

Labile trace elements in basaltic achondrites: Can they distinguish between meteorites from the Moon, Mars, and V-type asteroids?

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Abstract—We report data for 14 mainly labile trace elements (Ag, Au, Bi, Cd, Cs, Ga, In, Rb, Sb, Se, Te, Tl, U, and Zn) in eight whole-rock lunar meteorites (Asuka [A-] 881757, Dar al Gani [DaG] 262, Elephant Moraine [EET] 87521, Queen Alexandra Range [QUE] 93069, QUE 94269, QUE 94281, Yamato [Y-] 793169, and Y-981031), and Martian meteorite (DaG 476) and incorporate these into a comparative study of basaltic meteorites from the Moon, Mars, and V-type asteroids. Multivariate cluster analysis of data for these elements in 14 lunar, 13 Martian, and 34 howardite, eucrite, and diogenite (HED) meteorites demonstrate that materials from these three parents are distinguishable using these markers of late, low-temperature episodes. This distinguishability is essentially as complete as that based on markers of high-temperature igneous processes. Concentrations of these elements in 14 lunar meteorites are essentially lognormally distributed and generally more homogeneous than in Martian and HED meteorites. Mean siderophile and labile element concentrations in the 14 lunar meteorites indicate the presence of a CI-equivalent micrometeorite admixture of 2.6%. When only feldspathic samples are considered, our data show a slightly higher value of 3.4% consistent with an increasing micrometeorite content in regolith samples of higher maturity. Concentrations of labile elements in the 8 feldspathic samples hint at the presence of a fractionated highly labile element component, possibly volcanic in origin, at a level comparable to the micrometeorite component. Apparently, the process(es) that contributed to establishing lunar meteorite siderophile and labile trace element contents occurred in a system open to highly labile element transport.

INTRODUCTION

Samples from asteroids, the Moon and Mars were ejected with greater than escape velocities from their parents by explosive impacts and arrived to Earth as meteorites. Lunar meteorites are, of course, unambiguously identified with that source by properties that they share with materials returned by manned (Apollo) and unmanned (Luna) missions. These meteorites are particularly important since they provide a more random lunar sampling of source regions, hence are more representative of the Moon's surface (cf. Warren 1994; Korotev et al. 2003a). Prior studies of such basaltic achondrites demonstrate that, in particular, meteorites of lunar and Martian origin and those identified as having come from V-type asteroid(s), the HED meteorites, can be distinguished as to source from each other because of differences in high-temperature genesis. Their properties indicate that each of the three derive from sources that accreted and differentiated under different physical and chemical conditions from

potentially different starting materials (Richter et al. 2006, and references therein) with independent chronologies (Halliday and Klein 2006, and references therein). These properties include mineralogic, petrologic, bulk refractory elemental and isotopic parameters (Richter et al. 2006, and references therein). Whether, and to what extent, materials from these three sources are distinguishable solely using markers of late, low-temperature episodes has not been studied to any appreciable extent.

Prior studies demonstrate that elements of moderate-to-high thermal lability (i.e., volatility and/or mobility) have proven particularly useful in providing low-temperature planetary thermal histories (Palme et al. 1988; Lipschutz and Woolum 1988, and references therein). In previous studies of six lunar meteorites, the feldspathic regolith breccias Allan Hills (ALH) A81005, MacAlpine Hills (MAC) 88105, Yamato (Y-) 791197, Y-82192, Y-86032, and the anorthositic-bearing basaltic regolith breccia Y-793274, we reported data obtained by radiochemical neutron activation analysis

(RNAA) for a suite of 15 trace elements. This suite was comprised of siderophilic Co, Au and Sb; labile Se, Te, Bi, Ag, In, Tl, Zn, and Cd; and lithophilic Rb, Cs, Ga, and U (Verkouteren et al. 1983; Kaczaral et al. 1986; Dennison et al. 1987; Wang and Lipschutz 1990; Lindstrom et al. 1991a, 1991b). Data from these studies permitted the quantitation of the amount of admixed meteoritic debris (reported in % CI equivalents) and KREEP (potassium, rare earth elements and phosphorous) in each sample, and the identification of Y-791197 as a sink for labile elements mobilized via lunar volcanism and subsequently recondensed in a thermodynamically open system.

The determination and interpretation of labile trace element composition presents a unique set of challenges, particularly for labile element-depleted achondrites. RNAA has long been the analytical standard for labile element determination. During the past decade, inductively coupled plasma mass spectrometry (ICPMS) has developed into a very accurate and precise geochemical analytical tool (cf. Lipschutz et al. 2007). The analytical figures of merit of ICPMS rival that of RNAA and, because its high sample throughput capabilities facilitate the measurement of multiple blanks and application of time-dependent external drift corrections, ICPMS offers the possibility that it can supplant RNAA as the technique of choice for quantifying our suite of highly labile trace elements (cf. Wolf et al. 2005), even in lunar samples.

Interpreting and modeling labile element composition has its own unique set of challenges due to these elements' low concentrations and relatively complex geochemical behaviors. To address issues of interpretation, a variety of chemometric methodologies have been adopted in which meteorite sample sets, representative of their source parents, are treated using multivariate-based statistical analysis with labile element concentrations providing average low-temperature thermal history information (Wolf and Lipschutz 1995; Wolf and Lipschutz 1998). The fundamental basis of this approach is that complex higher-dimensionality data are best suited for multivariate analysis (Aitchison 1986). Model-independent data exploratory methodologies have been developed that can reveal hidden compositional trends and provide insight and facilitate meaningful interpretation (Lavine 1992). Potentially, a promising method for this application is multivariate cluster analysis as demonstrated for Martian and eucrite meteorites in Wang et al. (1998).

In this paper, we report our progress in these areas most critical to incorporating labile element composition of lunar meteorites into lunar geochemistry: their determination and interpretation. We report new RNAA data for 14 trace elements in seven lunar meteorites and one Martian meteorite. We report ICPMS data for an additional lunar meteorite to supplement our existing RNAA data. With this more substantial lunar meteorite database in hand, we can now perform a study involving cluster analysis of basaltic

meteorites for which complete analytical data exist from three parents: the Moon [$n = 14$]; Mars [$n = 13$, including Dar al Gani (DaG) 476 reported here]; and meteorites generally accepted as deriving from 4 Vesta or other V-type asteroids (cf. McCord et al. 1970; Consolmagno and Drake 1977; Lipschutz et al. 1989), the howardites [$n = 5$], eucrites [$n = 20$], and diogenites [$n = 9$], collectively called the HED achondrites [$n = 34$]. Our larger lunar meteorite database allows us to examine potential relationships between labile element concentrations and lunar rock type and address whether lunar meteorite labile element concentrations are consistent with an unfractionated micrometeorite component, either CI (Baedeker et al. 1973; Krähenbühl et al. 1973), CM (Wasson et al. 1975), or a combination of these and some other fractionated source (Haskin and Warren 1991). As this would involve relatively late, low-temperature processes, the issue is uniquely suited to interpretation on the basis of labile element concentrations.

