</sup> *priv. comm.*, 2023]; thus, we restrict our analysis to the ~21-year time period 2002–2023.

Figure 1 shows the monthly averaged flux of Fe-group SEPs from 2002 to 2023 over two different 80 energy ranges: (i) the full energy range measured by ULEIS, 0.035 < E < 3.07 MeV/nuc, and 81 (ii) the approximate energy range within which Fe-group SEPs are expected to generate observable 82 tracks, 0.50 < E < 3.07 MeV/nuc (discussed below; see also Szenes et al. 2010). Note that Fe-group 83 SEPs with energies greater than this range will produce tracks deeper within a material once they 84 have shed sufficient excess energy and thus, could also contribute to track densities; however, the 85 steep slope of the energy distribution (discussed below) implies that the exclusion of such higher-86 energy particles does not overly affect our results. Both curves are similar in shape, displaying both 87 short-term variation due to individual impulsive CMEs and/or solar flares and long-term variation 88 corresponding to the 11-year solar cycle for solar cycles 23, 24, and the beginning of solar cycle 89 25. For both curves, the respective horizontal dotted lines denote the mean flux over this time 90 range, specifically  $3.2 \times 10^6$  cm<sup>-2</sup> yr<sup>-1</sup> str<sup>-1</sup> for the full energy range and  $3.8 \times 10^5$  cm<sup>-2</sup> yr<sup>-1</sup> str<sup>-1</sup> 91 for the E > 0.50 MeV/nuc range. Figure 2 shows the differential flux as a function of energy-92 per-nucleon for Fe-group SEPs observed by ULEIS averaged over the full time period presented in 93 Figure 1. As shown by the fitted curve, the differential spectrum is well described by a power law, 94  $J_{Fe}(E) = 2.3 \times 10^5 \cdot E^{-1.70} \text{ cm}^{-2} \text{ yr}^{-1} \text{ str}^{-1} (\text{MeV/nuc})^{-1}$ . Based on an analysis of lunar sample 95 64455, Blanford et al. (1974) found that a long-term-averaged SEP spectral slope of  $\gamma = -1.9$  was 96 consistent with the observed solar flare track density distribution versus depth. This spectral slope 97 is slightly steeper than that measured by ACE ( $\gamma = -1.70$ ), but within reason given the different 98 observational approaches. 99

We also verified the differential Fe-group flux measured by ACE by comparison to concurrent Fegroup measurements in a slightly lower energy range of 0.03-0.5 MeV/nuc by the Supra-Thermal

Energetic Particle (STEP) subsystem on the Energetic Particle: Acceleration, Composition, and Transport (EPACT) investigation on the Wind spacecraft (von Rosenvinge et al. 1995). Within quoted energy resolution and error bars, the differential Fe-group flux measured by Wind/STEP matches that reported by ACE.

106

100

101

# 3. INFERRING TRACK PRODUCTION RATES AT 1 AU

Using the time-averaged Fe-group SEP flux measured by ACE, we employ a simple analytical model 107 to calculate the SEP-induced track density as a function of depth in lunar and/or meteoritic materials 108 at 1 au. We obtained the electronic stopping power as a function of energy for Fe incident on an 109 forsterite grain (Mg:Si:Fe:O = 27:12:4:56; matching that of Szenes et al. (2010)) from the TRIM.SP 110 code (Ziegler et al. 2010), shown in Figure 3. In this energy range (E > 0.01 MeV/nuc), the electronic 111 stopping power dominates over the nuclear stopping power and peaks near 1.5 MeV/nuc. Previous 112 laboratory work has shown that track formation in insulators occurs only when incident particles 113 deposit energy above a given linear energy density threshold. Using a forsterite sample, Szenes et al. 114 (2010) have shown that 56 MeV Fe (1.0 MeV/nuc) ions leave tracks with nearly unit efficiency, while 115 48 MeV Ar (1.2 MeV/nuc) ions do not register any tracks. The 1.0 MeV/nuc Fe ions have a peak 116 electronic stopping power of 9.9 keV/nm (green line, Figure 3) while the 1.2 MeV/nuc Ar ions have an 117 electronic stopping power of 6.9 keV/nm (red line, Figure 3). Szenes et al. (2010) further present an 118 analytical formula for the threshold electronic stopping power,  $S_{et}$ , above which particles will induce 119 track formation and below which, they will not. From their experiments, Szenes et al. (2010) derive 120 a threshold value,  $S_{et} = 9.04 \text{ keV/nm}$  (horizontal line, Figure 3), consistent with the registration of 121 tracks from 1.0 MeV/nuc Fe but not 1.2 MeV/nuc Ar. Adopting this threshold, we estimate that 122 Fe SEPs must fall within an energy range, 0.50 < E < 3.2 MeV/nuc, in order to register track 123 formation within forsterite minerals. Note that other minerals will have slightly different electronic 124 stopping powers and thus, slightly different energy ranges to which they are susceptible to SEP 125 track formation. We also note that experimental and computational studies have shown that ions 126

