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Chemical alteration and REE mobilization in meteorites from hot and cold deserts

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Abstract—The effects of terrestrial weathering on REE mobilization are evaluated for a variety of uncommon meteorites found in Antarctica and in hot deserts. The meteorites analyzed include 7 non-cumulate eucrites, 10 shergottites, 3 nakhlites, 2 lunar meteorites, 4 angrites, 10 acapulcoites, 1 winonaite, and 1 brachinite. In-situ concentration measurements of lanthanides and selected other minor and trace elements were made on individual grains by secondary ion mass spectrometry (SIMS). In Antarctic meteorites, oxidation converts Ce^{3+} to Ce^{4+}, which is less soluble than the trivalent REE, resulting in Ce anomalies. The mineral most affected is low-Ca pyroxene. However, not all grains of a given mineral are, and distinct analyses of a single grain can even yield REE patterns with and without Ce anomalies. The effect is most pronounced for Antarctic eucrites in which Ce anomalies are observed not only in individual minerals but also in whole rock samples. Although Ce anomalies are observed in meteorites from hot deserts as well, the most characteristic signs of chemical alteration in this environment are a LREE enrichment with a typical crustal signature, as well as Sr, Ba and U contaminations. These can modify the whole rock REE patterns and disturb the isotope systematics used to date these objects. The LREE contamination is highly heterogeneous, affecting some grains and not others of a given mineral (mainly olivine and low-Ca pyroxene, the two minerals with the lowest REE concentrations). The major conduit for REE movement is through shock-induced cracks and defects, and the highest levels of contamination are found in altered material filling such veins and cracks. Meteorites that experienced low shock levels and those that are highly recrystallized are the least altered. *Copyright © 2003 Elsevier Ltd*

1. INTRODUCTION

Our understanding of the early stages of evolution of the solar system, though still far from complete, has greatly benefited from studies of meteorites conducted since the second half of the last century. Our increase in knowledge can be traced back to the development and use of a variety of increasingly sophisticated experimental techniques to analyze meteorites. It has also been facilitated by the discoveries of many meteorites, particularly those belonging to rare groups, in both cold (i.e., Antarctica) and hot (the most productive so far being the Sahara) deserts on earth.

However, meteorites found in these regions have spent considerable time on earth since their fall, up to ~2 Ma for Antarctic meteorites (Welten et al., 1997) and typically $> 10^4$ yr for hot desert meteorites (Wlotzka et al., 1995). Therefore, it is critical to evaluate the extent to which their initial properties have been modified by their long residence time on our planet. Of course, concerns about the integrity of samples are not unique to extraterrestrial objects. However, the situation is more acute for them because specimens are typically small and, unlike for terrestrial rocks, there is often no opportunity to sample areas that are unambiguously unaltered. In addition, many meteorites show the effects of shock, including fractures and dislocations that greatly facilitate the action of weathering.

In our earlier work on Antarctic meteorites (Floss and Crozaz, 1991), we studied the microdistributions of a number of trace elements including the rare earth elements (REE). Repeatedly, we encountered evidence of chemical alteration, even in seemingly unaffected areas. Indeed, we found that different portions of the same grain often show strikingly different effects. As we moved into the study of hot desert meteorites, we realized that terrestrial alteration effects in these meteorites are both different from and more prevalent than in Antarctic meteorites. Our first observations in the shergottites Dar al Gani (DaG) 476/489 were published by Crozaz and Wadhwa (2001).

In the present paper, we report on the terrestrial alteration we have observed in a wide variety of meteorite groups, and summarize the different effects terrestrial weathering has had on meteorites from Antarctica and hot desert regions. We emphasize that most of our observations are not the result of a concerted effort to systematically study the effects of weathering on a variety of meteorites. Instead, they are the chance observations we made while studying the petrogenesis of rare groups of meteorites whose members increasingly are found either in Antarctica or in hot deserts. In most cases, we made no attempt to analyze areas that were visibly most affected by alteration. Indeed we generally tried to analyze grains or areas as devoid of cracks and fractures as possible. The picture that emerges from this work is thus necessarily incomplete but sufficient to indicate that caution should be used when evaluating data on desert meteorites. We find that different types of meteorites are affected differently and that hot desert meteorites have generally experienced more severe chemical alteration than Antarctic meteorites.

2. SAMPLES AND EXPERIMENTAL METHODS

Thin sections representing 38 different meteorites were examined. After mineral identification and analysis of each section with the

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electron microprobe, the concentrations of rare earth elements (REE) and selected trace and minor elements were measured in situ in a variety of phases, with the Washington University modified CAMECA IMS-3f ion microprobe. The analytical methods are similar to those described by Zinner and Crozaz (1986) and Lundberg et al. (1988). All measurements were made using an O^- primary beam and energy filtering at low mass resolution to remove complex interferences. The resulting mass spectrum was deconvolved to remove simple molecular interferences that are not eliminated with energy filtering, to obtain concentrations of the elements K-Ca-Sc-Ti, Rb-Sr-Y-Zr, and Ba-REE. Details of the analysis program can be found in Alexander (1994) and Hsu (1995). The sensitivity factors used to determine absolute concentrations are the same as in Floss et al. (1998). For each mineral analyzed, a variety of masses were monitored; some are representative of the mineral's composition while others are diagnostic of potential contamination by adjacent minerals, inclusions or terrestrial alteration. The measurement program records the count rates at each mass throughout the analysis (which typically consists of 10 to 20 cycles through the selected set of masses). This makes it possible to determine elemental concentrations in the spot of interest as well as monitor possible contributions from adjacent phases or from inclusions. Errors quoted are 1σ due to counting statistics only. These are generally $<$ 10%, but may range up to 50% for some of the REE in olivine and orthopyroxene. Detection limits are variable, depending on the element and phase being analyzed, but can be as low as a few nanograms per gram in favorable cases. The magnitude of the Ce anomaly is defined here by Ce/Ce*, where Ce is the measured value and Ce* is the interpolated value obtained from La and Pr abundances. Although we have not calculated errors for the magnitudes of the Ce anomalies listed in Tables 1 and 2, and in the Electronic Supplement, we adopt here the convention used by Floss and Crozaz (1991) that only Ce/Ce* ratios $>$ 1.5 or $<$ 0.75 are considered anomalous. REE concentrations in the figures are normalized to the CI abundances of Anders and Grevesse (1989).

3. OBSERVATIONS AND DISCUSSION

Table 1 summarizes some of the relevant properties of the meteorites discussed in this paper, including their terrestrial ages, weathering classification and degree of terrestrial chemical alteration, expressed in terms of the absence or presence of Ce anomalies and/or LREE enrichments. Shock classifications are not listed, as these are not available for the vast majority of the meteorites we studied. Table 2 lists the REE concentrations of those phases shown the figures, while the REE (La–Gd) analyses of all phases that contain Ce anomalies and/or LREE enrichments (e.g., in hot desert meteorites) for the meteorites discussed here are available as an Electronic Supplement. Results for some of the meteorites have previously been reported, as indicated in Table 1 and in the text below; except for LEW 85300 (Floss and Crozaz, 1991) and DaG 476/489 (Crozaz and Wadhwa, 2001), these are limited to brief summaries of the terrestrial alteration effects observed.

For the purpose of clarity, observations and discussion of a given meteorite group will not be separated. Those groups in which chemical alteration is most pronounced will be discussed first. As will become evident, chemical changes are the result of the complex interaction between the meteorite's mineral and chemical compositions, its state of shock, degree of recrystallization, duration of exposure to weathering, and the environment it experienced since it fell on earth. In Antarctica where liquid water is scarcer than in hot deserts, fewer signs of alteration are visible. These include rust, evaporites present in a small fraction of Antarctic samples, and rare clay weathering products (Gooding, 1986). In contrast, in hot desert meteorites, which may have been buried in soil or wind-blown dust, clays and iron oxides and hydroxides often fill cracks and mineral fractures, together with terrestrial silica and carbonates. The potential for chemical alteration of hot desert meteorites is thus a priori more significant than for Antarctic meteorites.

3.1. Eucrites

Eucrites are among the oldest basalts known in the solar system. Their parent body may be the asteroid 4 Vesta (e.g., Binzel and Xu, 1993). The most common non-cumulate eucrites formed through the eruption of basaltic lavas at or near the surface of their parent body, but were subsequently metamorphosed and recrystallized to various degrees. Many eucrites are also breccias whose components may have been affected by shock. Eucrites are composed primarily of plagioclase and pigeonite, which together account for only 5 to 10% of the whole rock LREE abundances, 55 to 75% of the Eu, and 10 to 20% of the HREE abundances (Hsu and Crozaz, 1996). The remaining REE are concentrated in small and rare ($<$0.2 vol.%) calcium phosphates, mainly merrillite, which are heterogeneously distributed in the meteorites (Hsu and Crozaz, 1996).

3.1.1. Antarctic eucrites

We analyzed pyroxene and plagioclase in nine thin sections of seven Antarctic eucrites. They are Lewis Cliff (LEW) 85300 (polymict eucrite), Allan Hills (ALHA) 76005 (polymict breccia including unequilibrated clasts), Asuka (A-) 87272 (monomict eucrite with both coarse-grained regions with a subophitic texture and fine-grained recrystallized granulitic areas), Yamato (Y-) 86763 and Elephant Moraine (EET) 90020 (eucrites with a subophitic texture), A-881388 and A-881467 (granulitic eucrites with highly recrystallized textures). This sampling is unusual in that it includes five of the most highly metamorphosed eucrites known (the last five meteorites mentioned). These were selected to study the effects of extreme metamorphism on the distribution of trace elements in eucritic minerals (Floss et al., 2000; Yamaguchi et al., 2001). Four of these, Y-86763, EET 90020, A-881388, and A-881467 do not show signs of REE mobilization due to terrestrial alteration. However, the other three eucrites do, as described below.