EXPERIMENTAL

We received whole rock samples of six lunar meteorites: Asuka (A-) 881757,83 (98.0 mg), Elephant Moraine (EET) 87521,37 (42.7 mg), Queen Alexandra Range (QUE) 93069,17 (51.6 mg), QUE 94269,11 (72.3 mg), QUE 94281,20 (68.1 mg), and Yamato (Y-) 793169,81 (20.1 mg) from Dr. D. W. Mittlefehldt who had previously analyzed them using instrumental neutron activation analysis (INAA). Lunar meteorite DaG 262 (102.0 mg) was provided by Prof. H. Palme of the Universität zu Köln. Martian meteorite DaG 476 (98.1 mg) was provided by Dr. J. Zipfel of the Max-Planck-Institut für Chemie. In all cases samples were splits taken from homogenized powders prepared from larger interior samples selected as representative of bulk meteorite material. After receipt the samples were transferred from their SiO₂ glass INAA irradiation tubes, re-encapsulated, and irradiated at the University of Missouri Research Reactor (UMRR) with the appropriate RNAA flux monitors for 144–480 h (depending on sample size) at a flux of 8.0×10^{13} n·cm⁻²·s⁻¹. The RNAA technique in this study was that routinely employed in the Purdue laboratory, with monitor preparation, chemical processing and data reduction procedures unchanged from those described previously (Wolf 1993). Chemical yields for elemental separations obtained for samples and monitors for all elements exceeded 30% in all cases and in most cases ranged from 50–95%. As demonstrated in prior consortium studies, neutron fluences during re-irradiation of INAA-irradiated samples for RNAA were sufficiently higher than those for INAA so that prior activity could be ignored (Wang and Lipschutz 1990).

We received lunar sample Y-981031 as part of the consortium led by Dr. H. Kojima of the Japanese National Institute of Polar Research (NIPR). This 103.8 mg whole-rock powdered sample was dissolved and analyzed by

quadrupole ICPMS (PQ Excell, ThermoElectron Corporation, Waltham, MA, USA) using methodology and instrumentation described in Wolf et al. (2005). For this work, calibration involved the standard addition method with internal standardization. Determination of our suite of 14 trace elements by ICPMS involved three separate analytical groups; each designed to optimize standard addition spike concentrations and elemental solution stability for the 14 elements. Group A included higher concentration elements Ga and Zn; group B included HCl-stabilized trace elements Sb, Te and Au; and group C included HNO₃-stabilized trace elements Se, Rb, Ag, Cd, In, Cs, Tl, Bi, and U. Each standard addition procedure included triplicate analysis of a dissolution blank, one unspiked sample and two samples spiked with differing amounts of each analyte element. Approximately 5-gram aliquots of the dissolution blank and sample were spiked with sufficient internal standard spike containing 2 $\mu\text{g}\cdot\text{mL}^{-1}$ Rh and Re to establish nominal concentrations of 10 $\text{ng}\cdot\text{mL}^{-1}$ of these internal standard elements. For analysis of Group A elements, two 5 g aliquots of dissolved sample were spiked with 50 and 100 ng of the analyte elements Ga, and Zn, respectively. For analysis of group B and C elements, two 5 g aliquots of dissolved sample were spiked with 0.25 and 0.5 ng of the corresponding analyte elements, respectively. Element concentrations were calculated using the ICPMS data acquisition and control software (PlasmaLab Issue 1.06, ThermoElectron Corporation, Waltham, MA, USA).

Several standard chemometric and statistical data analysis methods were used to evaluate our data. All analyses were performed using JMP statistical software (JMP Version 6.0, SAS Institute, Inc., Cary, NC, USA). The functional forms of the trace element concentration data for our 14 lunar meteorites were examined using two forms of univariate goodness-of-fit tests: the Shapiro-Wilk (Shapiro and Wilk 1965) and the Kolmogorov-Smirnov tests (Neter et al. 1988). The Shapiro-Wilk test is designed specifically to test whether the concentrations of a given element in a sample set derive from a population in which concentrations are normally distributed. The Kolmogorov-Smirnov goodness-of-fit test was used to determine whether the distributions of individual trace elements could be specified as lognormal. This test is based on the cumulative sample function and is independent of the underlying cumulative distribution function being examined.

We used Ward's method of hierarchical multivariate cluster analysis with data normalization (Ward 1963) as a chemometric data exploratory method to objectively identify and group lunar, Martian and HED meteorites possessing similar trace element compositions. The technique is based on the principle that the distance between a pair of samples in multivariate measurement space is inversely related to their degree of similarity. The technique is agglomerative, in that every sample initially represents a cluster, and the process of

joining clusters continues until all samples are joined in a single cluster. Ward's method is based on the minimization of the within-cluster sum of squares over all partitions obtained when every possible combination of cluster pairs are joined. Cluster analysis is similar to the approach of performing partial analysis of data via bivariate scatter diagrams (cf. Fig. 10, Korotev et al. 2006) and grouping samples on the basis of a priori classification. However, cluster analysis differs fundamentally from this approach in that the clustering of samples is objectively based on compositional similarities alone and not a priori classification. More importantly, cluster analysis facilitates analysis of multivariate data such as labile trace element composition.

Finally, we used standard multivariate analysis of variance (MANOVA) and univariate one-way analysis of variance (ANOVA) to detect and identify systematic compositional differences between lunar, Martian, and HED meteorites and within the lunar meteorites (Aitchison 1986; Neter et al. 1988). For both MANOVA and ANOVA we tested the null-hypothesis of the following form: meteorites with different a priori classifications are indistinguishable on the basis of trace element composition. We interpreted observed differences on the basis of the cosmochemical properties of each element.

RESULTS

In Table 1 we list data for 14 trace elements in the eight whole-rock lunar meteorites and Martian meteorite analyzed in this study with our prior data for six whole-rock lunar meteorites. Where indicated, uncertainties are $\pm 1\sigma$ and upper limits are $<2\sigma$. All are RNAA data with the exception of Y-981031 which are ICPMS results. Our meteorite sample set includes 6 feldspathic regolith breccias (ALH A81005, DaG 262, MAC 88105, QUE 93069, QUE 94269, and Y-791197), 2 feldspathic fragmental breccias (Y-82192 and Y-86032), three anorthosite-bearing basaltic regolith breccias (QUE 94281, Y-793274, and Y-981031), polymict basaltic fragmental breccia (EET 87521), one gabbro (A-881757), and one unbrecciated basalt (Y-793169) as listed in Righter (2007). The lone Martian meteorite analyzed for this study, DaG 476, is classified as an olivine-orthopyroxene shergottite (Meyer 2003). The new trace element data bring our lunar and Martian meteorite database to 14 and 13 specimens, respectively.