#### POPPE ET AL.

with energies on opposite sides of the Bragg peak have different electronic stopping power thresholds 127 for track formation (the so-called 'velocity effect'; e.g., Constantini et al. 1992; Szenes et al. 2010; 128 Rymzhanov et al. 2019) which could affect the overall energy range for track formation. These 129 experiments have also shown that the effect is primarily manifested as higher electronic stopping 130 power thresholds (i.e., reduced track formation rates) at energies *above* the Bragg peak. However, 131 considering the steep slope of the differential flux shown in Figure 2, use of a constant  $S_{et}$  as opposed 132 to a non-linear threshold that takes into account the velocity effect is likely to have only a minor 133 effect on the overall track production rate calculated here. 134

In the analytic model, we calculate the track production rate,  $d\rho/dt$ , as a function of depth, z, by integrating the incident Fe-group SEP flux via,

$$\frac{d\rho(z)}{dt} = \pi \int_{E_{min}(z)}^{E_{max}(z)} J_{Fe}(E, z=0) \ dE,$$
(1)

where  $J_{Fe}(E, z = 0)$  is the differential Fe SEP flux at the surface of the grain as derived above and 138 shown in Figure 2,  $[E_{min}(z), E_{max}(z)]$  are the minimum and maximum energies of the upstream 139 distribution that are capable of registering tracks at depth z, and the factor of  $\pi$  accounts for the 140 exposed solid angle of a point on the lunar surface (see also Fraundorf et al. 1980). To determine 141  $[E_{min}(z), E_{max}(z)]$ , we numerically integrated the penetration of Fe SEPs into the mineral surface 142 using the electronic stopping power shown in Figure 3. This step allows the model to correctly 143 account for SEPs that are initially above the 3.2 MeV/nuc threshold, yet begin to produce tracks 144 at greater depths once they have shed sufficient energy to fall within the 0.50 < E < 3.2 MeV/nuc 145 range. For simplicity, we assume all SEPs to be normally incident to the surface. Finally, to compare 146 with the results of Keller et al. (2021), who measured the track density as a function of depth for the 147 2 Myr-exposed lunar rock 64455, we multiplied  $d\rho(z)/dt$  by  $2 \times 10^6$  yr to obtain the track density 148 versus depth,  $\rho(z)$ . 149

Figure 4 compares the analytic derivation for  $\rho(z)$  described above and the data reported from Keller et al. (2021). The analytical track density calculation based on the ACE-measured Fe SEP flux yields a maximum track density at the surface ( $z = 0.01 \ \mu$ m) of  $2.8 \times 10^{12} \ cm^{-2}$  with a gradual

137

decrease as a function of depth. At 100  $\mu$ m depth, the track density has fallen to approximately 3 × 10<sup>11</sup> cm<sup>-2</sup>. Comparing to the Keller et al. (2021) results, the ACE-calculated track density has a nearly identical shape with respect to depth, but is ~ 25× higher; the dashed curve denotes the ACE-calculated flux divided by 25 to illustrate this comparison. To first order then, the track production rates derived from in-situ Fe-group SEP measurements are in conflict with sample-derived track production rates reported in Keller et al. (2021). Below, we discuss possible reasons for this discrepancy.