3.1.1.1. LEW 85300 (Floss and Crozaz, 1991). Pyroxene and plagioclase from clasts as well as matrix were studied. Cerium anomalies are present in both minerals, in all clasts as well as in mineral and lithic fragments in the matrix. The majority of pyroxene analyses (32 out of 52) have positive Ce anomalies and six of the pyroxene grains have negative Ce anomalies. In plagioclase, Ce anomalies are also present but less common; only two out of seventeen grains we measured have Ce anomalies, both of them negative. As can be seen in Table 2 and Figure 1, which shows four analyses from a traverse across a single pyroxene grain, the distribution of Ce anomalies is heterogeneous, even on a scale of several hundred micrometers.

3.1.1.2. ALHA76005 (Hsu and Crozaz, 1996). In this eucrite, 16 out of 19 pyroxene measurements and 15 out of 26 plagioclase measurements have positive Ce anomalies. These numbers differ somewhat from those given by Hsu and Crozaz

Table 1. Meteorite terrestrial ages, weathering classification and degree of terrestrial chemical alteration.

				Plagioclase		Pyroxene		Olivine	
Meteorite	Class	Terrestrial Age (ka)	Weathering	Ce anomalies (Ce/Ce*)	LREE rich	Ce anomalies (Ce/Ce*)	LREE rich	Ce anomalies (Ce/Ce*)	LREE rich
ALHA76005§	Eucrite	120 ± 30 [1]	A	15 of 26 (1.5–13.6)		16 of 19 (2.0–9.8)		n.a.*	
EET 90020§	Eucrite	n.a.	A	0 of 16		0 of 17		n.a.*	
LEW 85300§	Eucrite	n.a.	A/B	2 of 17 (0.3–0.5)		38 of 58 (0.3–30.0)		n.a.*	
A-87272§	Eucrite	n.a.	n.a.	0 of 8		8 of 8 (2.3–7.9)		n.a.*	
A-881388§	Eucrite	n.a.	n.a.	0 of 3		0 of 7		n.a.*	
A-881467§	Eucrite	n.a.	n.a.	0 of 4		0 of 7		n.a.*	
Y-86763§	Eucrite	n.a.	n.a.	0 of 8		0 of 10		n.a.*	
ALHA77005§	Shergottite	210 ± 80 [2]	A	0 of 3		8 of 40 (0.6–4.3)		0 of 8	
EETA79001 A§	Shergottite	12 ± 2 [3]	Ae	0 of 2		2 of 21 (1.7–2.5)		0 of 8	
EETA79001 B§	Shergottite	12 ± 2 [3]	Ae	0 of 3		10 of 25 (0.4–29.0)		n.a.*	
LEW 88516§	Shergottite	22 ± 2 [4]	A/B	0 of 6		9 of 45 (0.4–8.5)		0 of 1	
QUE 94201§	Shergottite	290 ± 50 [5]	B/C	0 of 3		2 of 29 (0.15–0.2)		n.a.*	
Y-793605§	Shergottite	35 ± 35 [6]	n.a.	0 of 1		5 of 12 (1.5–3.7)		0 of 2	
DaG476/489§	Shergottite	n.a.	n.a.	0 of 2	0 of 2	5 of 25 (0.06–3.8)	18 of 25	0 of 2	2 of 2
Dhofar 019	Shergottite	340 ± 40 [7]	n.a.	1 of 2 (0.5)	0 of 2	0 of 13	7 of 13	1 of 1 (0.4)	1 of 1
NWA 480	Shergottite	n.a.	n.a.	0 of 2	0 of 2	0 of 10	8 of 10	n.a.*	
NWA 856	Shergottite	n.a.	n.a.	0 of 2	0 of 2	1 of 14 (0.6)	4 of 14	n.a.*	
NWA 1068	Shergottite	n.a.	n.a.	0 of 1	0 of 1	0 of 17	9 of 17	0 of 1	1 of 1
Y-000593/749	Nakhlite	n.a.	n.a.	0 of 3		7 of 12 (1.8–8.8)		1 of 1 (1.6)	
NWA 817	Nakhlite	n.a.	n.a.	n.a.	n.a.	0 of 7	0 of 7	0 of 2	2 of 2
NWA 998	Nakhlite	n.a.	n.a.	n.a.	n.a.	0 of 6	3 of 6	0 of 1	1 of 1
DaG 262	Lunar	80 ± 10 [8]	n.a.	0 of 3	0 of 3	0 of 2	1 of 2	n.a.*	n.a.
DaG 400	Lunar	17 ± 2 [9]	n.a.	0 of 3	0 of 3	n.a.	n.a.	0 of 1	1 of 1
LEW 86010§	Angrite	360 ± 60 [9]	A/B	0 of 7		0 of 9		0 of 7	
LEW 87051§	Angrite	300 ± 60 [9]	A	0 of 2		0 of 5		0 of 2	
A-881371§	Angrite	90 ± 60 [9]	n.a.	0 of 6		0 of 7		0 of 6	
Sahara 99555§	Angrite	<90 [9]	n.a.	0 of 4	0 of 4	0 of 10	0 of 10	4 of 11 (1.6–4.5)	3 of 11
ALHA81261§	Acapulcoite	<50 [10]	A/B	0 of 3		0 of 8		0 of 1	
ALHA81187§	Acapulcoite	24 ± 20 [9]	B/C	0 of 3		0 of 6		n.a.	
EET 84302§	Lodranite	43 ± 22 [9]	B/C	0 of 3		0 of 7		n.a.	
GRA 95209§	Lodranite	<40 [11]	B	0 of 3		0 of 6		n.a.	
GRA 98028	Acapulcoite	n.a.	C	0 of 3		0 of 9		0 of 1	
LEW 86220	Acapulcoite	113 ± 20 [9]	n.a.	0 of 11		0 of 15		n.a.	
LEW 88280§	Lodranite	<50 [10]	B	n.a.		0 of 6		n.a.	
MAC 88177§	Lodranite	n.a.	B/C	n.a.		0 of 4		n.a.	
Dhofar 125	Acapulcoite	n.a.	W1/2	0 of 2	0 of 2	0 of 15	0 of 15	0 of 1	1 of 1
NWA 725	Acapulcoite	n.a.	n.a.	0 of 3	0 of 3	0 of 12	0 of 12	0 of 1	0 of 1
HaH 193	Winonaite	n.a.	W3	0 of 3	0 of 3	2 of 10 (0.4–0.5)	0 of 10	0 of 2	0 of 2
Eagles Nest§	Brachinite	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	1 of 1 (0.1)	1 of 1

The presence or absence of Ce anomalies is shown for all meteorites; for hot desert meteorites the presence or absence of LREE-enrichments due to terrestrial alteration is also indicated; n.a.: no data is available; * these meteorites contain very little olivine to analyze, thus the lack of data. Ce/Ce* gives the magnitude of the Ce anomaly and is less than 1 for negative anomalies and greater than 1 for positive anomalies. Eagles Nest is the only meteorite in which phosphate shows negative Ce anomalies (0.2–0.6) and LREE enrichment. Some results (for meteorites labeled §) have been published in Hsu and Crozaz (1996): ALHA76005; Yamaguchi et al. (2001): EET90020; Floss and Crozaz (1991): LEW85300; Floss et al. (2000): A-87272, A-881388, A-881467, Y-86763; Lundberg et al. (1990): ALHA77005; Wadhwa et al. (1994): EETA79001; Harvey et al. (1993): LEW 88516; Wadhwa et al. (1998a): QUE 94201; Wadhwa et al. (1999): Y-793605; Crozaz and Wadhwa (2001): DaG 476/489; Crozaz and McKay (1990): LEW 86010; Floss et al. (2003a): LEW 87051, A-881371, Sahara 99555; Floss (2000): ALHA81261, ALHA81187, EET 84302, GRA 95209, LEW 88280, MAC 88177; Swindle et al. (1998): Eagles Nest. References: [1] Freundel et al. (1986); [2] Schultz and Freundel (1984); [3] Jull and Donahue (1988); [4] Jull et al. (1994); [5] Nishiizumi and Caffee (1996); [6] Nishiizumi and Caffee (1997); [7] Nishiizumi et al. (2002); [8] Nishiizumi et al. (1998); [9] Nishiizumi (pers. comm.); [10] Schnabel et al. (1998); [11] Terribilini et al. (2000).

(1996) due to the stricter definition of a Ce anomaly used in this paper. Examples are shown in Figure 2 and Table 2.

3.1.1.3. A-87272 (Floss et al., 2000). All eight pyroxene REE patterns from this eucrite exhibit positive Ce anomalies. However, none of the eight plagioclase analyses show a Ce anomaly.

3.1.2. The origin of Ce anomalies in Antarctic eucrites

Cerium anomalies are present not only in our in situ analyses but also in ~61% of the whole rock Antarctic eucrite measurements considered by Mittlefehldt and Lindstrom (1991). Clearly REE mobilization is common in eucrites from the Antarctic environment. Most of the REE in eucrites are initially located in the calcium phosphates and can only be mobilized by their dissolution (Floss and Crozaz, 1991; Mittlefehldt and Lindstrom, 1991). This occurs when the meteorites are exposed to weakly acidic waters while on the surface of the earth. Cerium can then be separated from the other REE because it partially oxidizes to Ce^{4+} which is more insoluble than the other REE. Eucrites are not the only Antarctic meteorites in

Table 2. REE concentrations (in μg/g) in silicates and vein material.

