

*Meteoritics & Planetary Science* 42, Nr 10, 1831–1840 (2007) Abstract available online at http://meteoritics.org

# <sup>3</sup>He, <sup>20,21,22</sup>Ne, <sup>14</sup>C, <sup>10</sup>Be, <sup>26</sup>Al, and <sup>36</sup>Cl in magnetic fractions of cosmic dust from Greenland and Antarctica

# A. J. T. JULL<sup>1\*</sup>, D. LAL<sup>2</sup>, S. TAYLOR<sup>3</sup>, R. WIELER<sup>4</sup>, A. GRIMBERG<sup>4</sup>, L. VACHER<sup>2</sup>, L. R. MCHARGUE<sup>1, 5</sup>, S. P. H. T. FREEMAN<sup>5</sup>, C. MADEN<sup>5</sup>, C. SCHNABEL<sup>5</sup>, S. XU<sup>5</sup>, R. C. FINKEL<sup>6</sup>, K. J. KIM<sup>1, 8</sup>, and K. MARTI<sup>7</sup>

<sup>1</sup>NSF Arizona AMS Laboratory, University of Arizona, Physics Building, 1118 East Fourth Street, Tucson, Arizona 85721, USA

<sup>2</sup>Scripps Institution of Oceanography, Geologic Research Division, University of California San Diego, La Jolla, California 92093, USA

<sup>3</sup>Cold Regions Research Laboratory, Hanover, New Hampshire 03755, USA

<sup>4</sup>Isotope Geology and Mineral Resources, NW C84, ETH Zürich, 8092 Zürich, Switzerland

<sup>5</sup>Scottish Universities Environmental Research Centre, East Kilbride, G75 0QF, UK

<sup>6</sup>Center for Accelerator Mass Spectrometry, Lawrence Livermore National Laboratory, Livermore, California 94550, USA

<sup>7</sup>Department of Chemistry, University of California San Diego, La Jolla, California 92093, USA

<sup>8</sup>Korea Institute of Geoscience and Mineral Resources, 30 Gajeon-dong, Yuseong-gu, Daejeon 305-350, Korea

\*Corresponding author. E-mail: jull@email.arizona.edu

(Received 15 March 2006; revision accepted 25 April 2007)

Abstract–We report on studies of the concentrations of cosmogenic nuclides in the magnetic fraction of cosmic dust particles recovered from the South Pole Water Well (SPWW) and from Greenland. Our results confirm that cosmic dust material from these locations contains measurable amounts of cosmogenic nuclides. The Antarctic particles (and possibly those from Greenland as well) also contain minor amounts of solar Ne. Concentrations of cosmogenic nuclides are consistent with irradiation of this material as small objects in space, with exposure ages similar to the expected Poynting-Robertson (P-R) lifetimes of 50–200 kyr for particles 25–100 µm in size.

# **INTRODUCTION**

Material from space of all size ranges impacts the Earth and accretes at the rate of approximately 30,000 tons/yr, a number estimated from the impact rate on the Long-Duration Exposure Facility (LDEF) (Love and Brownlee 1993). Smaller cosmic dust particles are expected to spiral in toward the sun due to Poynting-Robertson drag caused by photons (Burns et al. 1979; Liou et al. 1995), as well as the further effect of solar-wind drag (Burns et al. 1979; Liou et al. 1995). Upon entry into the terrestrial atmosphere, incoming cosmic material is heated to at least 600 K (grains of  $\sim 6 \mu m$  radius) and to an average of about 1200 K (Flynn 2001), depending on the particle size. Spalling of smaller material from larger objects can also be expected to contribute to the flux at the Earth's surface (Brownlee 2001; Lal and Jull 2002). Xue et al. (1997) estimated from fractionation studies that perhaps 80% of the original spalled material might be lost due to heating during entry. In addition, Taylor et al. (1998) estimated a 90% loss of original material using a direct measurement of particles arriving on Earth, but this measurement was from only one location and time period. Hence, the purpose of this paper is to investigate whether we gain new information on the sources of cosmic dust using cosmogenic nuclides.

In isotopic studies, Merrihue (1964) was the first to note that excess <sup>3</sup>He in marine sediments could be attributed to the infall of cosmic debris. Rajan et al. (1977) began studies that provided evidence of cosmogenic noble gas isotopes in cosmic dust. This result was confirmed by Hudson et al. (1980) and demonstrated independently by Ozima et al. (1984). Olinger et al. (1990) established that implanted solar wind and cosmogenic and solar noble gases could be identified in material separated from Greenland ice samples. Later work was carried out by Farley and Patterson (1995). Marcantonio et al. (1995), and Farley et al. (1997), and summarized by Winckler et al. (2004). Detailed studies of individual cosmic dust particles began with the recovery of magnetic cosmic spherules from the ocean floor (Bradley and Brownlee 1982) and later of interplanetary dust particles (IDPs) by high-altitude aircraft (Brownlee 1985). Nier and Schlutter (1990, 1993), Kehm et al. (1998, 2002, 2006), and Pepin et al. (1999, 2000, 2001) have all reported on cosmogenic and solar gases in IDPs.

The conventional definition is that IDPs are particles collected in the atmosphere, and cosmic dust is material that actually makes it to the Earth's surface. There is a limited amount of information on cosmogenic radionuclides in cosmic dust. Previously, Raisbeck et al. (1983, 1985),



Fig. 1. Separation scheme for magnetite-rich cosmic dust material from Antarctic ice from the South Pole Water Well (SPWW) (Taylor et al. 1998) and Greenland blue lake sediment (GR) (Maurette et al. 1990). Note that the strongly magnetic fractions were divided into two fractions. The samples with the suffix SM were used for the wetchemical extractions of  $^{14}C$ ,  $^{10}Be$ ,  $^{26}Al$ , and  $^{36}Cl$ ; the numbered fractions were used for the dry combustions.