160

### 4. DISCUSSION

The overestimation of the in-situ particle flux-derived track density derived from ACE relative to 161 the lunar sample-derived track density suggests that some process is acting to either suppress track 162 formation (relative to our current understanding of track formation) or erase tracks at some rate 163 after they have formed. Here, we discuss several possible hypotheses that could account for such an 164 effect, including (i) variations in the efficiency of SEP-induced track registration within meteoritic 165 minerals, (ii) thermal annealing of tracks, (iii) grain and track erosion processes, (iv) shielding of 166 SEP fluxes locally at the Moon compared to L1, (v) long-term variations in the SEP flux at 1 au, 167 and (vi) uncertainties in track-density measurement techniques; however, we note that each of these 168 hypotheses suffers in some critical way and a clear resolution is not yet in hand. 169

170

## 4.1. Track Registration Efficiency

Our calculations of track production rates based on in-situ observed particle fluxes require knowl-171 edge of the threshold electronic stopping power required for track registration (e.g., Szenes et al. 172 2010), which is likely to vary across different minerals. Thus, changes in the assumed threshold could 173 impact the total track production rate. To explore this, we repeated our calculations in Equation 1 174 using the same input Fe SEP flux but with progressively higher electronic stopping power thresholds 175 (i.e., implying a less sensitive mineral for track formation). We found that the  $25 \times$  lower track 176 production rate could only be achieved if the electronic stopping power threshold was increased to 177 nearly the maximum observed (i.e., 99.95% of the maximum), such that Fe SEPs only induced track 178

#### POPPE ET AL.

formation over an incredibly narrow range of energies ( $\approx 1.34 - 1.53$  MeV/nuc). We consider such 179 "fine-tuning" of the electronic stopping power threshold to be unrealistic, in particular in the face 180 of significant experimental evidence that SEP Fe ions over a broader range of energies can induce 181 track formation with unit efficiency (e.g., Fleischer et al. 1965; Seitz et al. 1970; Price et al. 1973; 182 Szenes et al. 2010). Additionally, such a narrow energy range for track formation would lead to the 183 formation of exceedingly short tracks ( $\sim 20$  nm); however, track lengths many tens of microns are 184 routinely observed in space-exposed minerals (e.g., Blanford et al. 1974; Bull & Durrani 1975; Keller 185 et al. 2021). Nevertheless, additional laboratory measurements that methodically characterize the 186 track registration efficiency in a variety of minerals over a broad range of incident energies could help 187 to better elucidate the exact energy range within which track formation occurs. 188

189

## 4.2. Track Annealing

SEP tracks within materials can be annealed via exposure to high temperatures, which promotes 190 atomic mobility within the crystal lattice. Early work by Price et al. (1973) suggested that at 191 maximum lunar surface temperatures ( $\sim 130$  °C), thermal annealing of SEP-induced damage tracks 192 could be effective on timescales of  $\sim 10^5 - 10^6$  years (see their Figure 9), which could plausibly affect the 193 comparison between ACE-measured and lunar-derived SEP track densities. However, the suggestion 194 by Price et al. (1973) relied on extrapolation of annealing at much higher temperatures and shorter 195 timescales and other experiments have not supported this. Tracks in most minerals do not show 196 appreciable annealing for temperatures below ~400 °C (e.g., Bull & Durrani 1975; Afra et al. 2014), 197 which is far above temperatures encountered on the lunar surface. Furthermore, as discussed in e.g., 198 Paul & Fitzgerald (1992), tracks undergoing annealing typically display a characteristic behavior in 199 which a single continuous track develops gaps along its axis as individual portions of the track anneal 200 (see their Figure 6). However, no such 'gapped' tracks indicative of thermal annealing have been 201 reported in lunar sample 64455 (Keller et al. 2021), suggesting that annealing of lunar samples-even 202 on geologic timescales—is not occurring. 203