	LEW 85300	LEW 85300	LEW 85300	LEW 85300	ALHA76005	ALHA76005	DaG 476	DaG 476
	px1	px2	px3	px4	pl	px	vein A	vein B
La	0.60 ± 0.03	1.4 ± 0.1	1.3 ± 0.1	0.62 ± 0.04	1.1 ± 0.1	0.050 ± 0.007	9.8 ± 0.5	5.5 ± 0.9
Ce	5.6 ± 0.2	1.1 ± 0.1	7.6 ± 0.2	1.4 ± 0.1	42 ± 1	1.8 ± 0.2	21 ± 1	12 ± 2
Pr	0.17 ± 0.01	0.37 ± 0.02	0.36 ± 0.02	0.20 ± 0.02	0.59 ± 0.04	0.06 ± 0.01	1.8 ± 0.1	1.1 ± 0.3
Nd	0.56 ± 0.02	1.3 ± 0.1	1.4 ± 0.1	0.81 ± 0.04	1.7 ± 0.1	0.29 ± 0.03	6.6 ± 0.3	4.1 ± 0.6
Sm	0.17 ± 0.02	0.37 ± 0.03	0.41 ± 0.03	0.25 ± 0.02	0.50 ± 0.07	0.15 ± 0.02	1.1 ± 0.1	0.7 ± 0.3
Eu	0.001 ± 0.002	0.013 ± 0.003	0.011 ± 0.004	0.009 ± 0.002	1.1 ± 0.1	0.006 ± 0.004	0.16 ± 0.10	n.d.
Gd	0.42 ± 0.03	0.54 ± 0.05	0.71 ± 0.06	0.35 ± 0.04	0.24 ± 0.05	0.26 ± 0.03	0.72 ± 0.16	n.d.
Tb	0.09 ± 0.01	0.14 ± 0.01	0.12 ± 0.02	0.07 ± 0.01	0.03 ± 0.01	0.08 ± 0.01	0.16 ± 0.04	n.d.
Dy	0.80 ± 0.04	1.2 ± 0.1	1.2 ± 0.1	0.66 ± 0.03	0.19 ± 0.06	0.60 ± 0.04	0.78 ± 0.08	n.d.
Ho	0.23 ± 0.02	0.29 ± 0.02	0.27 ± 0.02	0.18 ± 0.01	n.d.	0.12 ± 0.02	0.18 ± 0.03	n.d.
Er	0.87 ± 0.04	1.2 ± 0.1	1.1 ± 0.1	0.63 ± 0.03	n.d.	0.53 ± 0.03	0.49 ± 0.06	n.d.
Tm	0.17 ± 0.02	0.18 ± 0.01	0.18 ± 0.01	0.12 ± 0.01	n.d.	0.07 ± 0.01	0.11 ± 0.02	n.d.
Yb	1.6 ± 0.1	1.7 ± 0.1	1.5 ± 0.1	0.94 ± 0.04	n.d.	0.78 ± 0.07	0.37 ± 0.06	n.d.
Lu	0.24 ± 0.02	0.28 ± 0.02	0.24 ± 0.02	0.18 ± 0.02	n.d.	0.12 ± 0.02	n.d.	n.d.
Ce/Ce*	4.1	0.4	2.7		12.1	7.5		

	DaG 489	HaH 193	NWA 856	Y-000593	DaG 400	DaG 262	Sahara 99555	Sahara 99555
	vein	vein	px	px	ol	px	kir1	kir2
La	2.5 ± 0.3	13 ± 1	0.051 ± 0.004	0.36 ± 0.03	0.024 ± 0.003	0.19 ± 0.02	0.38 ± 0.02	0.42 ± 0.04
Ce	4.4 ± 0.5	29 ± 1	0.10 ± 0.01	5.6 ± 0.3	0.031 ± 0.005	0.48 ± 0.04	3.1 ± 0.2	1.8 ± 0.2
Pr	0.32 ± 0.06	2.7 ± 0.1	0.013 ± 0.001	0.34 ± 0.02	0.003 ± 0.001	0.08 ± 0.01	0.17 ± 0.01	0.13 ± 0.02
Nd	1.1 ± 0.1	9.3 ± 0.3	0.085 ± 0.004	1.8 ± 0.1	0.02 ± 0.005	0.37 ± 0.02	0.78 ± 0.04	0.70 ± 0.05
Sm	0.32 ± 0.09	1.5 ± 0.1	0.055 ± 0.004	0.58 ± 0.05	0.001 ± 0.001	0.19 ± 0.02	0.35 ± 0.04	0.27 ± 0.04
Eu	b.d.	0.26 ± 0.04	0.014 ± 0.006	0.14 ± 0.01	0.003 ± 0.009	0.06 ± 0.03	0.13 ± 0.01	0.10 ± 0.01
Gd	0.30 ± 0.07	0.91 ± 0.17	0.13 ± 0.01	0.58 ± 0.05	0.006 ± 0.005	0.26 ± 0.03	0.60 ± 0.05	0.42 ± 0.06
Tb	0.08 ± 0.02	0.14 ± 0.03	0.029 ± 0.002	0.10 ± 0.02	0.001 ± 0.001	0.06 ± 0.01	0.16 ± 0.01	0.08 ± 0.01
Dy	0.58 ± 0.06	0.57 ± 0.05	0.24 ± 0.01	0.70 ± 0.03	0.004 ± 0.003	0.60 ± 0.03	1.5 ± 0.1	0.65 ± 0.05
Ho	0.16 ± 0.03	0.13 ± 0.01	0.060 ± 0.004	0.14 ± 0.01	0.009 ± 0.002	0.18 ± 0.02	0.43 ± 0.03	0.18 ± 0.02
Er	0.31 ± 0.05	0.43 ± 0.04	0.20 ± 0.01	0.40 ± 0.03	0.03 ± 0.01	0.72 ± 0.03	1.7 ± 0.1	0.55 ± 0.05
Tm	0.09 ± 0.02	0.04 ± 0.01	0.026 ± 0.002	0.06 ± 0.01	0.005 ± 0.002	0.12 ± 0.01	0.28 ± 0.02	0.11 ± 0.02
Yb	0.43 ± 0.06	0.24 ± 0.04	0.23 ± 0.01	0.43 ± 0.04	0.08 ± 0.01	0.80 ± 0.07	2.4 ± 0.1	0.83 ± 0.10
Lu	n.d.	n.d.	n.d.	n.d.	0.008 ± 0.003	0.19 ± 0.02	0.37 ± 0.04	0.14 ± 0.02
Ce/Ce*				3.4			2.9	1.9

Ce/Ce* gives the magnitude of the Ce anomaly and is only shown for phases with Ce anomalies; pl: plagioclase; px: pyroxene; ol: olivine; kir: kirschsteinite; n.d.: not determined; b.d.: below detection.

which we see Ce anomalies in individual grains but they are the only group of cold desert meteorites that show this effect so extensively in their whole rock samples. A combination of properties of Ca-phosphate in eucrites is most probably responsible for this. First, not only are they particularly rich in REE (typically 10,000–20,000 × CI chondrites), but their correspondingly high U and Th concentrations (Crozaz, 1979) must have resulted in extensive radiation damage to the crystals since their formation ~4.5 Ga ago. In addition, phosphates in eucrites are much smaller than in other types of meteorites, another factor contributing to their easier dissolution. Many eucrites also experienced a significant shock event that created defects and thus further crystal damage. Acid dissolution of minerals, and in particular the Ca-phosphates, is certainly more likely to proceed more rapidly when there is damage to the crystal structure. It is thus likely that many phosphates from Antarctic eucrites are partially to completely dissolved and their REE mobilized.

3.1.3. Why are only some Antarctic eucrites affected?

Only a fraction of the Antarctic eucrites we analyzed show Ce anomalies, and only some 60% of the whole rock analyses show them (Mittlefehldt and Lindstrom, 1991). If weathering caused the Ce anomalies, one might expect a correlation with the weathering classification of the meteorites. This classification is based on visible signs of alteration in the form of rust, and is used to divide Antarctic meteorites into three groups—A (minor rustiness), B (moderate), or C (severe). Rust is mostly formed by alteration of metal and troilite grains, and the classification scheme was designed for use primarily on chondrites. Eucrites are metal- and sulfide-poor and, thus, show minimal rust even when most of the metal and troilite are altered. For this reason, the weathering classification used by Johnson Space Center does not adequately measure the degree of alteration of eucrites, and would not work for hot desert eucrites either.

Though we will, in the next sections and whenever possible, attempt to consider the impact of long residence times on earth, unfortunately, it is not possible to test whether alteration is correlated with the terrestrial ages of these meteorites for lack of data. Indeed, of all the eucrites we studied, only the terrestrial age of ALHA76005 was determined (Freundel et al., 1986; see Table 1). However, one property that distinguishes eucrites that contain silicates with Ce anomalies from those that do not is the presence of a network of shock-induced fractures and cracks. LEW 85300 was described by Hewins (1990) as heavily

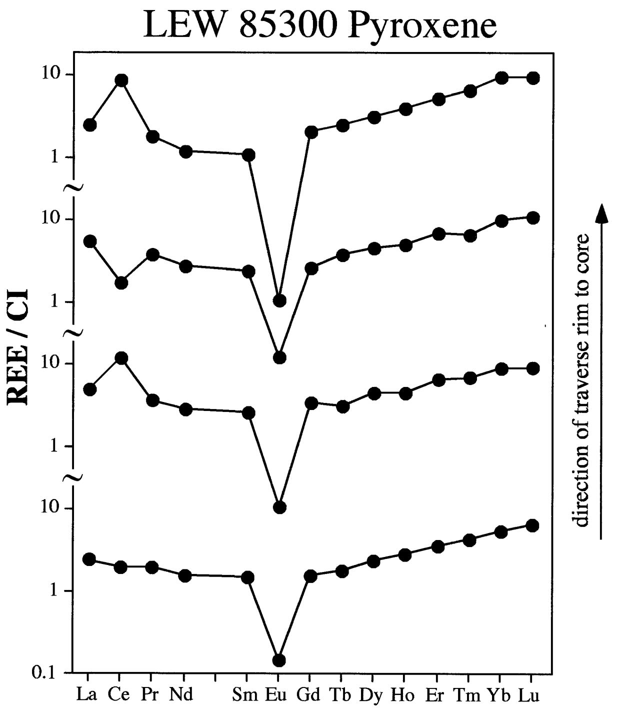

Fig. 1. Ce anomalies in CI chondrite-normalized REE patterns across a single pyroxene grain from LEW 85300 (Floss and Crozaz; 1991). REE concentrations are listed in Table 2.