Raisbeck and Yiou (1989), and Nishiizumi et al. (1991) carried out <sup>26</sup>Al and <sup>10</sup>Be studies on large cosmic spherules recovered from deep-sea sediments. Nishiizumi et al. (1991) also studied four particles from the Greenland ice cap; Nishiizumi et al. (1995, 2007) discussed <sup>10</sup>Be and <sup>26</sup>Al in individual cosmic spherules recovered from Antarctic ice. There have also been many other studies on cosmic dust in polar ice and marine sediments, but few focused on the cosmogenic nuclides, let alone radionuclides. Brook et al. (2000) studied the accretion of interplanetary dust in polar ice using <sup>3</sup>He. Furthermore, Farley and Patterson (1995) and Farley et al. (1997) looked at long-term changes of the in-fall rate of cosmic dust in the geologic past using <sup>3</sup>He in ancient oceanic sediments. Similarly to the work of Farley et al. (1997), Lal and Jull (2002) indirectly noted the presence of cosmic dust through observations of high levels of <sup>14</sup>C in one sample of 2 kg of Greenland ice melted as part of a program designed to study in-situ-produced cosmogenic <sup>14</sup>C in the ice. They calculated that cosmic dust material of 0.02 g could explain this result.

### PURPOSE

The purpose of this paper is to demonstrate that a fraction of the particles recovered in polar regions from the South Pole Water Well (SPWW) and from Greenland are extraterrestrial. We present the first preliminary studies of the concentrations of cosmogenic radionuclides (as opposed to stable nuclides) in 0.1–1 g amounts of the magnetic phase of dust particles recovered from polar regions, which could consist of a mixture of terrestrial and extraterrestrial material. We also report He and Ne data on aliquots of these samples. We will first show that at least part of the material from both locations is extraterrestrial. We then review the combined data and discuss whether the extraterrestrial material was mainly delivered by direct infall of cosmic dust or as fragments spalled from larger objects, as suggested by Lal and Jull (2002).

## **EXPERIMENTS**

#### Sample Selection

We obtained samples of micrometeorite debris from the SPWW, described in detail by Taylor et al. (1998, 2000, 2005, 2007). Taylor et al. (2000) estimated micrometeorite influx rates from cosmic spherules in SPWW and estimated an accretion rate of  $1100 \pm 200$  tons/yr for particles in the 50–300 µm size range. The samples studied here come from ice estimated to date from approximately 1100-1500 A.D. (Taylor et al. 2000). There is possible contamination in the samples from iron oxide from a failed pump, as well as by some copper and plastic. The samples from Greenland (GR) were collected from blue lakes by Maurette et al. (1987, 1990). The GR samples were rich in Fe and Si, whereas the samples from the SPWW were rich in Fe and Ni, suggesting an extraterrestrial component in SPWW material.

We initially processed the samples using a simple magnetic separation, removing what material could be collected with a hand magnet at a distance of  $\sim 10$  mm (Fig. 1). The material recovered was studied by scanning electron microscopy (SEM), which showed the angular nature of the grains (Fig. 2). We analyzed the chemical compositions and isotopic concentrations of the strongly magnetic fractions of the two samples. Figure 3 shows the elemental compositions of the etched solutions used in the wet dissolution of Lal et al. (1995), as discussed above.

#### **Sample Extractions**

#### $^{14}C$

For <sup>14</sup>C, we performed two different kinds of extractions. The first was a dry combustion of the sample in oxygen gas at <200, 200–400, 400–550, and 550–800 °C, followed by fusing the sample with Fe chips in an alumina crucible that had an oxygen atmosphere at T > 800 °C. Heating the >800 °C extractions was achieved using a radio-frequency (RF) induction furnace, similar to that described by Jull et al. (1998). The results of the high-temperature combustions are shown in Table 1. Below 400 °C, <sup>14</sup>C is released, which we identify as terrestrial organic material and contamination, as shown by Jull et al. (1998). Dry combustion releases cosmogenic <sup>14</sup>C in 400–550, 550–800, and >800 °C fractions (Jull et al. 1998). According to this approach, all these fractions thus contain



Fig. 2. Scanning electron microscope photographs of the material recovered from the strongly magnetic (SM) fractions shown in Fig. 1.



Fig. 3. Major-element analyses of solutions (in wt%) recovered from HF- and HF-HNO<sub>3</sub>-leaching of strongly magnetic (SM) grains recovered from Antarctic ice melted at the SPWW and Greenland blue lake sediment (GR). Two separate analyses (1 and 2) are shown for each fraction.

extraterrestrial <sup>14</sup>C; the 400–550 °C fraction might contain a mixture of terrestrial and extraterrestrial <sup>14</sup>C. The second approach used a wet chemical extraction method (Lal et al. 1995) with two sequential extraction steps using HF and HNO<sub>3</sub>. Results of the wet chemical extraction are shown in Table 2.

#### <sup>3,4</sup>He and <sup>20,21,22</sup>Ne

Noble gas measurements were performed at the ETH Zürich. Gases were extracted by a continuous-wave Nd-YAG infrared laser (Vogel et al. 2003). The loose grains of each sample were put into holes in the sample holder and covered with glass to prevent possible loss of grains during heating. No preheating prior to noble gas analyses was performed. To extract the noble gases, the samples were heated until all of the grains had assembled into a single molten sphere. Reheating these spheres proved there was complete noble gas extraction in the main extraction. Mass spectrometric analyses of He and Ne and data reduction were performed as described by Vogel et al. (2003 and references therein).

#### Other Nuclides: <sup>10</sup>Be, <sup>26</sup>Al, and <sup>36</sup>Cl

Extractions for the radionuclides <sup>10</sup>Be, <sup>26</sup>Al, and <sup>36</sup>Cl were performed by HF- and HNO<sub>3</sub>-dissolution of the cosmic dust grains, followed by the appropriate wet chemistry for these nuclides (Lal et al. 1995). Measurements of the radionuclides were made by accelerator mass spectrometry at the University of Arizona (Jull and Burr 2006), the Lawrence Livermore National Laboratory, and the Scottish Universities Environmental Research Centre (Maden et al. 2006).