204

# 4.3. Grain Erosion Mechanisms

Regolith grains exposed to space are subject to erosive processes, chief among which is sputtering 205 of individual atoms via incident charged particles (e.g., Biersack & Eckstein 1984; Szabo et al. 2018). 206 Decades of laboratory measurements have quantified the sputtering yield of silicate surfaces subject 207 to ion bombardment in the keV energy range (e.g., Biersack & Eckstein 1984). Using typical values 208 for the solar wind flux at 1 au and the combined proton and alpha sputtering yield, grains at 1 au 209 are eroded via charged-particle sputtering at a rate of  $\sim 7 \ \mu m/Myr$ . Thus, over the 2 Myr exposure 210 of lunar sample 64455, we would expect  $\sim 14 \ \mu m$  of erosion. To account for this erosion rate in the 211 accumulation of tracks, we developed a simple Monte Carlo model whereby tracks are numerically 212 created within a model grain with a depth profile determined from the ACE measurements as shown 213 in Figure 2 and simultaneously eroded from the top down (i.e., from the exposed grain surface) 214 at the 7  $\mu$ m/Myr sputtering rate. After  $\approx 1.3$  Myr, the track density versus depth profile reached 215 an equilibrium, shown in Figure 4 as the orange curve. Even when accounting for charged-particle 216 sputtering, the track density at the grain surface is  $\sim 1.3 \times 10^{12}$  cm<sup>-2</sup>, lower than the value without 217 sputtering,  $2.8 \times 10^{12}$  cm<sup>-2</sup>, yet still a factor of ~14 higher than that measured by Keller et al. (2021). 218 Thus, charged-particle sputtering, while likely reducing the track density somewhat, is insufficiently 219 intense to explain the observed discrepancy between ACE and lunar sample 64455. 220

221

# 4.4. SEP Shielding at the Moon

Discrepancies between the SEP flux measured by ACE at the Earth-Sun Lagrange 1 point and 222 the Moon could in theory arise due to local shielding of the lunar surface from SEPs. Remanent 223 crustal magnetic fields are widespread across the lunar surface, with magnitudes up to at least 224 hundreds of nanotesla (e.g., Mitchell et al. 2008). In-situ particle measurements have shown that 225 some crustal fields are of sufficient strength and coherency to reflect keV-energy solar wind protons 226 (e.g., Lue et al. 2011; Saito et al. 2012; Poppe et al. 2017) likely due to the formation of quasi-static 227 electric fields within the anomaly interaction regions (e.g., Fatemi et al. 2015; Deca et al. 2015). At 228 MeV energies, however, neither magnetic fields nor quasi-static electric fields are thought capable of 229 reflecting particles. MeV-scale electric fields are exceedingly unlikely to exist within such anomalies 230 and a 1 MeV/nuc  ${}^{56}$ Fe<sup>20+</sup> SEP in the presence of a 1000 nT field has a gyroradius of ~400 km, which 231

is much larger than the coherency scale of most magnetic anomalies. Thus, the presence of lunar
crustal magnetic fields are unlikely to provide any shielding to lunar soil from 1 MeV/nuc Fe-group
SEPs.

An additional source of discrepancy between L1-measured SEP fluxes and those at the lunar surface 235 could come from the Moon's transit through the terrestrial magnetotail each lunation; however, this 236 is also unlikely for two reasons. First, the Moon only spends approximately one quarter of its orbit 237 in the magnetotail which plainly cannot account for the factor of 25 difference discussed above. 238 Furthermore, recent analysis of in-situ particle measurements at the Moon have shown that SEPs 239 likely have broad access to the lunar environment even within the terrestrial magnetotail due to the 240 'open' nature of magnetotail lobe field lines to the solar wind (Liuzzo et al. 2023). Finally, shielding 241 of specific locations on the lunar surface by the solid obstacle of the Moon itself, while highly effective 242 at keV energies (e.g., Fatemi et al. 2012), appears to yield only small or even negligible results at 243 MeV energies (e.g., Xu et al. 2017). Nevertheless, in-situ SEP measurements placed at the lunar 244 surface, both on the nearside and farside for comparison, could help to better constrain any local or 245 regional shielding effects. 246