cracked due to the strong shock event it experienced. A network of microfractures is present in pyroxene but not in plagioclase (Floss and Crozaz, 1991). ALHA76005, in which a larger fraction of the pyroxene and plagioclase have Ce anomalies, has an extensive network of shock-induced microcracks in both minerals (Hsu and Crozaz, 1996). A-87272 appears to be strongly shocked though the plagioclase (maskelynite) is much less fractured and does not exhibit Ce anomalies (Floss et al., 2000). The other four meteorites (with no Ce anomalies in individual silicate grains) all experienced lower degrees of shock. Y-86763 is only weakly shocked; pyroxene and plagioclase show very weak mottled extinctions (Yamaguchi et al., 1997). In EET 90020, Yamaguchi et al. (2001) noted that both pyroxene and plagioclase generally show very sharp extinction, although some plagioclases show very weak mottled extinction. For A-881388 and A-881467, Yamaguchi et al. (1997) did not describe any shock features and noted that many plagioclases and pyroxenes show very sharp extinction, with some showing very weak mottled extinction. Thus, in these meteorites, the occurrence of Ce anomalies in the silicates appears to be correlated with the presence of a network of cracks and fractures that are most likely shock-induced. The major conduit for REE movement is inferred to be through these cracks and defects and, therefore, the mobilized REE probably do not enter the silicate mineral lattices, but rather are trapped in microcracks.

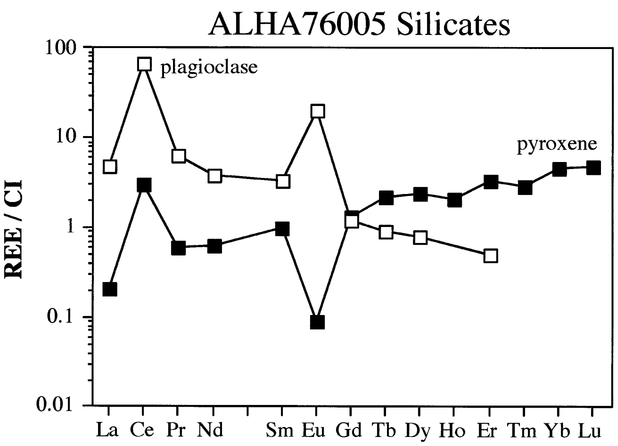

Fig. 2. Ce anomalies in CI chondrite-normalized REE patterns of plagioclase and pyroxene from ALHA76005 (Hsu, 1995). REE concentrations are listed in Table 2.

3.1.4. Isotopic disturbances in Antarctic eucrites

Finally, it should be noted that terrestrial REE mobilization in Antarctic eucrites can also disturb their isotopic systems. In this context, Patchett and Tatsumoto (1980) obtained Lu-Hf data for the eucrite ALHA77302, which did not plot on the 4.56-Ga isochron defined by non-Antarctic eucrites. They attributed the discordance to a loss of Lu during residence in Antarctica. Shimizu et al. (1984) presented Ce isotopic data for the eucrite ALHA78132, which are consistent with a recent resetting of the isotopic system, presumably by terrestrial weathering.

3.1.5. Hot desert eucrites

Having completed our petrogenetic studies of eucrites (Hsu and Crozaz, 1996; Floss et al., 2000; Yamaguchi et al., 2001), we did not analyze individual grains of any hot desert eucrites. There are, however, a few whole rock analyses of non-cumulate eucrites from the Sahara (Barrat et al., 2003) that seem to indicate that, unlike Antarctic eucrites, they have not suffered significant REE mobilization. Barrat et al. (2003) analyzed seven non-cumulate eucrites; one had no evidence of secondary terrestrial mineralization and the others showed various degrees of metal and troilite oxidation, silicate weathering, and filling of fractures by calcite and sometimes gypsum. Except for one specimen with a slight LREE enrichment, no significant weathering effects were observed for the REE, indicating negligible mobilization of these elements. In addition, the Hf/Sm ratios of all these eucrites are chondritic, in contrast with Antarctic eucrites where weathering processes have considerably altered the initial Hf/Sm values (a consequence of the greater mobility of Sm than Hf). From their whole rock analyses, Barrat et al. (2003) concluded that the most sensitive indicators of Saharan weathering are elevated Ba and Sr concentrations due to the formation of secondary carbonates and sulfates that fill fractures, and terrestrial Pb contamination. In another study, Warren (2002) saw severe alteration of the mesostasis material in eucrite NWA 1000 and marginal negative Ce anomalies in the whole rock REE pattern.

However, work on other types of meteorites (mainly the shergottites, see below) indicates that alteration in hot deserts can have important consequences for the distribution of REE.

3.2. Shergottites

There are currently four subgroups of meteorites (shergottites, nakhlites, Chassigny and the ALH 84001 orthopyroxenite) that are believed to have formed on Mars. The shergottites, the youngest meteorites known (Nyquist et al., 2001), are the most numerous of these, comprising at least 18 distinct meteorites. Only two, Shergotty and Zagami, were not found in deserts. There are two types of shergottites, basalts and lherzolites. None are brecciated, but all are severely shocked (much more so than eucrites). It is estimated that the basaltic shergottites experienced peak shock pressures in the range of ~30 to 35 GPa, while the lherzolitic shergottites were exposed to even higher pressures (~40–45 GPa; Stöffler et al., 1986; Stöffler, 2000; Nyquist et al., 2001).

As in eucrites, most of the REE in shergottites are sited in calcium phosphates, mainly merrillite. However REE abundances are much lower (~100 to <1000 × CI) than in eucritic phosphates (10,000–20,000 × CI) and, whereas REE abundances in whole rock non-cumulate eucrites typically are on the order of ~10 × CI, shergottites generally have much lower REE concentrations. Lanthanum in shergottites ranges from ~0.1 to ~20 × CI and the CI-normalized La/Yb ratio ranges from 0.05 to 1. The shape of the REE pattern probably reflects varying degrees of assimilation of a LREE-rich crust-like component by partial melts of a LREE-depleted mantle (Jones, 1989; Wadhwa, 2001; Herd et al., 2002).

3.2.1. Antarctic shergottites

We studied five shergottites found in Antarctica, the basaltic shergottite Queen Alexandra Range (QUE) 94201, EETA79001, which has two distinct basaltic lithologies in igneous contact, and three lherzolitic shergottites, ALHA77005, LEW 88516, and Y-793605. The majority of analyses were of pyroxenes as they are useful to infer the petrogenesis of these meteorites.

3.2.1.1. QUE 94201 (Wadhwa et al., 1998a). Ce anomalies in this meteorite are less common than in other Antarctic shergottites; only two out of 29 pyroxene analyses have Ce anomalies. The small proportion of pyroxenes with Ce anomalies could reflect a lesser degree of weathering than other Antarctic shergottites, despite the relatively long terrestrial age of this meteorite (Nishiizumi and Caffee, 1996; Table 1). Alternatively, it is possible that this meteorite has fewer microfractures to facilitate REE mobilization.

3.2.1.2. EETA79001 (Wadhwa et al., 1994). We used two different thin sections to analyze the two lithologies, A and B, of this meteorite. Lithology A contains olivine-orthopyroxene megacrysts in a fine-grained basaltic groundmass; lithology B is basaltic and more coarse-grained. The terrestrial age of this meteorite is only 12 ± 2 ka (Jull and Donahue, 1988). Pyroxenes with Ce anomalies are found in both lithologies. Two of the pyroxenes (out of 21) in lithology A have positive Ce anomalies (Table 1). Ten out of 25 pyroxenes in lithology B have Ce anomalies (5 are positive and the remaining negative). This difference in the extent of Ce anomalies between the two lithologies is most likely due to a combination of our sampling biases (see "Introduction") and the fact that meteorites, or parts thereof, of different structural states respond differently to shock. Therefore, we do not expect to observe a quantitative correlation between Ce anomalies and degree of shock. However, as we noted for the eucrites, Ce anomalies are primarily observed when a network of shock-induced microfractures is present to facilitate REE mobilization.

3.2.1.3. ALHA77005 (Lundberg et al., 1990), LEW 88516 (Harvey et al., 1993), and Y-793605 (Wadhwa et al., 1999). In all three lherzolites, Ce anomalies are more common than in the less shocked basaltic shergottites (Table 1). However, the fraction of pyroxene grains affected is far lower than in eucrites with Ce anomalies. As in the case of eucrites, pyroxene analyses revealed regions whose Ce anomaly size and sign vary on a micron scale (e.g., Harvey et al., 1993).

3.2.2. The significance of Ce anomalies in Antarctic shergottites

The same process responsible for Ce anomalies in eucrites was at work in shergottites, though on a smaller scale. Terrestrial alteration mobilized the REE from their main reservoir, the calcium phosphates, and the redistribution of these elements was facilitated by the extensive network of fractures and microcracks present in all shergottites. However, the intensity of this process is reduced in shergottites, despite the fact that they experienced higher shock pressures than the eucrites. Only one Ce anomaly has ever been reported in a whole rock sample of the ALHA 77005 Antarctic shergottite (Shimizu and Masuda, 1981), indicating again that chemical alteration is less significant than in eucrites. A plausible reason is to be found in the sizes and REE abundances of the phosphate grains in shergottites. Indeed, compared to eucritic phosphates, their larger sizes make them more difficult to weather and their lower REE contents reduce the chemical impact of their dissolution.