## **RESULTS AND DISCUSSION**

As noted in the Experiments section, we concentrated the samples of SPWW and GR using a magnetic concentration method, as shown in Fig. 1. To characterize the material, we first obtained SEM photographs (Fig. 2), then obtained the elemental composition (Fig. 3) of the etched solutions used in the wet dissolution technique of Lal et al. (1995), as discussed above. It is clear from Fig. 3 that sample SPWW consists mainly of Fe-Ni oxides. Although it is conceivable that

		Temperature	Sample weight	С	С		<sup>14</sup> C	<sup>14</sup> C
		(°C)	(g)	(mg)	(%)	$\delta^{13}C$	(dpm/kg)	σ
J683KA	SPWW	<200	0.1417	0.14	0.10%	-29.80	9.89	0.49
J683LA	SPWW	200-400	0.1417	2.78	1.96%	-25.80	107.05	0.95
J683MA	SPWW	400-550	0.1417	0.061	0.04%	-22.90	3.58	0.52
J683HA	SPWW	550-800	0.1417	1.711	1.21%	-8.10	5.89	0.67
R3087R	SPWW	>800	0.1417	0.138	0.10%	-23.50	1.95	0.49
J684K	GR	<200	0.4071	0.557	0.14%	-17.90	20.47	0.22
J684L	GR	200-400	0.4071	4.03	0.99%	(-25)	152.42	0.74
J684M	GR	400-550	0.4071	1.554	0.38%	(-25)	54.22	0.33
J684H	GR	550-800	0.4071	0.8	0.20%	-25.60	27.42	0.28
R3089R	GR	>800	0.3569	0.566	0.16%	-22.66	16.17	0.22
J685K	GR, mag <sup>a</sup>	<200	0.0501	0.071	0.14%	2.40	19.13	1.39
J685L	GR, mag	200-400	0.0501	0.871	1.74%	-20.80	254.48	1.82
J685M	GR, mag	400-550	0.0501	0.051	0.10%	Sample not	measured	
J685H	GR, mag	550-800	0.0501	0.0152	0.03%	-20.40	2.38	1.41
R3088R	GR, mag	>800	0.0396	0.054	0.14%	(-25)	5.76	1.75

Table 1. Combustions in oxygen at different temperatures for material recovered from the South Pole Water Well (SPWW) and Greenland (GR).

<sup>a</sup>Mag = highly magnetic fraction; see Fig. 1.

Table 2. <sup>14</sup> C and <sup>10</sup> Be from	acid etching of magnetic
fractions of cosmic dust.	

	<sup>14</sup> C	<sup>10</sup> Be	
	(dpm/kg)	(dpm/kg)	$^{14}C/^{10}Be$
South Pole Water Well			
1st leach: HF	$6.07\pm0.07$	$0.23\pm0.03$	26.4
2nd leach: HF-HNO <sub>3</sub>	$6.53\pm0.22$	$1.11\pm0.05$	5.88
Total	$12.60\pm0.23$	$1.34\pm0.06$	9.4
Greenland			
1st leach: HF	$62.9\pm0.6$	$0.13\pm0.06$	484
2nd leach: HF-HNO <sub>3</sub>	$3.7 \pm 0.5$	$0.01\pm0.01$	370
Total	$66.6\pm0.8$	$0.14\pm0.06$	476

oxidized fragments of stainless steel are one source of Ni in the SPWW material, the high Ni concentrations in these samples are very suggestive of an extraterrestrial component. In contrast to the Antarctic sample, the GR sample is very poor in Ni. We will discuss the radionuclide and noble gas data in the following sections.

# **Radionuclide Studies**

In order to understand the results from the studies of the SPWW and GR materials, it is first necessary to consider the expected production rates for the radionuclides in these materials. We consider the production rates of  $^{10}$ Be,  $^{14}$ C,  $^{26}$ Al, and  $^{36}$ Cl in Fe metal, Fe<sub>2</sub>O<sub>3</sub>, and Fe<sub>3</sub>O<sub>4</sub>, using the published results of Leya et al. (2000), who considered production rates in artificial irradiations of iron metal, and also Albrecht et al. (2000), who studied the production rates in the iron of mesosiderites. Furthermore, we discuss new calculations made by one of us (K. Kim) for small Fe<sub>2</sub>O<sub>3</sub> and Fe<sub>3</sub>O<sub>4</sub> objects <1 cm in size by using MCNPX computer code. These production rates are given in Table 3.

# South Pole Water Well Material

We first consider the SPWW sample. The total <sup>14</sup>C released at temperatures greater than 400 °C was  $11.4 \pm$ 1.0 dpm/kg, which is in good agreement with the value of  $12.6 \pm 0.2$  dpm/kg obtained from the combined wet extractions (Table 2). A maximum is observed in the 550-800 °C fraction, indicating good separation of the cosmogenic signal from any low-temperature organic contamination. The 10Be, 26Al, and 36Cl data from these same dissolution experiments are shown in Table 4. As noted, the adopted production rate ratios listed in Table 3 are derived from galactic cosmic ray (GCR) production rates for small dust particles estimated by various authors. Important to note is that for iron oxides, the mean measured ratio of 9.4 for <sup>14</sup>C/<sup>10</sup>Be is much higher than the production rate ratio of 0.84-0.85 for GCR, whereas the <sup>26</sup>Al/<sup>10</sup>Be ratio of 0.35 is close to the expected production ratio of about 0.31-0.33, and the <sup>36</sup>Cl/<sup>10</sup>Be ratio of 2.1 is somewhat higher than the expected production ratio of 1.4-1.5. This inconsistent picture can be explained by the presence of some <sup>14</sup>C from terrestrial contamination. The <sup>26</sup>Al/<sup>10</sup>Be ratio is evidence for GCR production for these nuclides. We do not have a good explanation for the enhanced <sup>36</sup>Cl, except perhaps that our modeling of the material as Fe oxides is inadequate. One could postulate a short exposure age, except that this would be inconsistent with the <sup>26</sup>Al/<sup>10</sup>Be.

# **Greenland Material**

The Greenland sample (GR) contained much higher <sup>14</sup>C activity in low-temperature fractions than the SPWW sample, suggesting contamination from surficial terrestrial organic material. The results show high <sup>14</sup>C (97.8 dpm/kg for the total

	Product	ion rate (cc STP/g.y)			
	In Fe metal	In Fe <sub>2</sub> O <sub>3</sub>	In silicates	-	
<sup>3</sup> He <sup>21</sup> Ne	$\begin{array}{c} 1.3 \times 10^{-14} \\ 3.0 \times 10^{-16} \end{array}$	$\begin{array}{c} 1.5\times 10^{-14} \\ 2.0\times 10^{-16} \end{array}$	$\begin{array}{c} 1.5\times 10^{-14} \\ 15\times 10^{-16} \end{array}$		
		Produc	ction rate (dpm/kg)		
	In Fe metal	In Fe <sub>2</sub> O <sub>3</sub>	In Fe <sub>2</sub> O <sub>3</sub>	In Fe <sub>2</sub> O <sub>3</sub>	In Fe <sub>3</sub> O <sub>4</sub>
Radius	10–25 cm (Leya et al. 2000)	10–25 cm (Leya et al. 2000)	<1 cm (This work)	~10–20 cm <sup>a</sup> (Albrecht et al. 2000)	<1 cm (This work)
<sup>14</sup> C	4.3	14	9.4	30	9
<sup>10</sup> Be	5.7	11.2	11	15	10.7
<sup>26</sup> Al	4.8	3.3	3.4	5.6	3.5
<sup>36</sup> Cl	23	16	15.5	17	16.1

Table 3. Estimated production rates of cosmogenic nuclides in cosmic dust.