We assess the quality of our new RNAA data by initially examining results of replicate analyses of aliquots of Allende (CV3) Standard Reference Meteorite and then by comparing our new lunar meteorite data with published results from prior analyses. Results of replicate RNAA of Allende by the two analysts contributing data to this study are listed in Table 2. These results are compared with the definitive Purdue mean for RNAA of the trace elements in this standard meteorite. Results for all but Se are within 1σ of the group mean: the

Table 1. Trace element concentrations in 14 whole-rock samples of lunar meteorites and one whole-rock Martian meteorite.

Meteorite	Type ^a	Au	Sb	Se	Te	Bi	Ag	In	Tl	Zn	Cd	Rb	Cs	Ga	U	Ref. ^c
ALHA81005,10	F	2.82	1.6	200	9.2 ± 1.8	0.73	2.4 ± 0.5	1.5 ± 0.2	2.27	4.68	19	340	19	2.53	110	A
MAC 88105,41	F	2.41	6.2	160	12.3	2.07	4	1.6 ± 0.1	7.25	7.76	14.8	625	91	3.26	107	B
Y-791197,88	F	24	4.9	300	70	12	16	10	6.4	58	190	1300	89	11	240	C
Y-82192,52C1	F	12.1	4.2	338	22.3	3.9	3.7	2.6 ± 0.2	5	4.63	20.2	230	19.9	2.86	50.6	D
Y-86032,75AM	F	1.86	4.3	283	29.3	2.99	5.6	3	4.8	5.32	31.7	248	10.5	3.89	59.1	E
Y-793274,62C	F/B	0.98	1.6	419	22	4.1	7.9	1.51	0.86	11.7	33.9	1280	72.6	4.91	283	F
A-881757,83	B	0.29	3.1	280	4.8 ± 1.9	1.6 ± 0.2	64 ^b	1.9 ± 0.3	9.62	1.44	12.3	662	36.6	3.26	124	G
DaG 262	F	5.51	13	312	32	7.55	32.7	10.8	2.22	32.9	176	960	53.9	2.94	239	G
EET 87521,37	F/B	0.26	6.6	119	186	5.8	2.17	0.56	1.5	2.06	9.8	590	27.3	5.15	255	G
QUE 93069,17	F	5.34	17	310	123	2.7	8.32	4.4	2.09	12.4	55.4	652	28.9	3.49	131	G
QUE 94269,11	F	4.31	4.1	289	21.7	1.0 ± 0.3	10.9	2.31	3.2	16.2	67.2	709	115	3.34	189	G
QUE 94281,20	F/B	1.8	1.9	184	8.5 ± 2.1	1.47	7.12	2.85	7.85	12.6	23.7	931	166	4.7	314	G
Y-793169,81	B	0.17	3.1	611	<13.2	2.7 ± 0.4	7.66	0.72 ± 0.26	4.26	8.59	4.6 ± 0.3	1120	60.8	5.14	217	G
Y-981031,71	F/B	9.1	10	217	42.6	0.4 ± 0.1	27.3	3.5 ± 0.9	18	10.9	48.7	1040	98.3	4.06	279	H
DaG 476	OOS	1.05	1.8	476	<8.8	2.97	19.4	13.5	10.8	89.3	37.1	435	9.70	8.64	18.5	G

^aType: F = lunar feldspathic; B = lunar basaltic; F/B = lunar polymict breccia containing both feldspathic and basaltic components; OOS = Martian olivine-orthopyroxene shergottite.^bDatum anomalously high and disregarded.^cReferences: A = RNAA by Verkooren et al. (1983); B = RNAA by Lindstrom et al. (1991a); C = geometric mean of duplicate RNAA by Kaczaral et al. (1986); D = RNAA by Dennison et al. (1987); E = RNAA by Wang and Lipschutz (1990); F = RNAA by Lindstrom et al. (1991b); G = RNAA, this study; H = ICPMS analysis, this study.

Table 2. Concentrations of 14 trace elements in Allende Standard Reference Meteorite as determined by replicate RNAA and in USGS standard rock BCR-1 as determined by ICPMS.

Element	Conc.	Allende MSW RNAA ^{a,b}	Allende SFW RNAA ^{a,c}	Allende Purdue mean RNAA ^{a,d}	BCR-1 ICPMS	BCR-1 literature value ^e
Ag	ppb	102 ± 4 (5)	102 ± 6 (11)	99.9 ± 8.5 (93)	31	27
Au	ppb	146 ± 5 (6)	139 ± 12 (12)	144.0 ± 9.6 (52)	<2	0.66
Bi	ppb	49.0 ± 1.5 (6)	47.3 ± 5.4 (7)	48.6 ± 3.2 (69)	50	47
Cd	ppb	494 ± 11 (14)	487 ± 58 (14)	504 ± 50 (99)	124	130
Cs	ppb	87.3 ± 3.0 (14)	78.3 ± 5.2 (15)	85.7 ± 5.8 (103)	950	960
Ga	ppm	6.00 ± 0.19 (11)	6.22 ± 0.49 (14)	6.15 ± 0.48 (90)	23.7	22
In	ppb	28.7 ± 1.2 (9)	29.2 ± 3.7 (11)	30.2 ± 3.3 (87)	95	92
Rb	ppm	1.08 ± 0.04 (15)	1.06 ± 0.10 (11)	1.10 ± 0.12 (95)	48.0	47.2
Sb	ppb	84.6 ± 4.5 (6)	87.1 ± 20.4 (9)	84 ± 13 (57)	600	620
Se	ppb	8.88 ± 0.17 (11)	7.34 ± 0.43 (14)	8.47 ± 0.98 (88)	93	88
Te	ppb	980 ± 49 (13)	969 ± 73 (14)	1000 ± 86 (109)	5.6	4.9
Tl	ppb	60.7 ± 3.3 (10)	61.2 ± 4.7 (9)	60.8 ± 4.4 (100)	320	300
U	ppb	—	—	—	1700	1750
Zn	ppm	117 ± 6 (15)	116 ± 4 (9)	116.7 ± 6.8 (105)	131	129.5

^aUncertainties are sample standard deviations. Numbers in parentheses are number of replicates.^bWang and Lipschutz (1990).^cWolf (1993).^dCalculated from Wolf (1993) and references therein.^eGovindaraju (1994).