3.2.3. Hot desert shergottites

In recent years, an increasing number of shergottites have been found in hot deserts. We discuss here five of these meteorites: DaG 476/489, Dhofar 019, NWA 1068, NWA 480, and NWA 856. All five are basaltic in composition, although the first three contain megacrysts of olivine (and, in the case of DaG 476/489, orthopyroxene).

3.2.3.1. DaG 476/489 (Crozaz and Wadhwa, 2001). A thin section of each of the paired shergottites DaG 476 and DaG 489 was studied. This is the first hot desert shergottite in which we observed terrestrial alteration effects and also the one we analyzed most comprehensively. In common with other shergottites, all silicate phases, and olivine and orthopyroxene in particular, exhibit fractures throughout. The largest of these are filled with calcium carbonate. There are several occurrences of sizable amorphous regions of general silicate composition, with elevated P and S concentrations, which are likely to be impact melt (Zipfel et al., 2000; Wadhwa et al., 2001a). These seem particularly susceptible to terrestrial weathering and are stained reddish-brown. These patches, as well as merrillite, maskelynite, pyroxene, and olivine were analyzed. Merrillite does

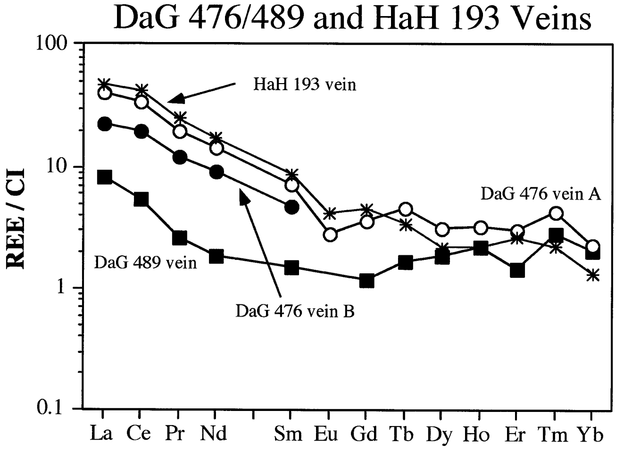

Fig. 3. CI chondrite-normalized REE patterns for amorphous alteration material from veins in the paired shergottites DaG 476 and DaG 489 (Crozaz and Wadhwa, 2001). Also shown is the REE pattern for amorphous alteration material from veins in the winonaite HaH 193. REE concentrations are listed in Table 2.

not show any signs of chemical alteration in this or any of the other hot desert shergottites. Maskelynite also appears to be largely unaltered. However, low-Ca pyroxene and olivine are frequently enriched in LREE. Of a total of 13 pigeonite and 12 orthopyroxene analyses, 18 (9 in each type of pyroxene) show elevated La, 4 with a negative and 1 with a positive Ce anomaly. Olivine also has elevated LREE concentrations that are correlated with Sr and Ba enrichments. Compared to Antarctic shergottites, the Sr concentrations in the olivine of the DaG meteorites are higher by a factor 10 to 100, and Ba concentrations are higher by a factor of 6 to 10. The amorphous patches have striking LREE enrichments (Fig. 3, Table 2) and contain elevated concentrations of a host of other elements, including Sr and Ba.

3.2.3.2. Dhofar 019. Like DaG 476/489, this meteorite was probably exposed to a shock pressure of 30 to 35 GPa (Nyquist et al., 2001). Olivine and pyroxene are inhomogeneously enriched in La, Sr, and Ba, with variations that correlate well with each other during a single measurement (Fig. 4). Out of 13 pyroxene analyses, 7 show LREE enrichments, and none have Ce anomalies. Maskelynite has a LREE-depleted pattern and of our two analyses, one has a negative Ce anomaly (Ce/Ce* ~ 0.5). One olivine grain also has a negative Ce anomaly. REE concentrations in individual minerals and whole rock samples are also reported by Taylor et al. (2002). These authors did not find any Ce anomalies in the pyroxenes they measured, but some of their maskelynite analyses do show negative Ce anomalies attributed to terrestrial alteration. A positive Ce anomaly was also observed in the smaller of two whole rock samples analyzed for trace elements by Taylor et al. (2002), another indication that terrestrial alteration modified REE patterns in this meteorite.

3.2.3.3. NWA 1068. Merrillite and maskelynite are not affected, but olivine is LREE-enriched, as are some of the pyroxene grains (9 out of 17). In pyroxene, the enrichment in LREE is only apparent in low-Ca pyroxene. Barrat et al. (2002a) reported elevated Sr, Ba, and Pb abundances in this meteorite.

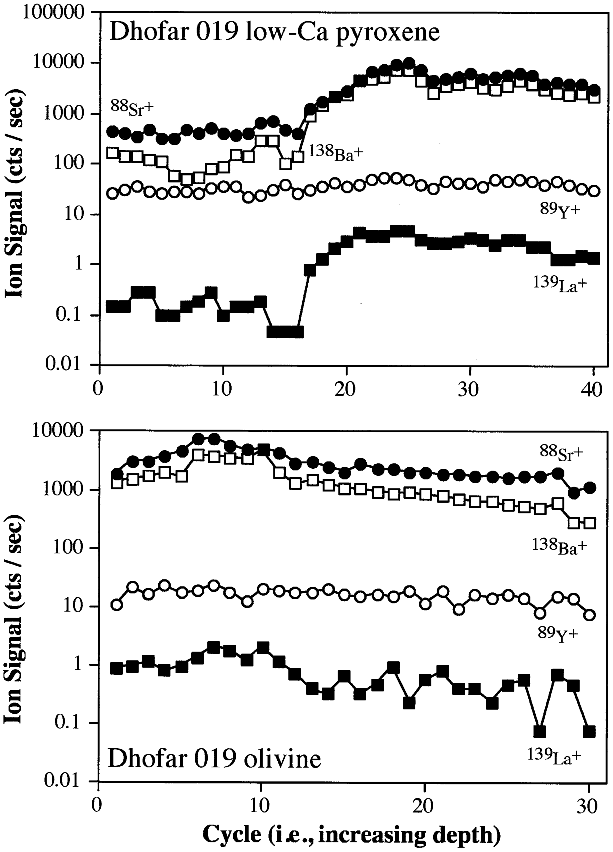

Fig. 4. Ion signals for ^{88}Sr$^+$, ^{89}Y$^+$, ^{138}Ba$^+$, and ^{139}La$^+$ during the analyses of a low-Ca pyroxene and an olivine in the Dhofar 019 basaltic shergottite. Higher cycle numbers indicate increasing depth of analysis, with 40 cycles representing a depth of ~15 to 20 μm. Errors on individual cycle results are similar in magnitude to the typical point to point variations from one cycle to the next.

3.2.3.4. NWA 480. Barrat et al. (2002b) reported that the Th/U ratio of this meteorite, which has a fresh appearance, is normal and that the Ba and Sr abundances in whole rocks are not outside the trend defined by unweathered shergottites. In the present work, no La enrichment is observed in individual grains of augite, but all but one of the low-Ca pyroxenes have elevated La due to terrestrial contamination.

3.2.3.5. NWA 856. This basaltic shergottite described by Jambon et al. (2002) is fractured at all scales. However, the Th/U ratio and the Sr and Ba abundances indicate that terrestrial weathering was not extensive. Out of 14 pyroxene analyses, only 4 show La enrichments (one is shown in Fig. 5 and Table 2) and 1 has a small negative Ce anomaly (Ce/Ce* ~ 0.6). The two augite grains analyzed were not affected.

Basically, we see the same alteration effects in all these shergottites, although they are more pronounced in some than in others. The composition of merrillite is not affected, as it is the main REE carrier. In most instances, the REE pattern of maskelynite does not show a dramatic upturn at La, because the

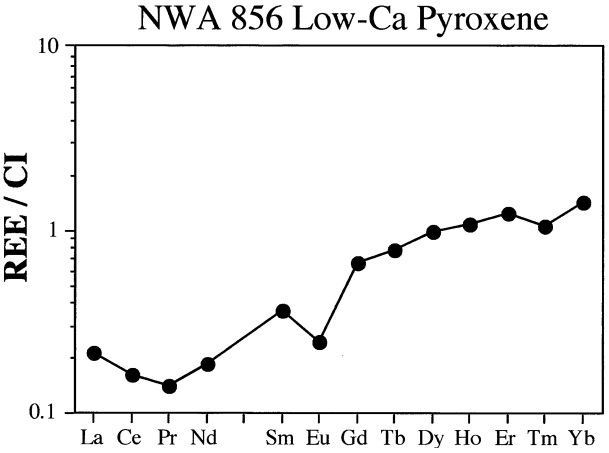

Fig. 5. CI chondrite-normalized REE pattern for a low-Ca pyroxene from the basaltic shergottite NWA 856. REE concentrations are listed in Table 2.

indigenous LREE pattern is usually either flat or LREE-enriched (one exception is the distinctly LREE-depleted maskelynite of Dhofar 019, some areas of which show negative Ce anomalies). On the other hand, olivine and low-Ca pyroxene, usually characterized by steep LREE-depleted patterns, can show a clear La enrichment and, occasionally, a Ce anomaly. However, not all grains of olivine and low-Ca pyroxene are affected. The fraction involved is generally correlated with the extent of the network of microfractures that facilitates REE redistribution. We first noted (Crozaz and Wadhwa, 2001) that the LREE enrichments in weathered locations are due to contamination with a component with average terrestrial crustal composition (Taylor and McLennan, 1985) as REE patterns for all of these are parallel. Dreibus at al. (2001) have since analyzed desert soils and caliche (a desert weathering product partly covering the meteorites) and found that both are LREE-enriched with REE patterns that are parallel to that of the amorphous silicate material from veins in the DaG meteorites. There is, thus, no question that the LREE enrichments observed in individual grains of olivine, pyroxene, and in weathered areas, are due to the introduction of a terrestrial component.

