## RADIONUCLIDE STUDIES OF STONY METEORITES FROM HOT DESERTS

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**ABSTRACT.** We summarize the use of radiocarbon produced by spallation in meteorites in space to determine their terrestrial age or residence time. This "age" gives us important information as it can be compared to the rates of weathering and infall of meteorites. The processes that affect the collection of meteorites in a given area can be related to the rates of infall of new meteorites, and the rate of removal by chemical weathering and physical erosion.

## INTRODUCTION

Meteorites can survive for long periods of time in arid environments, which makes the determination of their terrestrial age important to understanding compositional changes due to oxidation and weathering. This phenomenon was first recognized in the very long times that meteorites could survive in the cold, arid continent of Antarctica where residence times, or terrestrial ages, can be well over 50,000 yr (Jull et al. 2000; Jull 2006). In Antarctica, meteorites have been collected for many years as part of annual field expeditions and many measurements of  ${}^{14}C$  and  ${}^{14}C/{}^{10}Be$  (Jull et al. 1998a; Jull 2001, 2006; Welten et al. 2006) and other radionuclides including <sup>36</sup>Cl and <sup>41</sup>Ca (Welten et al. 2001, 2006, 2007; Folco et al. 2006; Nishiizumi and Caffee 2006; Nishiizumi et al. 2011) have been made. Most meteorites recovered are stony-iron meteorites, called chondrites, and these can be classified as H, L, and LL chondrites based on their iron metal content. Some chondrites rich in carbonaceous material are labeled as C chondrites. Terrestrial-age determinations on meteorites from hot desert environments (as opposed to the cold desert of Antarctica) can give us unique information about the terrestrial residence ages of meteorites (Jull et al. 1990; Bland et al. 1996, 1998, 2000; Jull 2001, 2006; Welten et al. 2006, 2007). Results from these studies suggest possible changes in meteorite infall rate and allow us to quantify the weathering of meteorites as a function of their terrestrial age (e.g. Gattacceca et al. 2011; Hezel et al. 2011). Other studies have focused on understanding storage effects, such as the adsorption of elements from soil (e.g. Al-Kathiri et al. 2005) and other geochemical changes that occur in the meteorite (Al-Kathiri et al. 2005; Zurfluh et al. 2011).

The idea of terrestrial-age measurements has a long history (e.g. Arnold et al. 1961; Suess and Wänke 1962), although it was not until the advent of accelerator mass spectrometry (AMS) that meteorite samples of a reasonable size could be processed. At the NSF-Arizona AMS laboratory, we make measurements of  ${}^{14}C$  and  ${}^{14}C/{}^{10}Be$ . The ratio of  ${}^{14}C/{}^{10}Be$  can be used to get better precision on terrestrial ages. For "hot" deserts, a dependence of the degree of oxidation on terrestrial age has been demonstrated for a number of desert locations from many parts of the world, yet others do not appear to show this dependence (Gattacceca et al. 2011). In this paper, we summarize our current understanding in this field and highlight some new developments.

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### **COSMIC-RAY EFFECTS**

Meteorites are exposed to relatively high levels of cosmic radiation as they travel through space (Leya and Masarik 2009) and this leads to enhanced cosmogenic isotope production. For example, the saturated radiocarbon content of a meteorite in space of  $2.2 \times 10^8$  atoms/g (equivalent to a production rate of 50.7 atoms/min/kg, or 26,650 atoms/g/yr) is about 1600 times the saturated <sup>14</sup>C content of a surface rock at sea level, which is about  $1.3 \times 10^5$  atoms/g or a production rate of ~16 atoms/year/g. At an elevation of 2000 m, the saturated level of <sup>14</sup>C would be about  $10^6$  atoms/g (Lifton et al. 2001), still 220 times less than produced in the meteorite in space. When a meteorite falls to Earth, it becomes shielded from most of this radiation, and the excess radionuclides produced in space decay with their characteristic half-lives.