SFW Se mean differs by 1.07σ . The new ICPMS methodology was assessed by analysis of USGS standard basalt BCR-1 to provide comparative data from a basaltic specimen. These results are also listed in Table 2. Wolf's (2005) ICPMS data for these elements in Murchison (CM2) show excellent agreement with accepted literature values.

Some published INAA data exist for ten elements in seven of the lunar meteorites and the DaG 476 Martian meteorite with which the RNAA and ICPMS results in this paper can be compared. No prior data exist for four of the most labile trace elements of our suite (Te, Bi, In, and Tl) or for QUE 94269. The most numerous prior data are for Au, Ga and U obtained primarily by INAA: many of the results reported for Au, Sb, Se, Ag, Cs and are only upper limits, thus are not as useful. Published results for A-881757 include: Zn, Ga, Se, Rb, Ag, Sb, Cs, Au, and U (Koeberl et al. 1993); Au, Zn and Ga (Warren and Kallemeyn 1993); and Au and U (Korotev et al. 2003b). For DaG 262 data exist for: Au, Sb, Cs, Ga, and U (Warren et al. 2005); Au, Se, Zn, Cs, Ga and U (Bischoff et al. 1998); and Au, Sb, Cs and U (Korotev et al. 2003a). Published results for EET 87521 include: Au and U (Korotev et al. 2003b); Au, Zn, Cd, Rb, Cs, Ga, and U (Warren and Kallemeyn 1989, 1991). Results for QUE 93069 include: Au, Sb, Se, Ag, Zn, Rb, Cs and U (Koeberl et al. 1996); and Au and U (Korotev et al. 1996). For QUE 94281 data exist for Ga and U (Dreibus et al. 1996); Au, Ga, and U (Arai and Warren 1999); Au and U (Korotev et al. 2003b); and Au, Cs, and U (Jolliff et al. 1998). Results for Y-981031 include Au, Cs, and U (Korotev et al. 2003b). Published data for Y-793169 include: Sb, Au, Se, Ag, Zn, Rb, Cs, Ga, and U (Koeberl et al. 1993); and Au, Zn, Cd, and Ga (Warren and Kallemeyn 1993). Prior data for Martian meteorite DaG 476 include: Au, Se, Zn, Ga, and U by INAA (Zipfel et al. 2000); and Zn, Rb, Cs, Ga, and U by ICPMS (Barrat et al. 2001).

The 14 trace elements reported in this study are inherently variable in basaltic meteorites due to heterogeneity in 10–100 mg samples and can result in real compositional differences greater than a factor of two between separate aliquots from a single specimen, even when analyzed by a single analyst using a single method. We assess RNAA and ICPMS results separately. The general application of our RNAA method for this suite of 14 trace elements in basaltic meteorites has been assessed previously (Verkouteren et al. 1983; Kaczaral et al. 1986; Dennison et al. 1987; Wang and Lipschutz 1990; Lindstrom et al. 1991a, 1991b). We assess potential systematic differences in our new data by comparison with previously published data derived from aliquots of the same meteorite. We calculate the percent relative difference between our data and the grand mean of all previously published analyses on an element-by-element basis. Due to the high level of variability intrinsic to these trace elements, we note here only differences greater than $\pm 50\%$ relative to the literature grand mean values as evidence of a possible

discrepancy. For the ten trace elements for which prior data exist, only Sb, Ag, Cd, and Rb exceed this threshold differing by -70% , -72% , $+50\%$, and -52% , respectively. While this level of bias is not evident for any of these elements in our Allende results (Table 2) this effect could be concentration or matrix dependent. Absolute Sb, Ag, and Cd concentrations are extremely low in lunar and Martian meteorites, <10 ppb in most cases. Absolute Rb concentrations are also low in lunar and Martian meteorites, typically ranging from ~ 100 – 1000 ppb. Limits of detection and quantitation for all of these elements are lower for RNAA than for INAA due to the decreased background associated with chemical separation. Several previously reported INAA results are near method limits of detection. The level of differences observed for these elements is also consistent with previously observed variabilities due to heterogeneity in basaltic meteorites.

Our ICPMS results agree reasonably well with the few INAA data available for Y-981031 (Korotev et al. 2003b). ICPMS results for U are lower by 7% relative to INAA data. Our ICPMS Cs value, 98.3 ppb, is consistent with the INAA upper limit of <150 ppb. The ICPMS Au result, 9.1 ppb, is higher than the INAA datum, 2 ppb. One might argue that the ICPMS Au might reflect uncertainty in background subtraction or contamination, but the absolute difference is consistent with heterogeneous distribution of this trace siderophile in lunar samples. A more complete comparison can be made from results of ICPMS analysis of USGS standard rock BCR-1 (Table 2). Concentrations of our 14-element suite in this study in BCR-1 determined by ICPMS, generally agree within $\pm 10\%$ relative to recommended values (Govindaraju 1994). The exceptions, Ag and Te, differ relative to recommended values by $+15\%$ and $+14\%$, respectively. Au was below ICPMS method limit of quantitation (<2 ppb). We accept our ICPMS results with caution, but without further question.

With respect to terrestrial alteration processes and their potential impact on trace element concentrations, all seven lunar meteorites and the one Martian meteorite analyzed in this work are finds and may potentially be impacted by terrestrial weathering or contamination. Two samples, DaG 262, and DaG 476 are Libyan, warm-desert finds while the remaining are Antarctic finds. The Libyan lunar and Martian specimens have both been described as moderately weathered (Bischoff et al. 1998 and Zipfel et al. 2000, respectively). Antarctic finds A-881757 and Y-793169 are unclassified with respect to weathering. All remaining Antarctic finds are weathering classification A/B or better with the exception of QUE 94281 which is Be. For our suite of elements, weathering should be highly reflected in concentrations of soluble lithophiles Cs, Rb, and U. We observe no evidence of discordance among our data for these elements that suggest compromise by terrestrial weathering subsequent to fall. Within our suite of 14 lunar meteorites, the data are internally consistent with no obvious outliers except possibly for a