In some cases, the LREE enrichment is also noticeable in whole rock analyses. Barrat et al. (2001) measured three aliquots of the DaG 476 meteorite and observed that all the REE patterns had an upturn to La, as well as high Sr, Ba, and U concentrations. But the most striking example of LREE enrichment in a hot desert meteorite was noted for the diogenite Tatahouine (Barrat et al., 1999). This meteorite fell in 1931. Samples were collected then and again in 1994, to evaluate the changes over 60 yr of residence in the desert. Not only were the samples collected in 1994 visibly altered and contained terrestrial calcite; they had also acquired significant enrichments of LREE, Rb, and Sr. It is no coincidence that DaG 476 and Tatahouine are so far the meteorites with the most disturbed LREE concentrations. They also originally had the most LREE-depleted whole rock patterns. Because their natural LREE concentrations are so low (La $\sim$ 0.5 $\times$ CI and $\sim$0.01 $\times$ CI, respectively, in whole rock samples of DaG 476 and Tatahouine), any terrestrial contamination will have a profound effect on the LREE budget. Furthermore, the rapid alteration of Tatahouine indicates that the intensity of terrestrial alteration cannot possibly be linear with time. Instead, as suggested by Bland et al. (1996a), the initial weathering must be the most substantial. These authors emphasized the importance of porosity to effectively alter meteorites. However, as pores gradually fill, the ability of water to circulate is reduced and a much slower oxidation regime sets in; the alteration process slows down.

3.2.4. *Other chemical changes due to weathering in shergottites (and other hot desert meteorites)*

A number of other changes are brought upon the meteorite as it resides in a hot desert. Contact with the terrestrial atmosphere introduces terrestrial heavy noble gases (Scherer et al., 1994). In carbonaceous chondrites from the Sahara, organic C, N, and H show depletions compared to their analogs from other regions of the world (Ash and Pillinger, 1995). With increasing oxidation and hydrolysis, Mg and Si abundances (Bland et al., 1996b), and Fe, Ni, and Co (Scherer et al., 1994) all decrease in chondrites. Increases in Sr, Ba, and U have already been mentioned as good indicators of desert alteration (Barrat et al., 2001). A host of other elements are probably also affected, but to a lesser extent. Therefore, caution should always be exercised when using whole rock or mineral separate data for hot desert meteorites. As far as the REE are concerned, the whole rock patterns are usually not significantly modified and valuable petrogenetic information can still be retrieved. In addition, using data for areas within individual grains, as we do, offers the advantage of identifying grains, or portions of grains, which have preserved their chemical integrity. However, there is a more troublesome consequence of hot desert alteration that needs to be addressed and is discussed below.

3.2.5. *Isotopic disturbances in shergottites from hot deserts*

Dating shergottites is not trivial even when they are found elsewhere than hot deserts. Interpretations of radiometric age data are complicated by their petrogenesis on a geologically active planet and complex postcrystallization histories, combined with a lack of geologic context for these samples (Nyquist et al., 2001). Furthermore, these rocks present the analytical challenges associated with isotopic analyses of relatively young samples having trace abundances of parent-daughter elements of interest.

Because shergottites have only recently been found in hot deserts, there are few chronological studies of these meteorites. However, the nature of terrestrial alteration in hot deserts, and its potential to contaminate the meteorites in a variety of elements, certainly imply that extreme caution will have to be exercised when attempting to date not only shergottites but any meteorite that experienced similar weathering conditions. Data on leachates, in particular, should be considered as suspicious.

Weathering can introduce ^{40}Ar from the terrestrial atmosphere and this needs to be corrected for appropriately. Pb contamination, as well as U enrichment, can disturb the U-Pb isotope systematics of these meteorites. The Rb-Sr and Sm-Nd methods also have their problems as illustrated by a growing number of recent isotopic studies. From these (Nyquist et al., 2000, on the shergottite Los Angeles; Jagoutz et al., 1999, and

Borg et al., 2000, on the shergottite DaG 476; Borg et al., 2001, on the shergottite Dhofar 019; Shih et al., 2002a, on the lunar meteorite Dhofar 287), a consistent picture emerges. These hot desert meteorites are so altered that their isotopic systematics are profoundly disturbed. Despite aggressive leaching to eliminate desert alterations and exclusion of the acid leachates from analysis, contamination problems still persist.

The Rb-Sr method is essentially useless in the hot desert meteorites. Whole rock and whole rock leachates are so overwhelmed by contamination that their $^{87}Sr/^{86}Sr$ ratio can approach the present seawater value (as shown for a lunar meteorite by Shih et al., 2002a). And, despite leaching, the pyroxenes are still dominated by the desert alteration. The only phase that is probably not significantly altered by the addition of terrestrial Sr is plagioclase because it is the main carrier of this element and typically contains a high concentration of it. However, plagioclase usually has a low abundance of Rb and, therefore, addition of even a modest amount of the highly mobile alkali elements during terrestrial weathering would easily affect the Rb/Sr ratio of this phase.

The Sm-Nd method has its problems too. Earlier, we noted (Crozaz and Wadhwa, 2001) that leachates of DaG 476 (Jagoutz et al., 1999) were dominated by a LREE-rich component introduced by terrestrial crustal contamination. This LREE contamination is, however, heterogeneously distributed in this meteorite. Indeed, Borg et al. (2000) prepared leachates of DaG 476 that do not appear to be contaminated. Further, it has been demonstrated that Sm-Nd data for aggressively leached whole rock and pyroxenes of hot desert meteorites can define a valid isochron (e.g., Shih et al., 2002a). It is thus possible, by careful analysis and consideration of the potential problems, to determine the ages of hot desert meteorites using the Sm-Nd method.

3.3. Nakhlites

The nakhlites are a small group of only six meteorites that are also thought to come from Mars. They are unbrecciated cumulate igneous rocks composed mainly of augite with olivine and intercumulus mesostasis. All are moderately shock metamorphosed, but have experienced lower shock levels (~20 GPa) than shergottites (30–45 GPa; Nyquist et al., 2001). Two of them, NWA 817 and NWA 998, were found in hot deserts, whereas the paired Y-000593/749/802 nakhlites were recently recovered from Antarctica.

3.3.1. *NWA 817 (Wadhwa et al., 2001b)*

This nakhlite from the Moroccan Sahara is similar to Nakhla but has a larger proportion of mesostasis (~20%; Mikouchi and Miyamoto, 2001; Sautter et al., 2002), which is also reflected in its higher whole rock REE content (Sautter et al., 2002). Only the olivine analyses show elevated LREE, which are attributed to terrestrial alteration. The absence of extensive LREE enrichment in phases other than olivine is consistent with other indicators of terrestrial alteration. These include the lack of weathering of the sulfides (Gillet et al., 2002) and the fact that only Pb and Ba abundances are elevated in this meteorite (Sautter et al., 2002). In addition, NWA 817 is much less fractured than Nakhla, another characteristic that explains its resistance to extensive alteration.

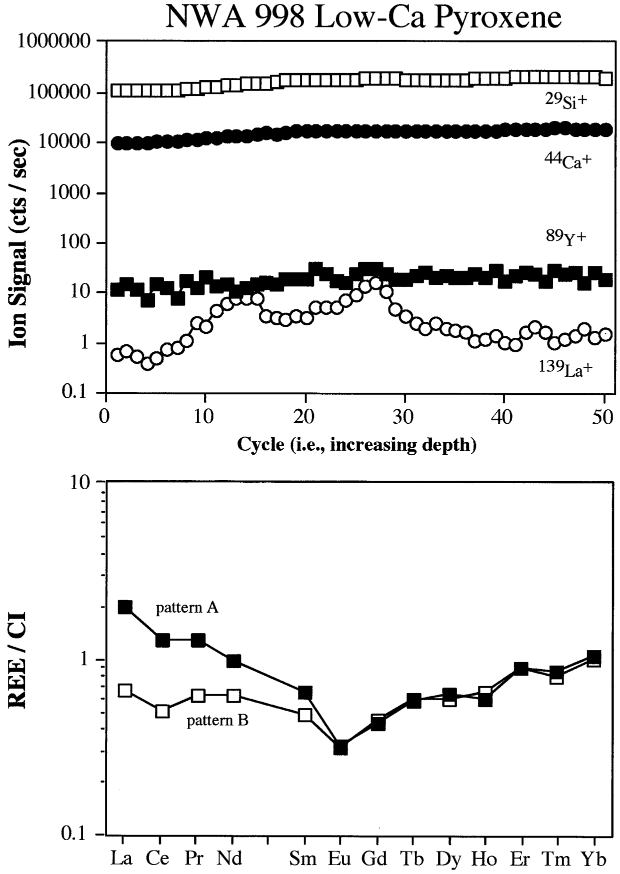

Fig. 6. Top: ion signals for $^{29}Si^+$, $^{44}Ca^+$, $^{89}Y^+$, and $^{139}La^+$ during the analysis of a low-Ca pyroxene in the NWA 998 nakhlite; higher cycle numbers indicate increasing depth of analysis. Errors on individual cycle results are similar in magnitude to the typical point to point variations from one cycle to the next. Bottom: CI chondrite-normalized REE patterns for this pyroxene for cycles 1 to 50 (pattern A) and for cycles 1 to 8 combined with 39 to 50 (pattern B).