<sup>a</sup>Estimated from the measured contents of mesosiderites; estimated sizes of radius = 10-20 cm.

Table 4. Specific activities of radionuclides derived from acid-etching.

	<sup>10</sup> Be	<sup>14</sup> C	<sup>26</sup> Al	<sup>36</sup> C1
Cosmic dust samples	(dpm/kg)	(dpm/kg)	(dpm/kg)	(dpm/kg)
SPWW	$1.34 \pm 0.06$	$12.6 \pm 0.2$	$0.47\pm0.04$	$2.89 \pm 0.20$
Ratio P/P <sub>10</sub> <sup>a</sup>	1.00	9.4	0.35	2.16
GR	$0.14 \pm 0.06$	$66.6 \pm 0.8$	$0.86\pm0.08$	Not measured
Ratio P/P <sub>10</sub>	1.00	50.8	6.14	
Production rate ratios in si	mall particles <sup>b</sup>			
Fe <sub>3</sub> O <sub>4</sub>	10.7	9.0	3.5	16.1
Ratio P/P <sub>10</sub>	1.00	0.84	0.33	1.50
Fe <sub>2</sub> O <sub>3</sub>	11	9.4	3.4	15.5
Ratio P/P <sub>10</sub>	1.00	0.85	0.31	1.41
SiO <sub>2</sub>	14.84	13.98	44.09	0.00
Ratio P/P <sub>10</sub>	1.00	0.94	2.97	0.00

<sup>a</sup>P = production rate of radionuclide;  $P_{10}$  = production rate of <sup>10</sup>Be. <sup>b</sup>From Table 3.

of all fractions >400 °C). The wet chemical results show in very high <sup>14</sup>C (66.6  $\pm$  0.80dpm/kg). Both of these results suggest terrestrial contamination of the <sup>14</sup>C signal. The <sup>10</sup>Be (0.14  $\pm$  0.06 dpm/kg) and <sup>26</sup>Al (0.86  $\pm$  0.08 dpm/kg) shows less <sup>10</sup>Be but more <sup>26</sup>Al than the SPWW sample. The <sup>26</sup>Al/<sup>10</sup>Be ratio of 6.1  $\pm$  2.6 is higher than the expected production ratio of 0.31–0.33 in iron oxides and also than the expected ratio of ~3 in silicates from GCR irradiation. The value of ~3 is also expected from eolian dust, which could be otherwise considered as a source of these low levels of <sup>26</sup>Al and <sup>10</sup>Be. Hence, we suggest this is also evidence of a small extraterrestrial component, but the very small signals and large errors mean this interpretation is not as strong as in the case of SPWW.

# He and Ne Isotopic Abundances

The results from He and Ne analysis of the SPWW and GR samples are given in Table 5. In Fig. 4, we show a plot of the Ne isotopic data. The <sup>20</sup>Ne/<sup>22</sup>Ne ratios of both SPWW samples clearly fall between the terrestrial (air) and the solar wind (SW) composition. Similarly, essentially all

<sup>3</sup>He in SPWW samples appears to be solar in origin, as indicated by <sup>3</sup>He/<sup>4</sup>He ratios that are much higher than the atmospheric value of  $1.4 \times 10^{-6}$  and are close to that of ~2.2 × 10<sup>-4</sup> for the component formerly known as "SEP," but now identified to be isotopically fractionated solar wind (Grimberg et al. 2006). The trapped noble gases therefore indicate that the Antarctic sample contains at least some extraterrestrial grains that were irradiated by solar wind. However, the amounts of solar <sup>20</sup>Ne are orders of magnitude lower than in solar-gas-rich micrometeorites (Olinger et al. 1990). No cosmogenic <sup>3</sup>He or <sup>21</sup>Ne are discernible in the SPWW samples, though this may well be because cosmogenic contributions are swamped by the solar He and Ne. However, no meaningful cosmic-ray exposure age can be given for the SPWW samples.

The case for solar noble gas contribution in the Greenland sample is less clear. One aliquot shows a trapped  ${}^{20}\text{Ne}/{}^{22}\text{Ne}$  ratio higher than the atmospheric ratio by slightly more than its  $2\sigma$  uncertainty (Fig. 4), but the second sample has trapped Ne with a  ${}^{20}\text{Ne}/{}^{22}\text{Ne}$  ratio very close to atmospheric. The  ${}^{3}\text{He}/{}^{4}\text{He}$  ratios of both aliquots are also much lower than the solar range, indicating there is also no discernible contribution

Table	5	Noble	gas	data
raute	5.	110010	gas	uata

		<sup>4</sup> He	<sup>20</sup> Ne							
Sample	Mass	(cm <sup>3</sup> STP/g)	$(cm^3 STP/g)$	<sup>3</sup> He/ <sup>4</sup> He	Error	<sup>20</sup> Ne/ <sup>22</sup> Ne	Error	<sup>21</sup> Ne/ <sup>22</sup> Ne	Error	<sup>4</sup> He/ <sup>20</sup> Ne
SPWW a	1.49 mg	1.99E-07	2.94E-09	2.78E-04	2.69E-05	11.93	0.62	3.35E-02	8.29E-03	68
SPWW b	0.90 mg	1.14E-06	7.30E-08	2.67E-04	9.22E-06	11.53	0.10	3.13E-02	1.07E-03	16
GR a	1.37 mg	7.61E-05	3.07E-09	1.55E-06	1.76E-07	11.70	0.72	4.63E-02	5.88E-03	24,800
GR b	3.89 mg	7.44E-05	5.76E-09	1.74E-07	1.71E-07	9.82	0.11	4.17E-02	3.55E-03	12,900
Solar wind				~4.5E-4		13.8	0.10	3.28E-2	5.0E-4	650

Subsamples a and b are aliquots. Sample masses given in mg; gas concentrations in cm<sup>3</sup> STP/g. Uncertainties of gas concentrations are on the order of 5%. Stated errors ( $1\sigma$ ) of isotopic ratios include measurement statistics and mass discrimination uncertainties. Solar wind composition (Wieler 2002) given as reference.