Cosmic rays come from 2 different sources: galactic and solar cosmic rays (GCR and SCR). Both of these are composed primarily of protons and alpha particles. GCR originate from beyond the solar system and have a flux of less than 2 particles/cm<sup>2</sup>/s, with GeV energies. SCR are energetic particles emitted by the Sun, with energies of tens to hundreds of MeV. They have a variable flux, typically around 100 particles/cm<sup>2</sup>/s (E>10 MeV), and a distinctly solar composition. Studies of depth profiles in the near surface (<1-2 cm) have been successfully used to derive an SCR flux in cases where production rates are well known. Constraints on the temporal variability of the SCR flux have also been determined by comparison of radionuclides with different half-lives (e.g. Jull et al. 1998a,b; Eugster et al. 2006). When cosmic-ray particles interact with matter, they produce a cascade of secondary particles that cause nuclear reactions and lead to the production of radioactive nuclides in the target (Leva et al. 2000; Reedy 2004; Leva and Masarik 2009). Meteorites are a rich natural archive of radionuclides produced by these processes, and many cosmogenic components in them can be measured with AMS. Radionuclides such as <sup>14</sup>C and <sup>10</sup>Be are primarily produced in rocks by highenergy spallation reactions on oxygen and to a much lesser extent on Mg and Si. Since the limit of the <sup>14</sup>C technique extends to about the last 50,000 yr, and <sup>10</sup>Be is about 5 million years, they are ideal for the study of terrestrial ages in desert environments (Jull et al. 1998a; Jull 2006).

For most meteorites, we are interested in the radionuclides produced by secondary neutrons and protons from GCR reactions, which have a characteristic depth dependence (e.g. Leya et al. 2000). In contrast to the case of a very large object such as the Moon or relatively large meteorites (radius >3 m), the GCR production profile in most meteorites actually increases to a depth of ~150 g/cm<sup>2</sup>. This is due to irradiation of the sample from all sides ( $4\pi$  irradiation), as opposed to  $2\pi$  irradiation, typical of larger objects. The time for a target to become saturated with a particular cosmogenic radioisotope is a function of the half-life of the radionuclide. A maximum saturation level for <sup>14</sup>C is achieved after ~5 half-lives, or 25,000 yr of continuous exposure. For <sup>10</sup>Be, saturation takes about 7–8 Ma. For <sup>26</sup>Al, saturation occurs after about 3–4 Ma and for <sup>129</sup>I, after ~80 Ma. The production of <sup>14</sup>C (or any other spallation-produced isotope) depends on the particle flux at the depth of the sample in the body, the energy distribution of nuclear particles at this depth, the excitation function for <sup>14</sup>C production from oxygen (and other elements) as a function of energy, and the chemical composition of the sample.

# METHODS

Meteorite samples are prepared for both <sup>14</sup>C and <sup>10</sup>Be measurements at the University of Arizona laboratory. For <sup>14</sup>C, the sample is first crushed, treated with 85%  $H_3PO_4$  to remove any carbonate weathering products, and then washed with distilled water. A weighed amount of the dried powder is mixed with Fe chips, which act as a combustion accelerator, and this mixture is placed in a ceramic crucible, which is preheated to 500 °C for 1 hr. After that time, the crucible is loaded into a

radio-frequency (RF) induction furnace system (Jull et al. 1993, 1998a, 2010) and the sample is heated in a flow of oxygen to melting for ~2 min. The oxygen gas flow is collected into a liquid nitrogen trap. After the sample has cooled, with oxygen continuing to be allowed to flow into the trap, the trap is opened to vacuum to remove oxygen gas, as it will be removed below its vapor pressure. The remaining material condensed in the trap is mainly  $CO_2$  and  $H_2O$ . This mixture is then passed over a dry ice (-78 °C) trap and into a gas sample collection vessel using cryogenic trapping. The gas is allowed to warm up in a known volume and the yield of the C as  $CO_2$  can be calculated. The gas is then converted to graphite over Fe and pressed into a target holder for measurement by AMS. After measurement, the number of <sup>14</sup>C atoms is calculated as summarized by Jull (2006).

For <sup>10</sup>Be measurements, we dissolved the sample in HF-HNO<sub>3</sub> and add 0.3 mg of Be as a carrier. Be is separated from the solution using an acetyl acetone complex into an organic solvent. The  $Be(OH)_2$  is precipitated and this material oxidized to BeO in a quartz tube using a Bunsen burner flame. Details of this method are given in McHargue et al. (1995).

### SATURATED ACTIVITY

In order to be able to compare results to production rates, it is conventional in these studies to quote the specific activity of the sample (in decays per minute per kilogram [dpm/kg]), partly for historical reasons, instead of <sup>14</sup>C atoms per gram of meteorite. Obviously, the two are easily related by the radioactive decay equation, with correction of the decay rate to units of dpm/kg:

 $dN / dt = -\lambda N$ 

where N is the number of atoms, t is time (here in decays per minute), and  $\lambda$  is the decay constant for <sup>14</sup>C of  $1.21 \times 10^{-4}$  yr<sup>-1</sup>.