3.3.2. *NWA 998*

This nakhlite was recently described by Irving et al. (2002). Like other nakhlites, it is composed predominantly of high-Ca pyroxene along with subordinate olivine. However, it is unusual among the nakhlites in having low-Ca pyroxene as a primary magmatic phase. We have made analyses of high- and low-Ca pyroxenes, olivine, plagioclase and apatite in this meteorite. Although a hand-sample of this nakhlite looks heavily altered and is very friable, no significant effects of terrestrial alteration were discerned in the high-Ca pyroxene, plagioclase and apatite. However, olivine has a slight LREE-enrichment (CI normalized La/Nd ~ 2) that is attributed to terrestrial contamination. In addition, three of the six low-Ca pyroxenes analyzed show LREE-enrichment. In fact, we noted distinct variations in the concentrations of La (and other LREE) during the course of one of these analyses (Fig. 6). As seen in Figure 6, these variations are independent of the abundances of Y (and HREE) and of major elements such as Si and Ca. This elimi-

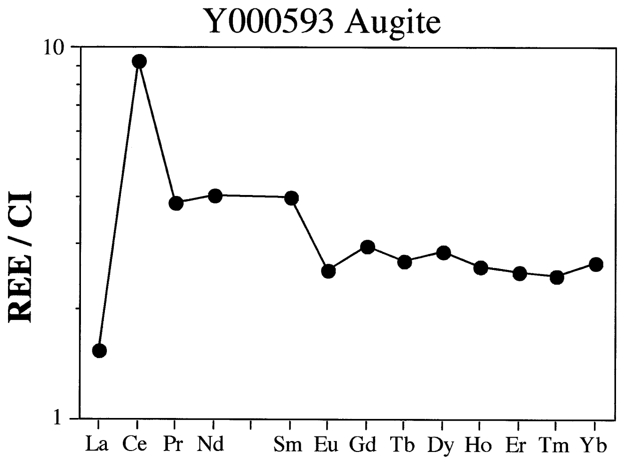

Fig. 7. CI chondrite-normalized REE pattern for an augite from the Y000593 nakhlite. REE concentrations are listed in Table 2.

nates the possibility of the LREE being contributed from silicate inclusions or other LREE-rich phases indigenous to this meteorite and strongly suggests that the LREE were added as a terrestrial contaminant, the distribution of which varies considerably on the micron scale.

3.3.3. Y-000593/749/802

These paired meteorites represent the first nakhlite and the largest achondrite recovered from the Antarctic (Kojima et al., 2002). Preliminary investigations show that the mineralogical and geochemical characteristics and crystallization and exposure ages of this Antarctic nakhlite are similar to those of previously known nakhlites (e.g., Imae et al., 2002; Mikouchi et al., 2002; Nakamura et al., 2002; Okazaki et al., 2002; Shih et al., 2002b). Although geochemical and isotopic (Rb-Sr and Sm-Nd) results obtained so far indicate that these paired meteorites suffered minimal weathering in the Antarctic, there is some evidence that Pb isotopic systematics may have been disturbed by the addition of terrestrial Pb (Yamashita et al., 2002). We analyzed minerals in one thin section each of Y000593 and Y000749. Ce anomalies are pervasive in all silicate phases except plagioclase. A small positive Ce anomaly (Ce/Ce* ~ 1.6) is present in our single olivine analysis. Large positive Ce anomalies are present in 7 of the 12 augites measured in the two sections (Ce/Ce* ~ 2–9; an example is shown in Fig. 7 and Table 2). Finally, a positive Ce anomaly is also observed in one of the two mesostasis regions analyzed (Ce/Ce* ~ 2). Although augite is the dominant phase present in this meteorite, some 90% of the Ce appears to reside in chlorapatite (Wadhwa et al., 1995). Thus, we would not expect the presence of positive Ce anomalies in individual augite grains to be reflected in whole rock analyses.

3.4. Lunar Meteorites

In addition to the lunar rocks brought back to earth by the Apollo missions, some 25 distinct (i.e., not paired) lunar meteorites have been found in hot and cold deserts. Almost all are regolith or impact melt breccias and most are dominated by feldspathic components, indicating that they originated from the lunar highlands. We studied two lunar regolith breccias from the Saharan desert, DaG 262 and 400 (Floss and Crozaz, 2001). Both rocks are anorthositic highland breccias (Bischoff et al., 1998; Zipfel et al., 1998) with affinities to ferroan anorthosites (FANs). Although these two meteorites are very similar, noble gas chemistry indicates they are not paired with each other (Scherer et al., 1998). They also have different terrestrial ages, 80 ka for DaG 262 (Nishiizumi et al., 1998) and 17 ka for DaG 400 (Nishiizumi, private communication).

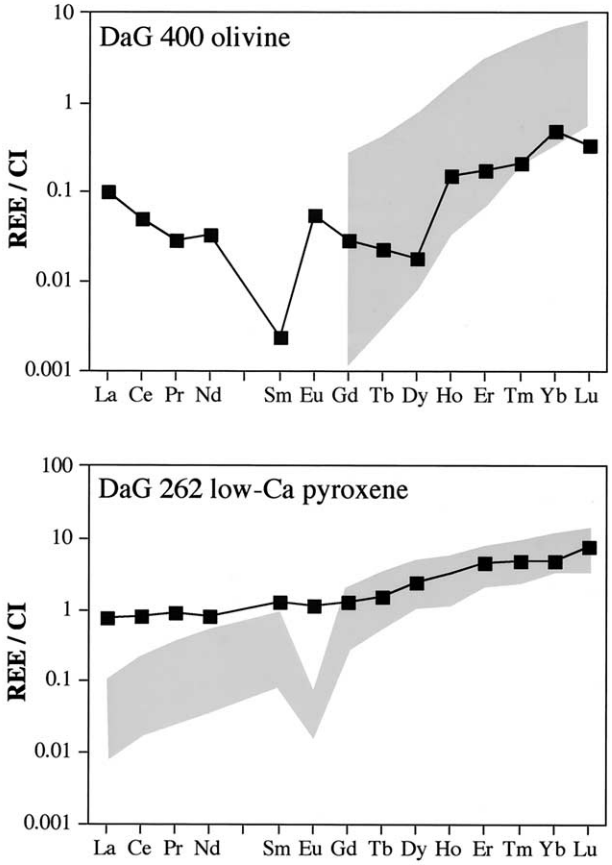

Fig. 8. CI chondrite-normalized REE patterns for olivine and low-Ca pyroxene from DaG 262 and DaG 400. Gray shaded areas show the range of compositions observed in typical ferroan anorthosites (Floss et al., 1998). REE concentrations are listed in Table 2.

Because of the anorthositic nature of these meteorites, we were only able to find a few mafic minerals that were large enough to analyze: one olivine in DaG 400, and one high-Ca pyroxene and one low-Ca pyroxene each in DaG 262. LREE abundances in the olivine are distinctly elevated and the low-Ca pyroxene is also LREE-enriched compared to its counterparts from the ferroan anorthosites (Fig. 8, Table 2). Both minerals also have Sr and Ba concentrations that are as much as 3 orders of magnitude higher than in olivine and low-Ca pyroxene from the FANs (Fig. 9). REE abundances in the high-Ca pyroxene fall within the range observed for FAN high-Ca pyroxenes (Floss et al., 1998), but both Sr and Ba concentrations in this grain are higher than expected (Fig. 9). We also measured six plagioclase grains from DaG 262 and DaG 400. All have REE and Sr concentrations within the range observed for FAN plagioclase (Floss et al., 1998), but two of the grains have elevated Ba concentrations (Fig. 9).

In addition to the individual minerals, we measured 17 clasts from the two meteorites. Most of these are feldspathic fine-grained to microcrystalline melt breccias, but mafic-rich granulitic clasts and cataclastic anorthosites were also analyzed. Concentrations of Sr and Ba are variable in the clasts, but tend to be higher than those observed in plagioclase, the main carrier of these elements among the silicate phases. Enrichments of Sr and Ba are also observed in whole rock analyses of these two meteorites (Bischoff et al., 1998; Zipfel et al., 1998), as are elevated U abundances (Bukovanska et al., 1999). Bischoff et al. (1998) furthermore noted the presence of several calcite-filled fractures in DaG 262 that are apparently of terrestrial origin, as well as occasional grains of barium sulfate. Despite this mineralogical evidence of terrestrial alteration and the elevated Sr and Ba concentrations in whole rock analyses, we observed only limited evidence of alteration of REE abundances in these meteorites, as noted above. This is most likely due to the fact that both DaG 262 and DaG 400 consist predominantly of feldspathic components. We expect terrestrial weathering to more profoundly affect the REE in lunar meteorites that did not form in the lunar highlands, i.e., are not dominated by a feldspathic component. Lunar meteorites dominated by mare components contain a higher proportion of mafic minerals that are more sensitive to alteration of the LREE than feldspar. In this context, Shih et al. (2002a) noted severe disturbance of the Rb-Sr system, as well as disturbance of the Sm-Nd systematics, in Dhofar 287, a lunar meteorite consisting of ~95% mare basalt component. We note also that DaG 262, with its much longer terrestrial age, is not more altered than DaG 400.

3.5. Angrites

The angrites are a group of seven very old achondrites (Wasserburg et al., 1977; Lugmair and Galer, 1992; Nyquist et al., 1994, 2003; Premo and Tatsumoto, 1995) characterized by unusual mineral assemblages and refractory element enrichments. All angrites, except for Angra dos Reis, have igneous (granular or basaltic) textures, and contain abundant olivine (including kirschsteinite) and anorthite, in addition to aluminian-titanian diopside (formerly called fassaite). Geochemical data also emphasize the distinct nature of Angra dos Reis (Crozaz and McKay, 1990). Three of the angrites come from Antarctica and two from hot deserts.