Fig. 4. Ne isotopic results for the strongly magnetic (SM) grains recovered from Antarctic ice melted at the SPWW and from GR blue lake sediments. Two separate sets of measurements on the same material, labeled "a" and "b," are shown.

of solar He. Nevertheless, the Ne data show that the GR samples also contain some extraterrestrial material. Both samples show excesses of <sup>21</sup>Ne above atmospheric composition, and the measured <sup>3</sup>He can likely also quantitatively be attributed to a cosmogenic origin (alternatively, if the <sup>3</sup>He were a remnant of solar wind implanted He, this would also prove there is an extraterrestrial contribution of material to GR). In the next paragraph, we attempt to estimate cosmic-ray exposure ages of the GR sample.

Note first that the possibility that the observed cosmogenic <sup>21</sup>Ne and <sup>3</sup>He in GR has been produced on the Earth's surface can be discarded. Although production rates are difficult to evaluate quantitatively, no values for terrestrial production of <sup>3</sup>He and <sup>21</sup>Ne on Fe have been determined experimentally, and the altitude- and shielding-history of the particles in the Greenland ice are ill-constrained. However, exposure ages on the Greenland ice cap would have to be on the order of millions of years at zero shielding, which is clearly inconsistent with the age of the ice at the sampling location (Maurette et al. 1987). It is also hardly conceivable that the cosmogenic noble gases in the GR sample are largely inherited from a previous exposure stage at some other

location, as this would also require extremely long preexposure durations at the low altitudes of potential source regions of aeolian dust.

To estimate exposure ages in space, let us first assume that 100% of the material of sample GR (including the roughly 40% silicate, Fig. 3) is extraterrestrial and that all measured <sup>3</sup>He in this sample is cosmogenic. The resulting nominal <sup>3</sup>He exposure ages of the two aliquots of this sample are then ~8000 and ~900 years, respectively. If the silicate portion is terrestrial, these values would increase by about ~20–60%; if part of the <sup>3</sup>He is noncosmogenic, they would decrease correspondingly. In any case, the calculated <sup>3</sup>He exposure ages of GR are very low.

To estimate a <sup>21</sup>Ne exposure age of GR samples is somewhat more complicated due to the ~40% silicate contribution, since <sup>21</sup>Ne production rates from Mg, Al, and Si are much higher than from Fe. The <sup>21</sup>Ne production rate for "silicates" given in Table 3 is the value estimated from Leya et al. (2000) for a Mg:Al:Si ratio, as shown in Fig. 3. If the silicate fraction is assumed to be extraterrestrial and to contain the large part of the cosmogenic Ne, the <sup>21</sup>Ne exposure age is about 4300 years for GR a and 15,700 years for GR b. On the other hand, if the silicates are assumed to be terrestrial, the <sup>21</sup>Ne<sub>cos</sub> concentrations of the Fe<sub>2</sub>O<sub>3</sub> fraction, and thus the <sup>21</sup>Ne exposure ages, become almost identical for both GR aliquots at about 55,000 years.

Nominal <sup>3</sup>He ages are thus considerably lower than the <sup>21</sup>Ne ages. This suggests loss of cosmogenic He and perhaps also Ne from the GR sample. The noble-gas exposure ages derived for this sample are thus lower limits only, both because of a likely substantial terrestrial contamination and noble-gas loss. If cosmogenic noble gases were lost from grain volumes, it is even more likely that the Greenland sample also lost surface-sited solar noble gases. Noble-gas loss may thus well explain an absence of solar gases in this sample. Loss of solar noble gases probably also occurred for the SPWW samples, as shown by their <sup>4</sup>He/<sup>20</sup>Ne ratios (67 and 16, respectively), which are much lower than the <sup>4</sup>He/<sup>20</sup>Ne ratio in the solar wind of ~600. Loss of noble gases from micrometeorites or IDPs has been observed previously (e.g., Nier and Schlutter 1990, 1993); such losses are expected based on heating models and the loss of other volatiles (Flynn 1989, 1995, 2001; Flynn and Sutton 1992).

#### Some Boundary Conditions for SPWW

We consider two possible explanations for the concentrations of radionuclides and noble gases observed in sample SPWW.

In the first approach, we assume that all grains were small IDPs in space and all the material in the analyzed SPWW samples is extraterrestrial. The solar noble gases may suggest that at least some grains were indeed exposed in space as IDPs. In the second approach, we estimate that at least 42% of the sample material is extraterrestrial, by simply comparing the measured amount of <sup>14</sup>C with an expected upper limit of 30 dpm/kg for a meteoroid with a radius of 10 cm (Wieler et al. 1996). Lower <sup>14</sup>C production rate estimates would give larger extraterrestrial fractions. In this case, the solar noble gases would be from regolith breccias irradiated by solar wind on their parent bodies. We adopt production rates obtained from new calculations using MCNPX computer code calculations, as described by Kim and Reedy (2004). The calculations for Fe<sub>2</sub>O<sub>3</sub> and Fe<sub>3</sub>O<sub>4</sub> for small particles are given in Table 4. Our adopted values can be compared with model predictions by Albrecht et al. (2000) and Leya et al. (2000).