In order to normalize the results, we need to compare the <sup>14</sup>C in the meteorite sample to a recently fallen meteorite of known age. We have done this using the Bruderheim meteorite, a known fall, as a standard. Results of 35 measurements of repeat samples of Bruderheim gave an average value of 51 dpm/kg with a standard deviation of  $\pm 6$  dpm/kg and an error in the mean of  $\pm 1$  dpm/kg. This allows us to establish an average value of the saturated level of <sup>14</sup>C in this meteorite in space. We then relate this to other meteorites by normalization to the oxygen content (Jull et al. 2000). Because we know that the <sup>14</sup>C production has a depth dependence in a meteorite, we assign a relatively conservative error of  $\pm 15\%$  to this possible depth error. In order to obtain better precision, we can normalize the <sup>14</sup>C to the <sup>10</sup>Be level if the meteorite is saturated in <sup>10</sup>Be. Other studies used Ne isotopes as a shielding indicator (Schultz et al. 2005). We can demonstrate this in the case of the Gold Basin meteorite strewn field (Kring et al. 2001). Here, a large number of meteorites from the same fall are spread over a wide area and represent different depths in the same large object (Figure 1). This method is only possible in some cases; for achondrites and some L-chondrites, <sup>10</sup>Be may not be saturated. In other cases, a different shielding parameter may be used (e.g. Schultz et al. 2005).

We compare the result from a specific sample to the expected saturated activity from Bruderheim, normalized for oxygen content, as almost all (>95%) of the production of <sup>14</sup>C by spallation is on oxygen (Jull et al. 1998a). In a meteorite that has been exposed in space long enough to be saturated, the production rate will equal the decay rate.

The <sup>14</sup>C terrestrial age can then be calculated as follows:

<sup>14</sup>C terrestrial age = 
$$\frac{-1}{\lambda_{14}} \ln\left(\frac{N_{meteorite}}{N_{saturated}}\right)$$

If we can also determine the <sup>10</sup>Be content for a shielding correction, and we can assume the sample is saturated, we can also determine the terrestrial age from the equation:

$$\frac{{}^{14}C}{{}^{10}Be} \text{ terrestrial } age = \frac{-1}{\lambda_{14} - \lambda_{10}} \ln \left( \frac{\frac{{}^{14}C}{{}^{10}Be} \text{ meteorite}}{\frac{{}^{14}C}{{}^{10}Be} \text{ saturated}} \right)$$

where  ${}^{14}C/{}^{10}Be$  (meteorite) is the measured atomic ratio of  ${}^{14}C$  to  ${}^{10}Be$  in the meteorite, and  ${}^{14}C/{}^{10}Be$  (saturated) is the expected value at saturation, usually taken as 2.5 to 2.65 (Welten et al. 2003, 2004).

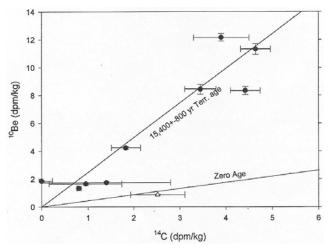


Figure 1 <sup>14</sup>C and <sup>10</sup>Be results for the Gold Basin meteorites

#### **Other Recent Falls**

We also have surveyed a number of other recent falls for their  $^{14}$ C content. This was in part to try to evaluate the variability of  $^{14}$ C among recently fallen meteorites. The measurements on these meteorites are shown in Table 1. These results show that the variability between samples is considerable, which is in part due to the uncertainty of the depth of the sample in the meteoroid, which can be compensated by using a depth indicator, such as Ne isotopes (Schultz et al. 2005) or  $^{10}$ Be if the meteorite is saturated in this nuclide (e.g. Jull et al. 1998a, 2001, 2010).

Table 1 Results of	C 011 501	lie recent rans.		
Meteorite	Class	Year of fall	$^{14}C (10^8 \text{ atom/g})$	<sup>14</sup> C (dpm/kg)
Allende	CV3	1978	2.51	$57.7\pm0.6$
Bjurböle	L/LL4	1899	2.08	$47.9\pm0.4$
Knyahinya	LL5	1866	2.40	$55.3\pm0.9$
Murchison	CM2	1969	3.22	$74.3\pm0.9$
Nuevo Mercurio	H5	1978	1.56	$35.9 \pm 1.1$
Saratov	L4	1918	1.99	$45.8\pm0.8$
Tamdakht	H5	2008	1.99	$45.8\pm0.9$
Bruderheim	L6	1962	2.22	$51 \pm 1$

Table 1 Results of <sup>14</sup>C on some recent falls.