247

#### 4.5. Long-term SEP Variability

Finally, we consider the possibility that the in-situ measurements from the ACE spacecraft during 248 the modern space age are not representative of the 2 Myr-averaged SEP flux presumably recorded 249 by sample 64455. In other words, was lunar sample 64455 exposed on the lunar surface during an 250 extended solar minimum after which the modern age is a kind of grand maximum in solar activ-251 ity? While variations over multiple time scales in solar and stellar behavior are a well-documented 252 phenomenon (e.g., Usoskin 2023) and solar cycles 17–23 (~1940-2000) are considered a 'Modern Max-253 imum', both sunspot measurements over the past  $\sim 300$  years (e.g., Usoskin et al. 2016b; Muscheler 254 et al. 2016; Carrasco et al. 2016) and cosmogenic radionuclide data over the past several millenia (e.g., 255 McCracken et al. 2013; Usoskin et al. 2016a) do not suggest that the current space-age measurements 256 are exceedingly atypical. That acknowledged, the current available history of solar activity ( $\sim 10^4$  yrs) 257 falls well short of characterizing the  $2 \times 10^6$ -year exposure age of lunar sample 64455 and thus, does 258

<sup>259</sup> not entirely rebut the question. Nevertheless, the idea that the recent 10,000 years are representative <sup>260</sup> of an extreme maximum nearly  $25 \times$  higher than the million-year average is not particularly tenable <sup>261</sup> and thus, we adopt the position that—at least to first order—the modern space-age measurements <sup>262</sup> taken by ACE are reasonably representative of the past two million years.

263

# 4.6. Uncertainties in Track-Density Measurement Techniques

As noted in the Introduction, recent TEM measurements of SEP-induced tracks in meteoritic 264 materials by Keller et al. (2021) have revised the sample-based track accumulation rate at 1 au 265 downwards by a factor of  $\sim 20$  relative to earlier chemical etching-based experiments by Blanford 266 et al. (1974). The earlier track accumulation rate from Blanford et al. (1974) is closer to the ACE-267 derived value (only a factor of  $\sim 4$  lower); however, as discussed in Keller et al. (2021), the TEM 268 measurements are believed to be a more accurate measurement of the track density. In particular, 269 the TEM measurements are made with relatively thin ( $\sim 100-150$  nm thick) slices of the Apollo 270 lunar sample thereby ensuring a 'local' measurement as a function of depth and with respect to the 271 typical track length ( $\sim 5-15 \ \mu m$ ), while the chemical etching approach used in Blanford et al. (1974) 272 requires an effective integration over depths of  $10-15 \ \mu m$ . Thus, chemical etching samples a much 273 larger volume which in turn yields an SEP track density that is likely biased to large values relative 274 to the TEM measurements. 275

We also note that transmission electron microscope (TEM) measurements can induce fading of 276 SEP-induced damage tracks in minerals (e.g., Fraundorf et al. 1980; Bradley et al. 1984). Such track 277 fading was particularly noted at electron energies of 100 keV with less pronounced fading at higher 278 energies of 200 keV, where interaction cross sections are typically lower. The TEM measurements by 279 Keller et al. (2021) were conducted at 200 keV electron energies where such fading is not expected to 280 be significant; however, a quantitative analysis of the degree of track fading at 200 keV irradiation 281 has not been fully undertaken. Nevertheless, we would not expect track fading from 200 keV TEM 282 irradiation to cause the erasure of  $\sim 95\%$  of SEP damage tracks, which is what would be required to 283 explain the difference between the Keller et al. (2021) results and the ACE in-situ measurements. 284