The ages calculated using these production rates are given in Table 6. The results for the SPWW sample suggest that the assumption that most of the material is cosmic dust can explain the observed values. The estimated exposure ages based on the three longer-lived nuclides are in agreement with Poynting-Robertson (P-R) lifetimes (Liou et al. 1995). We calculated the P-R lifetimes using the simplified equation for a circular orbit derived by Robertson (see McCracken and Alexander 1968):

$$t_{PR} = 7X10^6 \rho s(a^2 - 1)$$

where  $\rho$  is the density of the particle (g/cm<sup>3</sup>), s is the radius of the particle in  $\mu$ , and *a* is the distance in astronomical units. This results in the following lifetimes for particles of density ~3.0 to go from 3 AU to 1 AU: 25  $\mu$  = 46,700 yr; 50  $\mu$  = 93,300 yr; and 100  $\mu$  = 187,000 yr. These lifetimes can be compared favorably to the exposure times estimated in Table 6. Admittedly, the results for different nuclides are not all in good agreement, but this calculation is made to demonstrate the feasibility of this explanation. As noted above, estimates for sample GR based on cosmogenic noble gases are mostly lower than both SPWW radionuclide exposure ages and P-R lifetimes, which can be explained by noble-gas losses and sample dilution with terrestrial material.

# **Spalling From Large Objects?**

Finally, we consider the assumption that the material could be spalled off larger objects (e.g., Lal and Jull 2002). In this case, the production rates of <sup>10</sup>Be, <sup>26</sup>Al, and <sup>36</sup>Cl could be higher than those given in Table 3 (Leya et al. 2000; Wieler et al. 1996). For example, Nishiizumi et al. (2007) have Table 6. Estimated exposure times in space for cosmic dust from the SPWW, based on different nuclides and assumptions.

*	Pmodel	<sup>10</sup> Be	<sup>26</sup> Al	<sup>36</sup> Cl
SPWW samples				
Degree of undersaturation				
100% cosmic dust	А	0.089	0.08	0.17
	L	0.12	0.14	0.18
	М	0.12	0.13	0.18
42% cosmic dust	А	0.21	0.20	0.41
	L	0.29	0.33	0.43
	М	0.29	0.32	0.43
100%:				
Effective years of irradiation in space	(A)	174 ka	70 ka	72 ka
	(L)	238 ka	138 ka	77 ka
	(M)	238 ka	127 ka	76 ka
42%:				
Effective years of irradiation in space	(A)	440 ka	204 ka	201 ka
	(L)	639 ka	364 ka	217 ka
	(M)	639 ka	351 ka	217 ka
GR samples				
Degree of undersaturation				
100% cosmic dust	А	0.009	0.15	
	L	0.012	0.26	
	М	0.013	0.245	
Effective years of irradiation in space	(A)	19 ka	161 ka	
•	(L)	25 ka	309 ka	
	(M)	28 ka	288 ka	
A = A  lbracht at al. 2000; $I = I$ av	- 2000. M -	MONDY		

A = Albrecht et al. 2000; L = Leya 2000; M = MCNPX.

reported on <sup>10</sup>Be analyses of much larger individual silicaterich micrometeorites (~10 µg) from Tokkuti Point, Antarctica. These samples gave values for <sup>10</sup>Be ranging from ~6 to ~20 dpm/kg, indicating that the  $^{10}$ Be signal can be close to the saturated activity in a larger object and that spalled material can also be present in this size range.

It is not possible, in our opinion, to reconcile the observed values shown in our paper with spalled material, unless the samples are very diluted with terrestrial material to a much greater extent than suggested in this paper. However, it is clear that future work should focus on the analysis of single grains or groups of grains from the SPWW.

#### CONCLUSIONS

This preliminary study gives us confidence that we can apply more precise separations and selection of different types of particles (Taylor et al. 2007) to future samples of cosmic dust recovered from locations such as Greenland and Antarctic ice.

With the data at hand, we can make the following observations:

- Significant quantities of cosmic dust occur in the magnetic phases of SPWW and GR samples, as shown by the extraterrestrial cosmogenic nuclide component. The extraterrestrial component dominates the SPWW and must be the explanation for most of the cosmogenic nuclide signal observed in GR.
- 2. The Fe-Ni content and the radionuclide content of the SPWW sample, when taken together, strongly indicate an extraterrestrial component.
- 3. GR samples contain a large amount of terrestrial <sup>14</sup>C contamination, but also contain cosmogenic noble gases and radionuclides of extraterrestrial origin. It is unclear whether solar noble gases have been preserved in this sample.
- 4. The noble gas results suggest the following:

(i) Magnetic phases partly contain very small amounts of solar He and Ne, indicating that some fraction of the grains has probably been irradiated as small particles in space.

(ii) The particles have lost solar as well as cosmogenic He and Ne, probably not exclusively during atmospheric entry. Evidence for heating (e.g., Taylor et al. 2005; Flynn 1989, 1995, 2001) might also support this hypothesis.

Since our radionuclide results appear consistent with P-R lifetimes, we favor the conclusion that the data are consistent with the magnetic particles having been released in space from the break-up of meter-sized or larger meteoroids in space  $\sim 2-4 \times 10^5$  years ago and intercepted by the Earth. It appears that the history of these magnetic particles analyzed by us are analogues of the IDPs that are sampled in the stratosphere, except that the magnetite grains may have had longer exposure in space.

Acknowledgments–We are grateful to Dr. Michel Maurette for providing the Greenland sample. Thanks are also due to the AMS lab staff at various labs for technical assistance and to the reviewers of the manuscript, and in particular to Dr. C. Olinger for his helpful review and Prof. M. Caffee for his editorial reviews. This work was supported in part by the Leverhulme Trust, the UK Natural Environment Research Council, NASA grant NNG06GC23G, the U.S. National Science Foundation grant EAR0448461, and the Swiss National Science Foundation.

Editorial Handling-Dr. Marc Caffee

#### REFERENCES

Albrecht A., Schnabel C., Vogt S., Xue S., Herzog G. F., Begemann F., Weber H., Middleton R., Fink D., and Klein J. 2000. Light noble gases and cosmogenic radionuclides in Estherville, Budulan, and other mesosiderites: Implications for exposure histories and production rates. *Meteoritics & Planetary Science* 35:975–986.