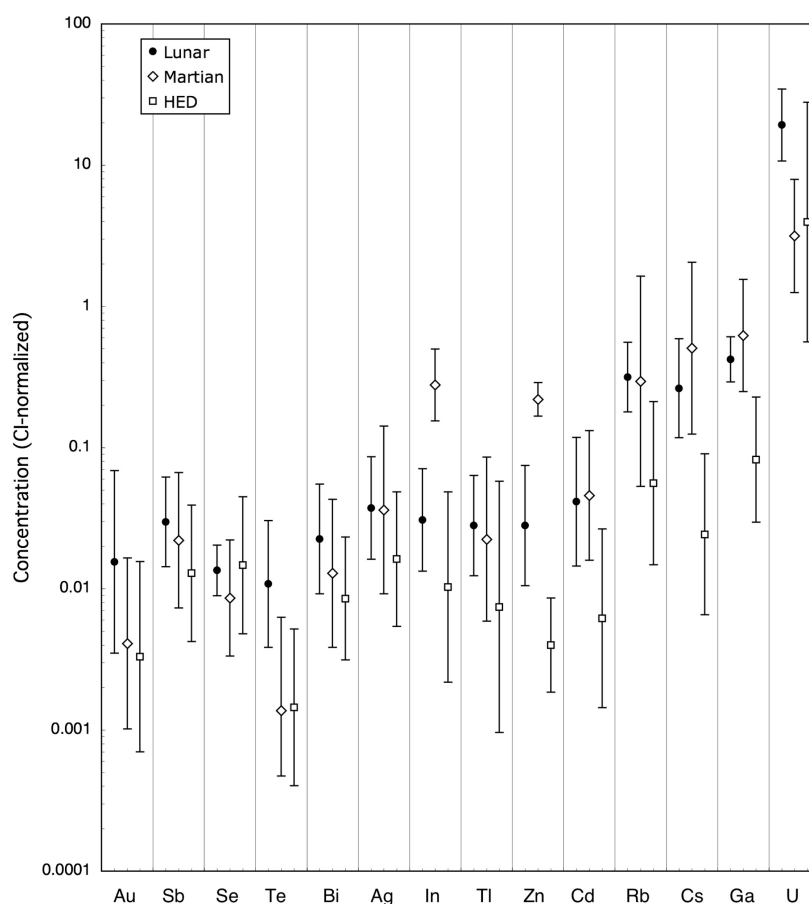


Fig. 1. Mean and standard deviation concentrations (CI-normalized) of 14 trace elements (Au, Sb, Se, Te, Bi, Ag, In, Tl, Zn, Cd, Rb, Cs, Ga, and U) in 14 lunar, 13 Martian, and 37 HED meteorites. Elements are ordered from the left as two siderophiles, eight thermally labile elements and four lithophiles ordered by increasing mobility or ease of vaporization and loss during simulated thermal metamorphism (cf. Lipschutz and Woolum 1988). Despite the variabilities in the three basaltic meteorite sample sets, some systematic differences are apparent. Nonetheless, none of the elements here can serve alone as a marker for lunar, Martian, or V-type asteroid origin.

single RNAA determination of Ag in A-881757. Because this sample was powder and the one anomalously high datum does not correlate with the other labile elements in the sample, it hints at possible contamination. We therefore, omit this sample's Ag data from our univariate calculations. Because multivariate analyses require a complete data set for each sample, we chose to omit Ag from these analyses rather than omitting sample A-881757.

DISCUSSION

Comparison of Lunar with Martian and HED Meteorites

For visualization, we plot in Fig. 1 means and standard deviations CI-normalized concentrations (Lodders 2003) of our trace element suite for 14 lunar meteorites (Verkouteren et al. 1983; Kaczaral et al. 1986; Dennison et al. 1987; Wang and Lipschutz 1990; Lindstrom et al. 1991a, 1991b; this study), along with those for 13 Martian (Biswas et al. 1980; Smith et al. 1984; Laul et al. 1986; Paul 1988; Treiman et al.

1994; Mittlefehldt et al. 1997; Wang et al. 1998; and this study) and 37 HED (Paul 1988; and Paul and Lipschutz 1990) meteorites. We list elements as: siderophile (Au and Sb), labile (Se, Te, Bi, Ag, In, Tl, Zn, and Cd), and lithophile (Rb, Cs, Ga, and U). We order labile elements by increasing mobility, i.e., ease of vaporization and loss from average chondritic matter heated at 1000 °C in a low pressure (initially 10^{-5} atm) H_2 environment. Results of Shapiro-Wilk and Kolmogorov-Smirnov goodness-of-fit tests both indicate that the distributions of the concentrations of the 14 trace elements in our lunar meteorite sample set are consistent with the hypothesis of lognormality. Thus, throughout this discussion we use log-transformed concentrations for cluster analysis and model-dependent hypothesis tests (MANOVA and ANOVA). Calculations of mean and standard deviation CI- and CM-equivalent material in lunar samples are performed using log-transformed concentrations followed by reverse-transformation. All means and standard deviations are geometric unless otherwise stated.