#### 12

### POPPE ET AL.

285

## 5. CONCLUSION

We have presented a calculation of track-inducing Fe-group SEPs measured at 1 au by the 286 ACE/ULEIS instrument, deriving a flux of  $6 \times 10^5$  cm<sup>-2</sup> s<sup>-1</sup> str<sup>-1</sup>. In comparison, the track ac-287 cumulation rate determined by laboratory analysis of lunar sample 64455, which was exposed to SEP 288 fluxes on the lunar surface for  $\sim 2$  Myr, is approximately 25 times lower at  $8 \times 10^3$  cm<sup>-2</sup> s<sup>-1</sup> str<sup>-1</sup>. 289 As discussed above in Section 4, we have considered several possibilities in attempting to explain 290 the difference between the ACE-measured fluxes and those calculated from analysis of lunar sample 291 64455. Despite this, no obvious solution for this disagreement is apparent. While previous work has 292 demonstrated the efficiency of track formation with various minerals at discrete individual energies 293 (e.g., Price et al. 1973; Szenes et al. 2010), we would suggest a more thorough investigation. In 294 particular, an experiment that documented the track registration efficiency across energies spanning 295 the range predicted to induce track formation (i.e.,  $\sim 0.5$ -3.0 MeV/nuc) would help to better calibrate 296 the range over which to integrate in-situ measured SEP fluxes. Such experiments could also examine 297 a variety of mineral phases in order to further constrain any composition-related variations in track 298 registration efficiency. Additionally, a search for other appropriately suitable lunar samples (whether 299 in the current Apollo collection or to be returned from the upcoming Artemis missions to the Moon) 300 whose SEP-induced track densities over a known lifetime could be compared to those derived from 301 64455 would provide an additional validation of the results reported in Keller et al. (2021). 302



Figure 1. The monthly averaged Fe SEP flux measured by ACE/ULEIS for two energy ranges: (blue) 0.03 - 3.0 MeV/nuc and (red) 0.50 - 3.0 MeV/nuc. Average values for each separate energy range are shown as dashed lines. The SEP track formation flux at 1 au inferred from Keller et al. (2021) is shown as the black dashed line.



Figure 2. The differential energy spectrum of Fe SEPs measured by ACE/ULEIS between 0.03 MeV/nuc and 3.0 MeV/nuc. The best-fit power law spectrum is denoted by the dashed red line. The approximate energy range in which Fe-group SEPs leave damage tracks in meteoritic materials is denoted by the vertical dotted lines.

A. P. gratefully acknowledges support from the NASA New Frontiers Data Analysis Program, grant
 #80NSSC18K1557. A. P. thanks G. Mason for useful discussions on the ACE/ULEIS instrument.



Figure 3. The electronic stopping power,  $S_e$ , for three incident ion species (Ar, red; Fe, green; Ni, blue) in a forsterite mineral. The green closed circle and red open circle represent experimental measurements by Szenes et al. (2010) that did and did not register tracks, respectively. Correspondingly, the minimum required  $S_e$  for track formation estimated by Szenes et al. (2010) is shown as the horizontal line.

## REFERENCES

306       Nuc. Instr. Meth. Phys. Res. B, 326, 126       317       Lunar Sci. Conf., 6, 3619         307       Biersack, J. P., & Eckstein, W. 1984, Appl. Phys.       318       Carrasco, V. M. S., Aparicio, A. J. P.,         308       A, 34, 73       319       J. M., & Gallego, M. C. 2016, Solar         309       Blanford, G. E., Fruland, R. M., McKay, D. S., &       3045, doi: 10.1007/s11207-016-0998-         310       Morrison, D. A. 1974, Proc. 5th Lunar Conf., 3,       321         311       2501       321         312       Bochsler, P. 1987, Physica Scripta, T18, 55       Lunar Sci. Conf., 6, 3619
<ul> <li>Biersack, J. P., &amp; Eckstein, W. 1984, Appl. Phys.</li> <li>A, 34, 73</li> <li>Blanford, G. E., Fruland, R. M., McKay, D. S., &amp;</li> <li>Morrison, D. A. 1974, Proc. 5th Lunar Conf., 3,</li> <li>2501</li> <li>Bochsler, P. 1987, Physica Scripta, T18, 55</li> <li>Biersack, J. P., &amp; Carrasco, V. M. S., Aparicio, A. J. P.,</li> <li>Carrasco, V. M. S., Aparicio, A. J. P.,</li> <li>J. M., &amp; Gallego, M. C. 2016, Solar</li> <li>3045, doi: 10.1007/s11207-016-0998-</li> <li>Constantini, J. M., Brisard, F., Flame</li> <li>et al. 1992, Nuc. Instr. Meth. Phys.</li> </ul>
<sup>323</sup> 568 <sup>313</sup> Bradley, J. P., Brownlee, D. E., & Fraundorf, P. <sup>314</sup> 1984, Science, 226, 1432, <sup>315</sup> doi: 10.1126/science.226.4681.1432 <sup>320</sup> 568 <sup>324</sup> Crozaz, G., Drozd, R., Hohenberg, C. <sup>325</sup> 1972, Proc. 3rd Lunar Sci. Conf., 3,