- Alexander C. M. O'D., Taylor S., Delaney J. S., Ma P., and Herzog G. F. 2002. Mass-dependent fractionation of Mg, Si, and Fe isotopes in five stony cosmic spherules. *Geochimica et Cosmochimica Acta* 66:173–183.
- Bradley J. P. and Brownlee D. E. 1982. Analytical SEM/TEM studies of individual grains of disaggregated CP interplanetary dust (abstract). 13th Lunar and Planetary Science Conference. pp. 65– 66.
- Brook E. J., Kurz M. D., Curtice J., and Cowburn S. 2000. Accretion of interplanetary dust in polar ice. *Geophysics Research Letters* 27:3145–3148.
- Brownlee D. E. 1985. Cosmic dust—Collection and research. *Annual Review of Earth and Planetary Sciences* 13:147–172.
- Brownlee D. E. 2001. The origin and properties of dust impacting the Earth. In Accretion of extraterrestrial matter throughout Earth's history, edited by Peucker-Ehrenbrink B. and Schmitz B. New York: Kluwer Academic Press. pp. 1–12.
- Burns J. A., Lamy P. L., and Soter S. 1979. Radiation forces on small particles in the solar system. *Icarus* 40:1–48.
- Farley K. A. 1995. Cenozoic variations in the flux of interplanetary dust recorded by <sup>3</sup>He in a deep-sea sediment. *Nature* 376:153– 156.
- Farley K. A. and Patterson D. B. 1995. A 100 ka periodicity in the flux of extraterrestrial <sup>3</sup>He to the sea floor. *Nature* 378:600– 603.
- Farley K. A., Love S. G., and Patterson D. B. 1997. Atmospheric entry heating and helium retentivity of interplanetary dust particles. *Geochimica et Cosmochimica Acta* 61:2309–2316.
- Flynn G. J. 1989. Atmospheric entry heating: A criterion to distinguish between asteroidal and cometary sources of interplanetary dust. *Icarus* 77:287–310.
- Flynn G. J. 1995. Thermal gradients in IDPs: Effects of endothermic phase transition (abstract). 26th Lunar and Planetary Science Conference. pp. 405–406.
- Flynn G. J. 2001. Atmospheric entry heating of interplanetary dust. In *Accretion of extraterrestrial matter throughout Earth's history*, edited by Peucker-Ehrenbrink B. and Schmitz B. New York: Kluwer Academic Press. pp. 107–127.
- Flynn G. J. and Sutton S. R. 1992. Trace elements in chondritic stratospheric particles: Zinc depletions as a possible indicator of atmospheric entry. Proceedings, 22nd Lunar and Planetary Science Conference. pp. 171–184.
- Grimberg A., Baur H., Bochsler P., Buhler F., Burnett D. S., Hays C. C., Heber V. S., Jurewicz A. J. G., and Wieler R. 2006. Solar wind neon from Genesis: Implications for the lunar noble gas record. *Science* 314:1133–1135.
- Hudson B., Flynn G. J., Fraundorf P., Hohenberg C. M., and Shirek J. 1981. Noble gases in stratospheric dust particles: Confirmation of extraterrestrial origin. *Science* 211:383–386.
- Jull A. J. T., Courtney C., Jeffrey D. A., and Beck J. W. 1998. Isotopic evidence for a terrestrial source of organic compounds found in Martian meteorites ALH 84001 and EETA79001. *Science* 279: 366–369.
- Jull A. J. T. and Burr G. S. 2006. Accelerator mass spectrometry: Is the future bigger or smaller? *Earth and Planetary Science Letters* 243:305–325.
- Kehm K., Flynn G. J., Sutton S. R., and Hohenberg C. M. 1998. Combined noble-gas and trace-element measurements in single IDPs from the L2036 collector (abstract #1970). 29th Lunar and Planetary Science Conference. CD-ROM.
- Kehm K., Flynn G. J., Sutton S. R., and Hohenberg C. M. 2002. Combined noble-gas and trace-element measurements on individual stratospheric interplanetary dust particles. *Meteoritics* & *Planetary Science* 37:1323–1335.
- Kehm K., Flynn G. J., and Hohenberg C. M. 2006. Noble-gas

exposure ages of individual interplanetary dust particles. *Meteoritics & Planetary Science* 41:1199–1217.

- Kim K. J. and Reedy R. C. 2004. Production rates of cosmogenic nuclides in the Knyahinya L chondrite (abstract #1359). 35th Lunar and Planetary Science Conference. CD-ROM.
- Lal D. and Jull A. J. T. 2002. Atmospheric cosmic dust fluxes in the size range 10<sup>-4</sup> to 10 cm. *The Astrophysical Journal* 579:1090– 1097.
- Lal D., Jull A. J. T., and Donahue D. J. 1995. Evidence for a noncosmogenic implanted <sup>14</sup>C component in lunar samples. *Earth* and Planetary Science Letters 136:693–702.
- Leya I., Lange H. J., Neumann S., Wieler R., and Michel R. 2000. The production of cosmogenic nuclides in stony meteoroids by galactic cosmic-ray particles. *Meteoritics & Planetary Science* 35:259–286.
- Liou J.-C., Zook H. A., and Jackson A. A. 1995. Radiation pressure, Poynting-Robertson drag, and solar wind drag in the restricted three-body problem. *Icarus* 116:186–201.
- Love S. G. and Brownlee D. E. 1993. A direct measurement of the terrestrial mass accretion rate of cosmic dust. *Science* 262:550– 553.
- Love S. G. and Brownlee D. E. 1994. Peak atmospheric entry temperatures of micrometeorites. *Meteoritics* 29:69–71.
- Maden C., Anastasi P. A. F., Dougans D., Freeman S. P. H. T., Kitchen R., Klody G., Schnabel C., Sundquist M., Vanner K., and Xu S. 2006. SUERC AMS ion detection. *Nuclear Instruments* and Methods in Physics Research B 259:131–139.
- Marcantonio F., Kumar N., Stute M., Anderson R. F., and Seidl M. A. 1995. Comparative study of accumulation rates derived by Th and He isotope analyses of marine sediments. *Earth and Planetary Science Letters* 133:549–555.
- Maurette M., Jéhanno C., Robin E., and Hammer C. 1987. Characteristics and mass distribution of extraterrestrial dust from the Greenland ice cap. *Nature* 328:699–702.
- Maurette M., Hammer C., and Pourchet M. 1990. Multidisciplinary investigations of new collections of Greenland and Antarctic micrometeorites. In *From mantle to meteorites: A garland of perspectives*, edited by Gopalan K., Gaur V. K., Somayajulu B. L. K., and MacDougall J. D. Bangalore: Indian Academy of Sciences. pp. 87–126.
- Maurette M., Olinger C., Michel-Levy M. C., Kurat G., Pourchet M., Brandstätter F., and Bourot-Denise M. 1991. A collection of diverse micrometeorites recovered from 100 tonnes of Antarctic blue ice. *Nature* 351:44–47.
- McCracken C. W. and Alexander W. M. 1968. Interplanetary dust particles. In *Introduction to space science*, edited by Hess W. N. and Mead G. D. New York: Gordon and Breach. pp. 447–499.
- Merrihue C. 1964. Rare gas evidence for cosmic dust in modern Pacific red clay. Annals of the New York Academy of Sciences 119:351–367.
- Nier A. O. and Schlutter D. J. 1990. Helium and neon isotopes in stratospheric particles. *Meteoritics* 25:263–267.
- Nier A. O. and Schlutter D. J. 1993. The thermal history of interplanetary dust particles collected in the Earth's stratosphere. *Meteoritics* 28:675–681.
- Nishiizumi K., Arnold J. R., Fink D., Klein J., Middleton R., Brownlee D. E., and Maurette M. 1991. Exposure history of individual cosmic dust particles. *Earth and Planetary Science Letters* 104:315–324.
- Nishiizumi K., Arnold J. R., Brownlee D. E., Caffee M. W., Finkel R. C., and Harvey R. P. 1995. <sup>10</sup>Be and <sup>26</sup>Al in individual cosmic spherules from Antarctica. *Meteoritics* 30:728–732.
- Nishiizumi K., Nakamura T., Caffee M. W., and Yada T. 2006. Exposure histories of 10 microgram individual Antarctic micrometeorites: Radionuclide measurements and chemical and