Given the variability and overlap of labile element

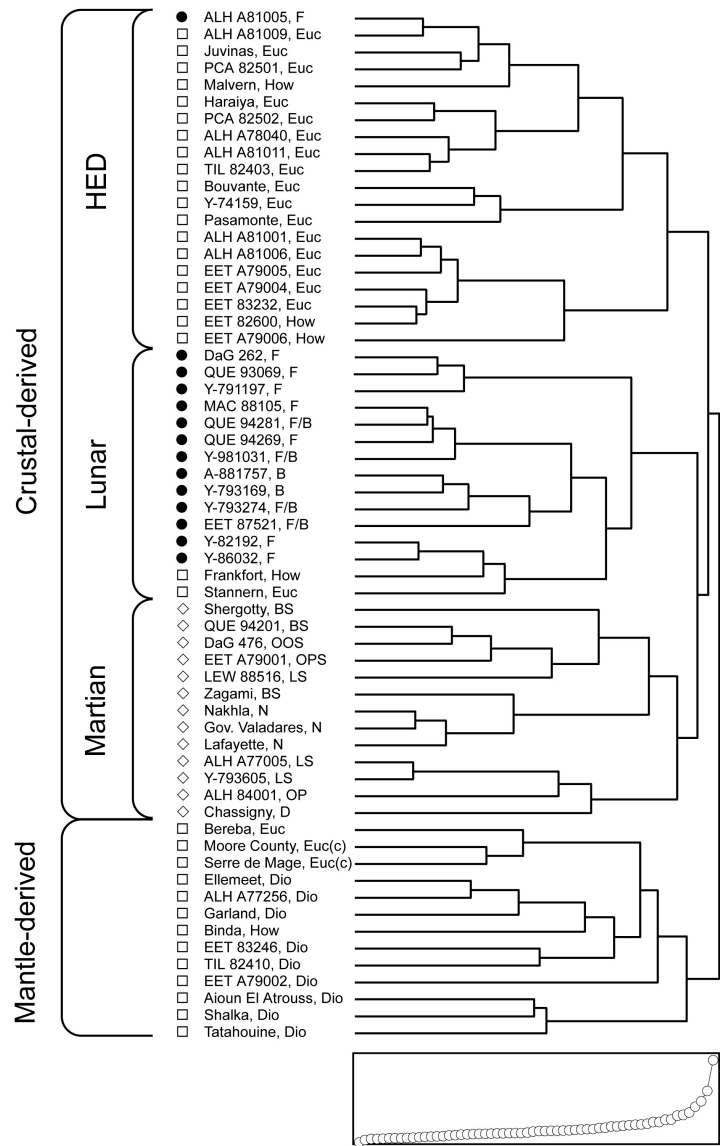


Fig. 2. Cluster analysis using 13 trace elements of our suite for 14 lunar meteorites (●), 13 Martian meteorites (◇), and 34 HED meteorites (□). Lunar meteorites are sub-classified by rock type: feldspathic (F), basaltic (B), and polymict breccias containing both feldspathic and basaltic components (F/B). Martian meteorites are sub-classified as: basaltic shergottite (BS), dunite (D), ilherzolitic shergottite (LS), clinopyroxenite nakhilite (N), olivine-orthopyroxene shergottite (OOS), olivine-phyric shergottite (OPS), or orthopyroxenite (OP). Each HED meteorite is listed as a howardite (How), eucrite (Euc), or diogenite (Dio). With few exceptions cluster analysis reveals compositional separation first on the basis as to crust or mantle origin (independent of parent body) and second as to lunar, Martian, and HED parent body origin.

concentrations in lunar, Martian, and HED meteorites, in contrast to compositional parameters reflecting high-temperature planetary processes, obviously no individual labile element clearly distinguishes between them (Fig. 1). Considering these elements' low concentrations, diverse geochemical behaviors, and their different responses to thermal events during planetary formation and evolution, this is not surprising. Cluster analysis of our trace elements suite, in principle, offers the possibility that these three sample sets can be distinguished by markers of late, low-temperature episodes.

Figure 2 depicts cluster analysis results for 61 basaltic achondrites from the Moon [$n = 14$], Mars [$n = 13$], and the HED parent body [$n = 37$] using log-transformed concentrations of the 13 elements in our suite. Standard output for cluster analysis takes the form of a dendrogram and scree plot. From left to right, clustering starts with the two compositionally most similar meteorites. Individual meteorites or clusters are subsequently grouped as determined by within-cluster sum-of-squares analysis. The length of each horizontal line segment (branch) joining cluster pairs indicates the degree of compositional similarity between the

pair, with, e.g., shorter lengths representing greater compositional similarity. Additional samples or clusters are subsequently joined in order of greatest compositional similarity. The scree plot (named after this plot's resemblance to a geologic detritic cone, given below the dendrogram) graphically indicates the relative distance in compositional space that was bridged to join the corresponding clusters. A sharp increase in slope in the scree plot represents the linking of two relatively dissimilar clusters. These increases can suggest natural cutting points for data evaluation. While criteria for interpreting the graphic output are somewhat subjective, the technique provides valuable insight by identifying compositionally similar specimens.

From the dendrogram (Fig. 2) two observations are noteworthy: (1) a parent body-independent distinction between crustal-derived and mantle-derived meteorites; and (2) distinction between meteorites from the Moon, Mars, and HED parent body. The greatest compositional difference in our 61 achondrites occurs between 48 exclusively crustal-derived samples of meteorites from all three parent bodies and 13 primarily mantle-derived meteorites from the HED parent body. This 13-sample cluster includes all 9 diogenites, one howardite (Binda) and three eucrites (Bereba and the two cumulate eucrites in our database, Moore County and Serre de Mage). Clustering of mantle-derived samples reflects their systematic depletion in all 13 elements studied, most significantly in the 4 lithophiles (Rb, Cs, Ga, and U) and the 6 most labile elements (Bi, Ag, In, Tl, Zn, and Cd). Whether this depletion is due solely to fractionation by elemental incompatibility during magmatism or a combination of incompatibility and volatility is uncertain.

The second noteworthy feature in Fig. 2 is compositional differences involving our 61 basaltic achondrites distinguishing among meteorites from the lunar, Martian, and HED parents. The 48 crustal-derived meteorites split into three separate clusters containing 20, 15, and 13 members: these correspond to eucrites/howardites, lunar meteorites and Martian meteorites. Only three of these 48 samples are misclassified as to source. The lunar feldspathic regolith breccia ALHA81005 is grouped in the cluster of 16 eucrites and 3 howardites. Stannern eucrite and Frankfort howardite are included with the other 13 lunar meteorites. Misclassifications within these two clusters indicate a modest compositional similarity between lunar and HED meteorites. The misclassifications are readily reconciled from the siderophile and labile element concentrations of these samples. The grouping of ALH A81005 with the eucrites/howardites reflect this particular lunar meteorite's low labile element concentrations relative to other lunar meteorites (Table 1). Similarly, grouping Stannern and Frankfort with lunar meteorites is readily accounted for: these meteorites contain the highest or near highest levels of Au, Sb, Ag and Zn of all other HED meteorites (Paul and Lipschutz 1990; and Paul 1988). It is notable that these two HED meteorites are grouped with lunar meteorites Y-82192 and Y-86032, two

terrestrially-paired feldspathic samples (Wang and Lipschutz 1990; Lindstrom et al. 1991) that are fragmental breccias and hence, do not contain materials diagnostic of the lunar surface (cf. Korotev et al. 2006). The associations of lunar, Martian, and HED meteorites by multivariate cluster analysis (Fig. 2) closely match those deduced from markers of *high-temperature* processes. Of course, here these three sample sets are distinguishable on the basis of moderately to highly labile elements, thus reflecting distinguishable average *low-temperature* thermal histories. Finally, it is notable that both Martian and lunar meteorites cluster only nominally according to traditional (SNC) sub-types and lunar rock types, respectively. This observation indicates that despite their significantly different mineralogies, samples from each respective parent body share related late, low-temperature thermal histories.