# **RESULTS AND DISCUSSION**

# **New Results**

Although a large number of results have been published as discussed in the following sections, here we highlight several results on interesting meteorites that have not previously been published. One interesting meteorite that deserves special attention is the Carancas (Peru) fall.

#### Carancas, Peru Meteorite

The Carancas meteorite was a spectacular fall that occurred on 15 September 2007, which created a 13-m-diameter crater (Borovika and Spurný 2008; Tancredi et al. 2008). Some meteorite fragments were recovered from the crater area. The meteorite is classified as an H4-5 chondrite. Although it is expected that these fragments represent the impacting meteoroid, we undertook <sup>14</sup>C and <sup>10</sup>Be measurements to determine the levels present. The results are shown in Table 2.

Table 2 Results of <sup>14</sup>C and <sup>10</sup>Be measurements on the Carancas meteorite.

<sup>14</sup> C (dpm/kg)	<sup>10</sup> Be	<sup>14</sup> C/ <sup>10</sup> Be	Age
$53.8 \pm 1.5$	$17.3\pm0.2$	3.08	Recent fall

It is interesting to note that the value observed in Carancas is in agreement with the saturation values measured in Bruderheim. However, since Bruderheim is an L6 chondrite and contains more oxygen, we would expect there to be less <sup>14</sup>C in Carancas based on our simple oxygen-normalization model. This only serves to emphasize the need to correct for the size of the meteorite using <sup>10</sup>Be. In this case, we calculate that the impactor must have had a radius of at least 30–40 cm, which is consistent with observations on the size of the impact crater (Borovika and Spurný 2008).

### Meteorites from Atacama, Chile

In addition to the meteorites discussed in the published results, we compared the terrestrial ages of 5 meteorites collected near Pampa, Antofagasta, Chile. These results are shown in Table 3, which summarizes the different <sup>14</sup>C contents and estimated terrestrial ages, but we also compare the results on the weathering products of the meteorites. In the case of weathering products, <sup>14</sup>C from the atmosphere is incorporated into secondary weathering carbonates, and is not produced by spallation. The results shown for the weathering carbonates are uncalibrated conventional radiocarbon years BP (<sup>14</sup>C yr BP).

Table 3 Meteorites from Pampa, Chile.					
N		In situ <sup>14</sup> C	Terrestrial	Carbonate <sup>14</sup> C	
Name	Class	(dpm/kg)	<sup>14</sup> C age (ka)	age (yr)	
Pampa A	L6	$2.46\pm0.20$	$25.1 \pm 1.5$	post-bomb	
Pampa B	L4/5	$3.9 \pm 1.0$	$21.3\pm2.7$	$9070\pm420$	
Pampa C	L4	$9.7 \pm 1.9$	$13.8\pm2.1$	$5590 \pm 120$	
Pampa D	L5	$9.3\pm1.6$	$14.2\pm1.9$	—	
Pampa G	L5	$9.1 \pm 1.1$	$14.3\pm1.6$	>15,300	

There are 2 interesting results from these studies. First, the carbonate weathering products are usually more recent in <sup>14</sup>C age than the meteorite. This suggests the source of the <sup>14</sup>C in the weathering products is the atmosphere, not the surrounding old soil carbonate. However, Pampa G suggests that

this can also be a source. Al-Kathiri et al. (2005) have argued that local soil chemistry is very important in the uptake of soil ions into meteorites lying on the ground. Second, the distribution of  $^{14}$ C ages suggests that the meteorite collection does not reflect the expected exponential behavior of the terrestrial ages, despite the low number of samples. This is similar to the conclusion of Gattacceca et al. (2011) for Atacama Desert meteorites.

#### Saudi and Omani Meteorites

Franchi et al. (1995) were some of the first to note the importance of the Arabian Peninsula as a source of meteorites. Indeed, this paper is somewhat prescient given the amount of work done since that time (e.g. Al-Kathiri et al. 2005; Jull et al. 2008; Gnos et al. 2009; Hezel et al. 2011; Zurfluh et al. 2011). We have reinvestigated the <sup>14</sup>C and in some cases <sup>10</sup>Be for a number of meteorites from this region, most of which were collected in the 1930s to 1950s (Franchi et al. 1995).