morphological analyses (abstract #2129). 38th Lunar and Planetary Science Conference. CD-ROM.

- Olinger C. T., Maurette M., Walker R. M., and Hohenberg C. M. 1990. Neon measurements of individual Greenland sediment particles: Proof of an extraterrestrial origin and comparison with EDX and morphological analyses. *Earth and Planetary Science Letters* 100:77–93.
- Ozima M., Takayanagi M., Zashu S., and Amari S. 1984. High <sup>3</sup>He/ <sup>4</sup>He ratios in ocean sediments. *Nature* 311:449–451.
- Pepin R. O., Palma R. L., and Shlutter D. J. 1999. Evidence for a dominant component of solar energetic particle (SEP) helium and neon in a suite of interplanetary dust particles (abstract #1864). 30th Lunar and Planetary Science Conference. CD-ROM.
- Pepin R. O., Palma R. L., and Schlutter D. J. 2000. Noble gases in interplanetary dust particles, I: The excess <sup>3</sup>H problem and estimates of the relative fluxes of solar wind and solar energetic particles in interplanetary space. *Meteoritics & Planetary Science* 35:495–504.
- Pepin R. O., Palma R. L., and Schlutter D. J. 2001. Noble gases in interplanetary dust particles II: Excess <sup>3</sup>H in cluster particles and modeling constraints on interplanetary dust particle exposures to cosmic radiation. *Meteoritics & Planetary Science* 36:1515– 1534.
- Rajan R. S., Brownlee D. E., Tomandl D., Hodge P. W., Farrar H., and Birtten R. A. 1977. Detection of <sup>4</sup>He in stratospheric particles gives evidence of extraterrestrial origin. *Nature* 267:133–134.
- Raisbeck G. M., Yiou F., Klein J., Middleton R., Yamakoshi Y., and Brownlee D. E. 1983. <sup>26</sup>Al and <sup>10</sup>Be in deep-sea stony spherules: Evidence for small parent bodies (abstract). 14th Lunar and Planetary Science Conference. pp. 622–623.
- Raisbeck G. M., Yiou F., and Brownlee D. E. 1985. Unusually high concentration of <sup>10</sup>Be in a cosmic spherule: Possible evidence for irradiation outside the planetary solar system (abstract). *Meteoritics* 20:734–735.
- Raisbeck G. M. and Yiou F. 1989. Cosmic-ray exposure ages of cosmic spherules (abstract). *Meteoritics* 24:318.
- Taylor S., Lever J. H., and Harvey R. P. 1998. Accretion rate of cosmic spherules measured at the South Pole. *Nature* 392:899– 903.
- Taylor S., Lever J. H., and Harvey R. P. 2000. Numbers, types, and compositions of an unbiased collection of cosmic spherules. *Meteoritics & Planetary Science* 35:651–666.
- Taylor S., Alexander C. M. O'D., Delaney J., Ma P., Herzog G. F., and Engrand C. 2005. Isotopic fractionation of iron, potassium, and oxygen in stony cosmic spherules: Implications for heating histories and sources. *Geochimica et Cosmochimica Acta* 69: 2647–2662.
- Taylor S., Herzog G. F., and Delaney J. S. 2007. Crumbs from the crust of Vesta: Achondritic cosmic spherules from the South Pole Water Well. *Meteoritics & Planetary Science* 42:223–233.
- Vogel N., Wieler R., Bischoff A., and Baur H. 2003. Microdistribution of primordial Ne and Ar in fine-grained rims, matrices, and dark inclusions of unequilibrated chondrites— Clues on nebular processes. *Meteoritics & Planetary Science* 38: 1399–1418.
- Wieler R. 2002. Noble gases in the solar system. Reviews of Mineralogy and Geochemistry 47:21–70.
- Wieler R., Baur H., and Signer P. 1986. Noble gases from solar energetic particles revealed by closed system stepwise etching of lunar soil minerals. *Geochimica et Cosmochimica Acta* 50:1997– 2017.
- Wieler R., Graf T., Signer P., Vogt S., Herzog G. F., Tuniz C., Fink D., Fifield L. K., Klein J., Middleton R., Jull A. J. T., Pellas P., Masarik J., and Dreibus G. 1996. Exposure history of the Torino meteorite. *Meteoritics & Planetary Science* 31:265–272.

Winckler G., Anderson R. F., Stute M., and Schlosser P. 2004. Does interplanetary dust control 100 kyr glacial cycles? *Quaternary Science Reviews* 23:1873–1878.

Xue S., Shih C.-Y., Wisemann H., Brownlee D. E., Hall G. S., Herzog

G. F., and Nyquist L. E. 1997. Isotopic and elemental composition of chromium, iron, and nickel in type I deep-sea spheres (abstract). 28th Lunar and Planetary Science Conference. pp. 595–596.