We did not find evidence of terrestrial alteration in any of the three Antarctic angrites (LEW 86010, LEW 87051 and A-881371), despite their relatively long terrestrial ages (Nishiizumi, private communication; Table 1). This is probably due to the fact that these meteorites all have low shock levels and, thus, do not have an extensive system of microfractures along which the REE can be mobilized. Nevertheless, we note that Lugmair and Galer (1992) found the LEW 86010 angrite to be extensively contaminated with terrestrial Pb. In addition, although Sm-Nd systematics in this angrite appear to be unaffected, these authors suggested that there may be addition of alkali elements concomitant with Pb contamination, which may also affect Rb-Sr systematics.

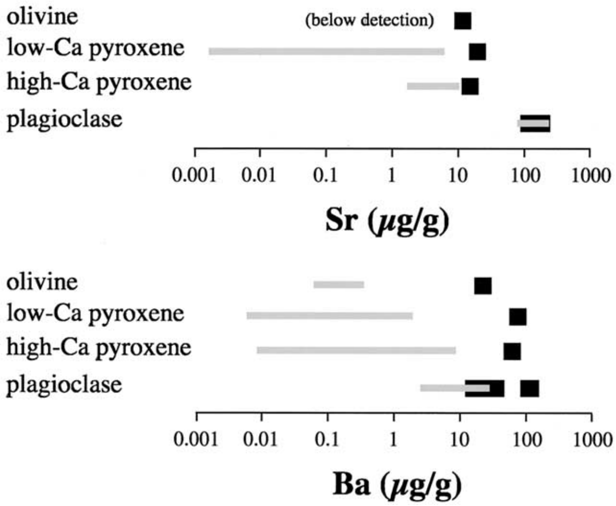

Fig. 9. Sr and Ba concentrations (μg/g) in minerals from DaG 262 and DaG 400. Shaded lines show the range of concentrations observed in ferroan anorthosites (Floss et al., 1998). Sr concentrations are below detection in FAN olivines.

We studied one of the hot desert angrites, Sahara 99555, and find that it does contain evidence of chemical changes due to terrestrial alteration (Floss et al., in press). No aluminian-titanian diopside, anorthite or phosphate grains that we analyzed are affected, but several grains of olivine and kirschsteinite exhibit LREE enrichments, together with positive Ce anomalies (Fig. 10, Table 2). The presence of terrestrial contamination in this meteorite is particularly interesting because, unlike some of the other hot desert meteorites we have studied, Sahara 99555 has a very fresh unweathered appearance in thin section. Thus, we note that chemical changes occur even without visible evidence of alteration.

3.6. Acapulcoites and Lodranites

Primitive achondrites are an important source of information about early differentiation processes on asteroids. These meteorites typically exhibit achondritic textures, but have retained at

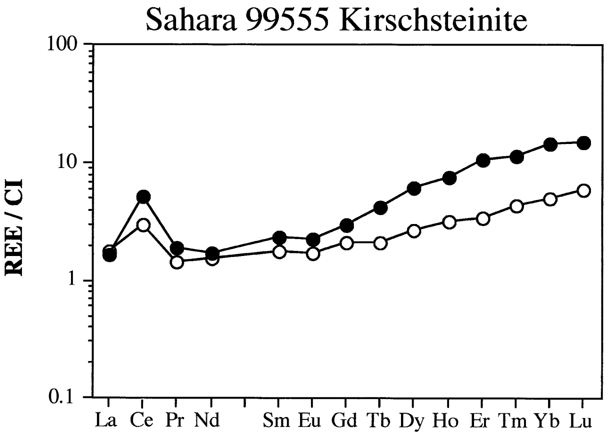

Fig. 10. CI chondrite-normalized REE patterns for two kirschsteinite grains from Sahara 99555. REE concentrations are listed in Table 2.

least some primitive compositional characteristics. One of the best defined groups of primitive achondrites are the acapulcoites and related lodranites. These meteorites are thought to have originated on a common parent body that experienced variable degrees of heating, resulting in complex partial melting and melt migration processes (McCoy et al., 1996, 1997a, 1997b; Mittlefehldt et al., 1996; Floss, 2000).

Our petrogenetic studies of the acapulcoites–lodranites (Floss, 2000, 2002) so far include investigation of a number of meteorites from Antarctica (ALHA81261, ALHA81187, EET 84302, Graves Nunataks [GRA] 95209, GRA 98028, LEW 86220, LEW 88280 and MacAlpine Hills [MAC] 88177). We do not observe evidence for terrestrial alteration in any of these meteorites. In addition, we have studied the hot desert acapulcoites, Dhofar 125 and NWA 725 (Floss, 2002). In these meteorites as well, there is little indication of terrestrial contamination. One olivine grain from Dhofar 125 shows somewhat elevated La and Ce concentrations, but Sr and Ba abundances are low. Other phases do not appear to be altered. The low degree of terrestrial alteration noted in most of these meteorites is probably due to the fact that they are highly recrystallized with compact granular textures. In addition, most appear to have experienced only low shock levels and are not extensively fractured. Thus, there are few conduits available in these meteorites for REE remobilization or the introduction of terrestrial contaminants.

3.7. Winonaites

The winonaites are similar to acapulcoites and lodranites in that they are essentially chondritic in mineralogy and bulk chemical compositions, but have achondritic textures (Kimura et al., 1992; Benedix et al., 1998; Yugami et al., 1998).

One of the winonaites we are currently studying is Hammadah al Hamra (HaH) 193, from Libya. HaH 193 is dominated by orthopyroxene with lesser amounts of plagioclase, olivine, clinopyroxene and the amphibole fluoro-edenite (Floss et al., 2003). Our section of HaH 193 also shows moderate to extensive reddish-brown staining of the silicates and has a large fracture filled with weathered amorphous Ca-Si-rich material. The primary minerals in HaH 193 show little evidence for terrestrial alteration: only 2 orthopyroxene grains (out of 15 silicate grains measured) have negative Ce anomalies. However, the amorphous material filling the fracture is distinctly LREE-enriched with a pattern that is very similar to the REE pattern observed in amorphous vein material from the shergottite DaG 476 (Fig. 3, Table 2), studied by Crozaz and Wadhwa (2001). These REE patterns are consistent with the REE patterns seen in average crustal material (Taylor and McLennan, 1985). Furthermore the HaH 193 vein material has elevated Sr (800–8000 μg/g) and Ba (100–200 μg/g) concentrations, again similar to those seen in the DaG 476 veins. Crozaz and Wadhwa (2001) argued that the vein material in DaG 476 was likely to be impact melt that had been preferentially altered due to its amorphous nature. It is not clear whether the material filling the large vein in HaH 193 has a similar origin, since this meteorite is not extensively shocked and the material is more Ca-rich than typical impact melt is likely to be. Nevertheless, whatever its origin, its current REE signature clearly reflects extensive terrestrial alteration. The fact that the primary silicates in HaH 193 show only little evidence for terrestrial contamination probably reflects, as in the acapulcoites, the highly recrystallized nature and low shock level of this meteorite.

3.8. Brachinites

Brachinites are yet another group of primitive achondrites. Only six are known (Brachina, Eagles Nest, ALH 84025, the paired EET 99402/99407, Hughes 026, and Reid 013). They have near chondritic major element whole rock compositions, but distinct igneous textures, and are very old (Wadhwa et al., 1998b).

Eagles Nest, one of the brachinites, is a hot desert meteorite. It was found in central Australia and is highly weathered. Cracks and vugs are numerous in its interior. We analyzed its apatite and olivine and found them to be highly LREE-enriched with deep negative Ce anomalies (Ce/Ce* $\sim$ 0.1; Swindle et al., 1998). This, again, implies terrestrial weathering. The whole rock analyses (Swindle et al., 1998) also show LREE enrichment and negative Ce anomalies, and much higher REE concentrations than expected in such a mafic meteorite. LREE-rich material from the Australian desert obviously contaminated the meteorite and also contributed terrestrial K that disturbed the K-Ar isotopic system. Using the chemical composition of Eagle Nest to infer its petrogenesis is, therefore, excluded.

4. CONCLUSIONS

This work has shown that meteorites found in Antarctica as well as in hot deserts have experienced different degrees of terrestrial alteration since their fall on earth.

In Antarctica, remobilization of the lanthanides occurred mainly while the meteorites were exposed on the ice. Oxidation of Ce^{3+} to the more insoluble Ce^{4+} caused the separation of this element from the other REE, resulting in Ce anomalies in the REE patterns, mainly of pyroxene. While it is difficult to correlate this effect to the weathering grades or the terrestrial ages of the meteorites, the Ce anomalies are clearly related to the presence of networks of fractures and microcracks that facilitate the mobilization of the REE. These networks are most probably the result of shock events experienced by the meteorites. However, not all grains of a given phase in the same meteorite are affected and even portions of a single grain may have been altered differently. This process was most efficient in eucrites whose small highly REE-U-Th-rich phosphates dissolve easily. Cerium anomalies are also widespread in shergottites, but are less common in other types of meteorites.

Hot desert meteorites have spent less time on earth since their fall, but often have been rapidly weathered, particularly if they were heavily shocked. As pores in the meteorites gradually filled in, the alteration process slowed down. Ce anomalies can also occur in these meteorites but the most important effect is a LREE enrichment that is most prominent in low-Ca pyroxene, olivine, and heavily weathered areas. The LREE contamination is most visible in the shergottites and the brachinite Eagles Nest. Meteorites that experienced low shock levels or are highly recrystallized, such as the angrites, acapulcoites/lodranites, and winonaites do not show significant LREE contamination. Basaltic hot desert lunar meteorites are expected to be more affected than those from the lunar highlands.

Isotopic disturbances have been observed in Antarctic meteorites but are more prevalent in hot desert meteorites. Contamination of the latter by Sr, Pb, and U restricts the use of the Rb-Sr and U-Pb methods to date these objects. Successful application of the Sm-Nd method requires extensive leaching of the whole rock and mineral separates followed by careful analysis and screening of the data for any residual problems.

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