The distinguishability of lunar, Martian, and HED meteorites is further supported by hypothesis testing. Using MANOVA we find that for our suite of 13 trace elements, our 14 lunar, 13 Martian, and 34 HED meteorites possess significantly different compositions ($p < 0.0001$). We obtain identical results when comparisons are limited to subsets of 4 lithophile elements Rb, Cs, Ga, and U ($p < 0.0001$), 2 siderophile elements Au and Sb ($p < 0.0001$), 7 labile elements Se, Te, Bi, In, Tl, Zn, and Cd ($p < 0.0001$), and even the 2 most labile elements Zn and Cd ($p < 0.0001$). Using ANOVA we find that the mean concentrations of a majority of elements considered here differ significantly in the lunar, Martian, and HED suites: 13 of our 14 elements (i.e., all but Se) differ at the 95% confidence level. Mean concentrations of all elements except Se are higher in the lunar than in the HED sample set (Fig. 1) indicating a relatively labile element rich component in the former, not found in HEDs. Comparison of lunar with Martian meteorites however, does not indicate a similar systematic difference: mean concentrations of Au, Sb, Se, Te, Bi, Ag, Tl, and Rb are higher in the former; and In, Zn, Cd, Cs, and Ga are higher in the latter. Within both the Martian and HED meteorite sample sets, no clear trend is apparent involving siderophile and labile element composition and corresponding cosmochemical affinity, volatility, or mobility. HED meteorites are extremely labile element depleted and do not reflect any late, low-temperature signature in our elements. That the Martian meteorites are on average richest in high-lability elements associated with volcanic activity (i.e., In, Zn, and Cd) may hint at a volcanic component: however, Martian meteorites certainly experienced other complex low-temperature processes. Our data do not unambiguously reveal specific differences between the Martian and HED meteorites other than their different average thermal histories.

Lunar Meteorite Labile Element Composition

Clearly, the most distinguishing feature of the concentrations of the 10 siderophile and labile elements in our

Table 3. Mean and standard deviation % CI-normalized concentrations of 14 trace elements in lunar meteorites. Summary overall means and standard deviations are calculated for each element for our 8 feldspathic samples, 4 polymict breccias containing both feldspathic and basaltic components, and 2 basaltic samples. Arithmetic means and pooled standard deviations are calculated for each type of lunar meteorite for elements grouped as siderophiles (Au and Sb), labiles (Se, Te, Bi, Ag, In, Tl, Zn, and Cd), and for all 10 siderophiles and labiles overall. Elements are listed in order of increasing lability within each element group. We note that the CI-normalization scheme is used here for comparison purposes only.

Elements	Feldspathic [n = 8]		Feldspathic/basaltic [n = 4]		Basaltic [n = 2]		Overall lunar [n = 14]	
	\bar{x}	s	\bar{x}	s	\bar{x}	s	\bar{x}	s
Au	3.5	2.4	1.0	4.4	0.2	1.5	1.6	4.4
Sb	3.6	2.1	2.5	2.5	2.0	1.0	3.0	2.1
Overall: siderophiles	3.5	2.2	1.7	3.4	1.1	1.2	2.3	3.3
Se	1.4	1.3	1.1	1.7	2.1	1.7	1.3	1.5
Te	1.2	2.3	1.5	3.7	0.3	2.0	1.1	2.8
Bi	2.6	2.6	1.8	3.3	1.9	1.4	2.2	2.5
Ag	3.7	2.4	3.8	2.8	3.8 ^a	–	3.7	2.3
In	4.4	2.1	2.2	2.3	1.5	2.0	3.1	2.3
Tl	2.6	1.7	2.6	4.1	4.5	1.8	2.8	2.3
Zn	3.8	2.6	2.4	2.4	1.1	3.5	2.8	2.7
Cd	6.9	2.7	3.7	2.0	1.1	2.0	4.4	2.9
Overall: labiles	3.3	2.2	2.4	2.8	2.0	2.1	2.7	2.4
Overall: siderophiles and labiles	3.4	2.2	2.2	2.9	1.9	1.9	2.6	2.6

^an = 1.

14-meteorite lunar sample set is the resemblance of their interelement distribution to that seen in carbonaceous chondrites (Fig. 1). Originally observed in data from lunar rocks and soils retrieved through the course of Apollo missions, this feature is considered diagnostic of a micrometeorite component admixed in the lunar regolith, initially expressed as % CI equivalents on the basis of siderophile and labile element concentrations (Baedeker et al. 1973; Krähenbühl et al. 1973) and subsequently, argued to be more consistent with CM chondrite on the basis of siderophile/Ir ratios (Wasson et al. 1975). Interpretation of concentrations of highly labile elements in this context has proven difficult. All of the highly labile elements discussed here have been identified as potentially “vapor-mobilized” based on their high lability and overall low abundances in lunar rocks relative to soils and glasses and can thus be fractionated (Haskin and Warren 1991). Our data can be used to evaluate and potentially deconvolute these two putative processes in lunar meteorites.

Quantitation of the micrometeorite component in our lunar meteorites is dependent on several interrelated factors: the composition of the micrometeorite source; the effect of labile element-dependent fractionation (including labile element loss on the lunar surface or gain from intralunar sources); and sample selection. Because labile element fractionation is element- and potentially sample-dependent, identifying systematic differences between different lunar meteorite rock types might clarify the impact of this process on our data. Our approach is statistically based, assuming that our lunar meteorites [n = 14] are random samples from the Moon’s surface and reflect a time-integrated average of these

processes. We initially utilize therefore, our entire lunar meteorite sample set (F, F/B, and B rock types) in our analyses and examine systematic variations, first between the lunar sub-types and second within our suite of labile elements across each sub-type.

In Table 3 we show summary statistics for % CI-normalized concentrations for each lunar meteorite rock type and overall, for each element and elements grouped as siderophiles (Au and Sb), labiles (Se, Te, Bi, Ag, In, Tl, Zn, and Cd), and an overall mean for all 10 siderophile and labile elements. While the data are highly variable, we observe two trends: (1) mean elemental concentrations tend to increase as B < F/B < F for each element grouping; and (2) mean element concentrations tend to increase subtly with lability, specifically in the F samples. This second trend is much less evident in the F/B and B rock types. MANOVA indicates that when compared on the basis of our 2 siderophile elements Au and Sb, the compositional difference between lunar rock types B, F/B, and F is moderately significant ($p = 0.05$). However, when MANOVA is based on concentrations of 7 labile or 4 lithophile elements, we find no evidence of a compositional difference ($p = 0.62$, and $p = 0.26$, respectively). ANOVA reveals that only 2 of our 14 trace elements, Au and Cd, differ at the >95% confidence level: U differs at the 94% confidence level. Qualitatively, the concentrations of the lithophiles increase as F < B < F/B, consistent with the F/B samples possessing a higher incompatible element component. However, this trend is not statistically significant on a multivariate basis.