Figure 4. The predicted track density as a function of depth for an objects exposed for 2 Myr at 1 au as determined by the ACE in-situ measurements (solid line). Solid dots reproduce the measurements of Keller et al. (2021) along with the ACE-predicted flux lowered by a factor of 25 (dashed line). The orange line is the result of a Monte Carlo model for track formation taking into account a charged-particle erosion rate of  $7 \ \mu m/Myr.$ 

- Deca, J., Divin, A., Lembège, B., et al. 2015, J. 326
- Geophys. Res.: Space Physics, 120, 6443 327
- Fatemi, S., Holmström, M., & Futaana, Y. 2012, 328 J. Geophys. Res., 117, 1 329
- Fatemi, S., Lue, C., Holmström, M., et al. 2015, J. 330
- Geophys. Res.: Space Physics, 120 331

333

- Fleischer, R. L., Price, P. B., & Walker, R. M. 332 1965, Annu. Rev. Nucl. Sci., 15, 1
- -. 1975, Nuclear Tracks in Solids: Principles and 334
- Applications (University of California Press) 335

- Fraundorf, P., Flynn, G. J., Shirck, J., & Walker, 336 R. M. 1980, Proc. Lunar Planet. Sci. Conf. 337 11th, 11, 1235 338
- Goswami, J. N. 1981, Nature, 293, 124, 339 doi: 10.1038/293124a0 340
- Keller, L. P., & Berger, E. L. 2014, Earth Planets 341 Space, 66, doi: 10.1186/1880-5981-66-71 342
- Keller, L. P., Berger, E. L., Zhang, S., & 343
- Christoffersen, R. 2021, Meteorit. Planet. Sci., 344 56, 1685, doi: 10.1111/maps.13732 345
- Keller, L. P., & Flynn, G. J. 2022, Nature Astron., 346 6, 731, doi: 10.1038/s41550-022-01647-6 347

- Kuchner, M. J., & Stark, C. C. 2010, Astron. J.,
  140, 1007
- Liou, J.-C., & Zook, H. A. 1999, Astron. J., 118,
  580
- <sup>352</sup> Liuzzo, L., Poppe, A. R., Lee, C. O., Xu, S., &
- Angelopoulos, V. 2023, Geophys. Res. Lett., 50,
   doi: 10.1029/2023GL103990
- <sup>355</sup> Lue, C., Futaana, Y., Barabash, S., et al. 2011,
- 356 Geophys. Res. Lett., 38
- Mason, G. M., Gold, R. E., Krimigis, S. M., et al.
  1998, Space Sci. Rev., 86, 409
- 359 McCracken, K. G., Beer, J., Steinhilber, F., &
- Abreu, J. 2013, Solar Phys., 286, 609,
  doi: 10.1007/s11207-013-0265-0
- <sup>362</sup> Meyer, J. 1985, Astrophys. J. Supp. Ser., 57, 151
- Mitchell, D. L., Halekas, J. S., Lin, R. P., et al.
- <sup>364</sup> 2008, Icarus, 194, 401
- <sup>365</sup> Muscheler, R., Adolphi, F., Herbst, K., & Nilsson,
- 366 A. 2016, Solar Phys., 291, 3025,
- 367 doi: 10.1007/s11207-016-0969-z
- Paul, T. A., & Fitzgerald, P. G. 1992, Am.
  Mineral., 77, 336
- <sup>370</sup> Poppe, A. R., Halekas, J. S., Lue, C., & Fatemi, S.
- <sup>371</sup> 2017, J. Geophys. Res.: Planets, 122
- <sup>372</sup> Poppe, A. R., Lisse, C. M., Piquette, M., et al.
- <sup>373</sup> 2019, Astrophys. J. Lett., 881,
- doi: https://doi.org/10.3847/2041-8213/ab322a
- Price, P. B., Lal, D., Tamhane, A. S., & Perelygin,
  V. P. 1973, Earth Plan. Sci. Lett., 19, 377
- <sup>377</sup> Price, P. B., & O'Sullivan, D. 1970, Proc. Apollo
- <sup>378</sup> 11 Lunar Sci. Conf., 3, 2351