For example, the meteorite Ghubara found in Oman has been studied on several occasions, so we can compare results. The other results were obtained in the late 1990s from museum collections, but have never been formally published. Intriguingly, in comparison to more recent collections, these meteorites show a preponderance of young falls. This may reflect a collection bias in the original meteorite collection, or a bias towards larger sample sizes. Hofmann et al. (2009) collected several samples in Saudi Arabia in 2008 as part of a Swiss-Saudi collaboration, but no terrestrial ages have yet been obtained.

	Year		<sup>14</sup> C	$^{14}C$	<sup>10</sup> Be	<sup>14</sup> C/ <sup>10</sup> Be age
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Meteorite	collected	Class	$(10^8 \text{ atoms/g})$	(dpm/kg)	(dpm/kg)	(kyr)
Ghubara BM1954 <sup>a</sup>	1954	L5	1.56	$35.9 \pm 1.07$		
Ghubara <sup>b</sup>		L5	1.48	$34.6 \pm 0.5$	$20.7\pm1.2$	$3.4 \pm 0.6$
Ghubara <sup>b</sup>		L5	2.04	$46.9\pm0.4$	$20.7\pm1.2$	$0.8\pm0.6$
Ghubara BM1954 <sup>c</sup>				$39 \pm 3$		
Ghubara BM1956 <sup>c</sup>				$39 \pm 3$		
Ghubara BM1958 <sup>c</sup>				$38 \pm 3$		
Ad Dhabubah	1932	H5	1.07	$24.7\pm0.3$		$5.2 \pm 1.3$
Al Ghamin	1960	L6	0.41	$9.4 \pm 0.1$		$14.0 \pm 1.3$
Ash Shalfah	1961	L6	0.94	$2.1 \pm 0.2$		$26.1\pm1.6$
As-Suay'dan	1960	L4	1.31	$30.1 \pm 0.2$		$4.4 \pm 1.3$
Bir Hadi	1958	H5	1.31	$30.1 \pm 0.3$		$3.6 \pm 1.3$
Hajmah (a), Oman	1958	Ureilite	0.16	$3.7 \pm 0.7$		$22 \pm 2$
Hajmah (c), Oman	1958	L5-6	0.355	$8.1 \pm 0.3$		$15.1 \pm 1.3$
Jiddat al Harasis, Oman 1957	1957	H4		$0.48\pm0.02$		$37.8 \pm 3.7$
Suwahib (AinSalah)	1932	H6	0.083	$1.9 \pm 0.2$		$26.3 \pm 1.5$
Suwahib (Buwah)	1932	H3	2.03	$46.6\pm0.5$		Recent fall
Tarfa, Oman	1954	L6		$8.2 \pm 0.2$		$15.2 \pm 1.3$
Umm Tina	1932	L6		$0.45\pm0.30$		$39\pm 6$

Table 4 Summary of results on desert meteorites from early collections in Saudi Arabia and Oman.

<sup>a</sup>Run in 2008.

<sup>b</sup>Run in 1995.

°Results of Ferko et al. (2002), who measured  $39 \pm 3$ ,  $39 \pm 3$ , and  $38 \pm 3$  dpm/kg on 3 different samples.

# Summary of Published Results on Meteorites

In the past decade, many new samples have been recovered from Australia, Oman, and sites in the Atacama in Chile. These locations are rich in meteorite finds. An example of a meteorite found in the field by the Swiss-Omani team is shown in Figure 2.

Radionuclide Studies of Stony Meteorites from Hot Deserts



Figure 2 A meteorite found in the field in Oman (courtesy B Hofmann)

We have determined the <sup>14</sup>C terrestrial ages for many samples from these locations. The results indicate that weathering is generally dependent on terrestrial age, as noted earlier (Wlotzka et al. 1995; Bland et al. 1996; Al-Kathiri et al. 2005). In most cases, we see a typical age distribution similar to that shown in Figure 3 for meteorites from Western Australia, where we observe an exponential drop-off of a number of meteorites with increasing terrestrial age.

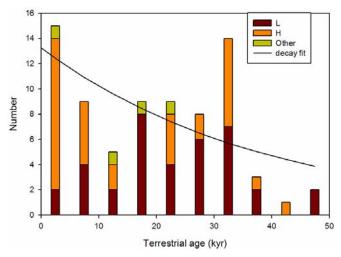


Figure 3 Terrestrial age distribution of meteorites from Western Australia (Jull et al. 2010). Note that meteorites of different classes (L, H, and other) are shown separately, but the general plot is one of an exponential decay curve. Some of the excess number of meteorites at 30–35 kyr is probably due to pairing of meteorites.