Based on these observations and tests, we limit our investigation of the composition of the micrometeorite

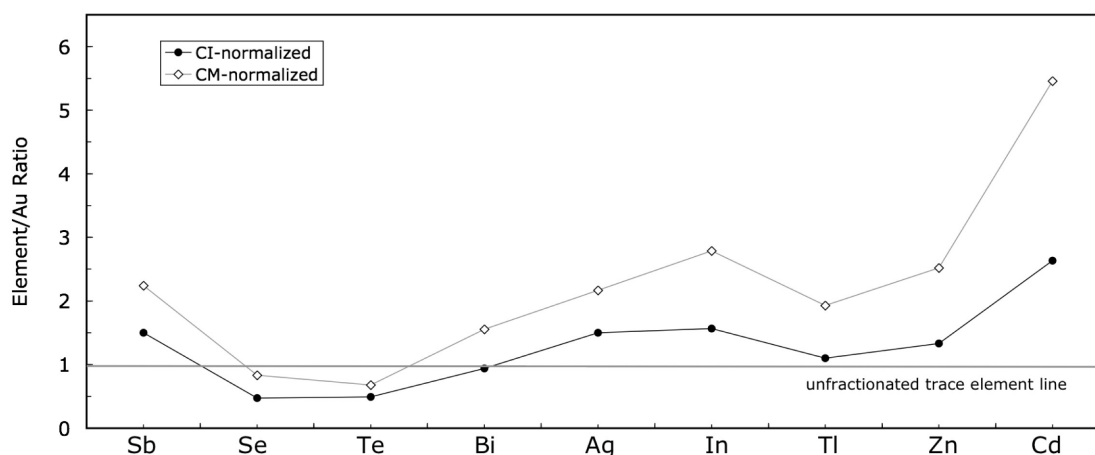


Fig. 3. Mean siderophile and labile element/Au ratios for 14 lunar meteorites normalized to CI chondrite (Lodders 2003) and CM chondrite (Wasson and Kallemeyn 1988). Data are plotted in order of increasing element lability. Patterns resulting from both normalization schemes are essentially flat with a slight trend toward higher relative concentrations of higher lability elements. While neither of these results unambiguously identifies the composition of the micrometeorite component as CI or CM, both hint at an additional fractionated highly labile element component.

component by examining samples containing significant siderophile concentrations: our 8 feldspathic samples. In Fig. 3 we plot the mean element/Au ratio for 9 siderophile and labile elements, normalized to concentrations in CI and CM chondrites and examine the inter-element trends. We use Au as an internal standard because it is our least labile siderophile and has been used as a marker for the micrometeorite component (cf. Korotev et al. 2006). Figure 3 shows that both the CI- and CM-normalized patterns are essentially flat with a slight trend toward enrichment of higher lability elements, particularly Ag, In, Tl, Zn, and Cd. The CM-normalized pattern shows a slightly more pronounced trend toward enrichment of higher lability elements due to CM chondrites' higher mean Au and lower mean labile element concentrations relative to CI chondrites. The mean element/Au ratios for CI- and CM-normalized are 1.3 ± 0.7 and 2.2 ± 1.4 , respectively. While neither of these results unambiguously identifies the composition of the micrometeorite component, both hint at subtle fractionation of the highly labile elements. When the ratio of the mean of the most labile (Ag, In, Tl, Zn, and Cd) to mean of the remaining labile (Sb, Se, Te, and Bi) elements is calculated, the ratios of these CI- and CM-normalized results are 1.9 ± 0.8 and 2.2 ± 1.6 , respectively. While highly uncertain, the observation of increasing element concentration with lability is consistent with results from studies performed on Apollo-16 soils demonstrating that concentrations of the highly labile elements In, Zn, and Cd, increase with decreasing grain size; indicating a particle surface correlation resulting from open system deposition of these elements during regolith evolution (Boynton et al. 1976). More recently, Zn and Cd isotope fractionation studies of lunar glass and soil samples have identified isotopic fractionations consistent with deposition of these labile elements associated with volcanism (Moynier et al. 2006;

Schediwy et al. 2006). As mature regolith contains greater proportion of glass fragments and smaller particles (cf. Korotev et al. 2006) increasing amounts of both micrometeorite and fractionated components might be expected in these meteorite samples. In this light, the compositions of three feldspathic regolith breccias are particularly noteworthy: DaG 262, QUE 93069, and Y-791197. Table 1 clearly shows that these samples contain the highest or near highest concentrations of siderophile and labile elements, particularly Au, In, Zn, and Cd. High concentrations of siderophile elements in DaG 262 and QUE 93069 have been noted previously (Bischoff et al. 1998; Korotev et al. 2003a). The regolith breccia Y-791197 is compositionally very heterogeneous and its enrichment in highly labile elements was attributed to the condensation of volcanic exhalations in it (Kaczaral et al. 1986): in this respect it is similar in trace element composition to Apollo 66095 ("Rusty Rock"). That cluster analysis (Fig. 2) groups these specimens, indicates similar compositions and hints at similar low-temperature thermal histories. Possibly Y-791197, DaG 262, and QUE 93069 contain a fractionated labile element component, volcanic in origin, to slightly varying degrees. However, while our analyses hint at a fractionated highly labile element component added to an unfractionated micrometeorite component, the variability of these data preclude unambiguous determination of the precise composition of either.

CONCLUSIONS

Multivariate cluster analysis proves to be an effective technique for identifying groups of meteorites possessing relatively similar labile trace element compositions and hence, similar *low-temperature* thermal histories. When

applied to 14 lunar, 13 Martian, and 34 HED meteorites, cluster analysis demonstrates that concentrations of 13 mainly labile trace elements distinguish these groups as they are defined by compositional parameters that reflect their different *high-temperature* histories. Cluster analysis further demonstrates nominal labile trace element compositional similarities that exist within HED sub-types (howardites, eucrites and diogenites). Martian and lunar meteorites only nominally cluster as to sub-types suggesting that despite their different high-temperature thermal histories, samples from each respective parent body share related late, low-temperature thermal histories.

Concentrations of 14 trace elements—siderophiles (Au and Sb), labile elements (Se, Te, Bi, Ag, In, Tl, Zn, and Cd) and lithophiles (Rb, Cs, Ga, and U)—in whole-rock samples of 14 lunar meteorites exhibit lognormal distributions without significant outliers. New ICPMS data for Y-981031 accord with RNAA data for these elements indicating that ICPMS is a valid technique for determining labile trace elements even at low achondritic levels. Mean siderophile and labile element concentrations in the 14 lunar meteorites indicate the presence of a carbonaceous chondrite-like micrometeorite admixture of 2.6% CI-equivalents. When only feldspathic samples are considered, our data show a slightly higher value of 3.4% CI-equivalents consistent with an increasing micrometeorite content in regolith samples of higher maturity (Korotev et al. 2006). Further examination of labile element concentrations in the 8 feldspathic samples suggests the presence of an additional fractionated highly labile element component, possibly volcanic in origin, at a level comparable to the micrometeorite component. The high variability intrinsic to this suite of labile elements precludes unambiguous determination of the micrometeorite composition, or the precise ratio of fractionated to unfractionated components in our samples. However, these observations indicate that the processes contributing to lunar meteorite siderophile and labile trace element content occur in a system open to labile element transport.

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