- Rymzhanov, R. A., Gorbunov, S. A., Medvedev,
  N., & Volkov, A. E. 2019, Nuc. Instr. Meth.
  Phys. Res. B, 440, 25,
- doi: 10.1016/j.nimb.2018.11.034
- Saito, Y., Nishino, M. N., Fujimoto, M., et al.
  2012, Earth Planets Space, 64, 83
- 385 Sandford, S. A. 1986, Icarus, 68, 377,
  doi: 10.1016/0019-1035(86)90045-X
- Seitz, M., Wittels, M. C., Maurette, M., Walker,
  R. M., & Heckman, H. 1970, Rad. Effects, 5, 143
- 389 Stone, E. C., Frandsen, A. M., Mewaldt, R. A.,
- <sup>390</sup> et al. 1998, Space Sci. Rev., 86,
- doi: 10.1007/978-94-011-4762-0\_1
- <sup>392</sup> Szabo, P. S., Chiba, R., Biber, H., et al. 2018,
- <sup>393</sup> Icarus, 314, 98, doi: 10.1016/j.icarus.2018.05.028
- Szenes, G., Kovács, V. K., Pécz, B., & Skuratov,
  V. 2010, Astrophys. J., 708, 288,
  doi: 10.1088/0004-637X/708/1/288
- Thiel, K., Bradley, J. P., & Spohr, R. 1991, Nucl.
  Tracks Radiat. Meas., 19, 709,
  doi: 10.1016/1359-0189(91)90298-V
- <sup>400</sup> Usoskin, I. G. 2023, Living Rev. Sol. Phys., 20,
  <sup>401</sup> doi: https://doi.org/10.1007/s41116-023-00036-z
- <sup>402</sup> Usoskin, I. G., Gallet, Y., Lopes, F., Kovaltsov,
  <sup>403</sup> G. A., & Hulot, G. 2016a, Astron. Astrophys.,
  <sup>404</sup> 587, doi: http:
- 405 //dx.doi.org/10.1051/0004-6361/201527295
- <sup>406</sup> Usoskin, I. G., Kovaltsov, G. A., Lockwood, M.,
  <sup>407</sup> et al. 2016b, Solar Phys., 291, 2685,
  <sup>408</sup> doi: 10.1007/s11207-015-0838-1
- von Rosenvinge, T. T., Barbier, L. M., Karsch, J.,
  et al. 1995, Space Sci. Rev., 71, 155

- <sup>411</sup> Xu, X., Angelopoulos, V., Wang, Y., et al. 2017,
- 412 Astrophys. J., 849,
- 413 doi: 10.3847/1538-4357/aa9186

- <sup>414</sup> Ziegler, J. F., Ziegler, M. D., & Biersack, J. P.
- <sup>415</sup> 2010, Nuc. Instr. Meth. Phys. Res. B, 268, 1818
- <sup>416</sup> Zinner, R. 1980, in The ancient Sun: Fossil record
- <sup>417</sup> in the Earth, Moon, and meteorites, ed. R. O.
- <sup>418</sup> Pepin, J. A. Eddy, & R. B. Merrill (Pergamon
- <sup>419</sup> Press), 201–226