The terrestrial age distribution from an Omani site is more complex, showing a bimodal behavior, with a suite of younger dates consistent with infall and older dates >25 kyr, suggesting some removal or sorting process at this location, which is geologically different from all other Oman find sites (fossil dunes). Previously, Al-Kathiri et al. (2005) summarized the terrestrial ages of 53 meteorites from Oman, which showed an approximately exponential distribution of ages, but with a deficiency of ages <10 kyr. Recently, Zurfluh et al. (2011) have extended the Omani study to over 100

meteorites. Independently, Hezel et al. (2011) have reported on the age distribution of meteorites from the nearby United Arab Emirates (UAE). Both the UAE and Omani meteorites show a deficiency of younger terrestrial ages, although this trend is more distinct in the case of Oman, as shown in Figure 4.

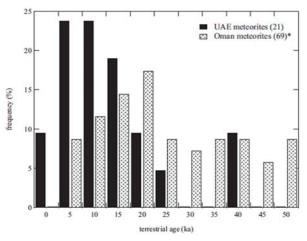


Figure 4 Comparison of terrestrial ages for the United Arab Emirates (UAE) and 69 meteorites from Oman (Al-Kathiri et al. 2005; Hezel et al. 2011). Note the deficiency of meteorites <10 kyr in age.

In contrast, results from the Atacama Desert indicate a wide range of terrestrial ages, and the <sup>14</sup>Cage distribution is essentially flat, with no exponential drop-off as expected from previous work (Figure 3). This suggests a much longer residence time at this location (Gattacceca et al. 2011).

Gattacceca et al. (2011) also plotted the weathering grade (see Figure 5) of meteorites from different collection areas, and noted that the Atacama Desert meteorites showed less weathering in general, even though they had a wide range of terrestrial ages up to >50 kyr. This suggests that there are other processes involved, either some meteorites are removed by some process, perhaps weathering of meteorites in this region takes a longer time, or this region represents some type of erosional lag deposit. A related question is the low degree of weathering of San Juan ordinary chondrites, especially given the otherwise severe weathering conditions of this environment. We presume that a lack of water is the key to the observed reduced weathering rate in this terrain.

# Interpreting Meteorite Cumulative Age Distributions

The differential plots (number of meteorites vs. time) in Figures 3 and 4 indicate deficiencies of meteorites at certain periods of time. These could be related to various processes, such as removal, weathering, or infall-rate changes. If we assume constant infall, then plot the cumulative age distributions, we can estimate the decay constant due to weathering, and relate this to climatic influences. We suggest that local exponentially shaped disruptions of a linear cumulative age distribution are caused by climate changes. Since different climates induce different chemical weathering rates and affect mostly young meteorites (as shown by Bland et al. 1998), only that portion of the age distribution that is close to the time of the climate change is affected.

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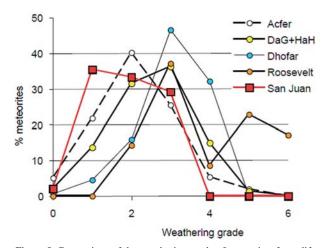


Figure 5 Comparison of the weathering grade of meteorites from different desert environments. Acfer, Algeria; DaG = Dar al Gani, Libya; HaH = Hamadah al Hamra, Libya; Dhofar, Oman; Roosevelt = Roosevelt County, New Mexico, USA; San Juan = San Juan field, Atacama Desert (adapted from Gattacceca et al. 2011).

## CONCLUSIONS

In summary, the terrestrial age of a meteorite is a very useful parameter, since it gives information that can be compared to chemical and physical changes in a meteorite over time due to weathering. These effects are very important, given the number of meteorites now recovered from desert environments. In order to understand the processes involved, we need large numbers of meteorites collected from a specific terrain. We can then begin to understand the processes of weathering and how they are related to local chemistry and availability of water, as well as local erosion processes. Recent studies emphasize the importance of these studies, since weathering may affect the chemical (Al-Kathiri et al. 2005) as well as isotopic composition (Schwenzer et al. 2012) of meteorites from deserts, whether in a hot arid desert or in the cold desert of Antarctica.

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