GEOCHEMICAL INVESTIGATION OF FIVE LUNAR METEORITES:
IMPLICATIONS FOR THE COMPOSITION, ORIGIN
AND EVOLUTION OF THE LUNAR CRUST

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Abstract: The total number of lunar meteorites is now eleven, and the number of distinct finds now stands at eight. We report new data for the bulk compositions of five lunar meteorites, including Y-793274, a unique regolith breccia composed of 2/3 mare material of VLT (very-low-Ti) affinity and 1/3 highlands material compositionally similar to Apollo 16 regolith. Mixing-deconvolution of the bulk-rock composition shows that for any reasonable assumed highlands-component composition, the TiO$_2$ content of the mare component must be <1.2 wt%. A unique clast from the Y-791197 highlands regolith breccia is also probably of VLT-mare affinity, based on its REE pattern, Sc/Sm, Mn/Sm, and Eu/Al ratios, and the low Ti/(Ti+Cr) ratio of its pyroxene. VLT affinities have previously been inferred for numerous mare clasts observed in thin section studies of highlands meteorites, and three other recently-discovered lunar meteorites have either VLT or borderline VLT/"normal" mare compositions. Apparently, the abundance of VLT-basaltic matter in the lunar crust is greater than previously supposed; and the fundamental dichotomy of lunar magmatism into distinct nonmare and mare styles may have been less abrupt than commonly envisaged. Alternatively, the prevalence of VLT varieties of mare basalt among the lunar meteorites might be a sign of source-crater pairing. However, it seems probable that at least three, and more likely five or more, separate craters are represented; and at least one of the sources is probably on the farside. In terms of major-element composition, the four highlands regolith breccia meteorites resemble the Apollo 16 regolith. The Si content and Ca/Al ratio of the upper lunar crust appear to be monotonously close to 210 mg/g and 0.53 (molar), respectively. However, concentrations of incompatible elements, including K, Th, and U, are far lower in the highlands meteorites than in regolith samples from the central nearside. This trend implies that inferred "lower limits" on the bulk-Moon content of U (and associated refractory lithophile elements) should be relaxed.

1. Introduction

The Moon is a geochemically differentiated body, of great interest as an object for comparison to the Earth. Although the Moon differentiated in its own distinctive style, the lunar crust is comparable in mass and volume to the Earth's crust. In some ways, the lunar crust is also comparable in complexity. Lunar crustal rocks range from anorthosites to magnesian troctolites, norites, and REE-rich "KREEP" basalts, with smaller proportions of other rock types such as magnesian dunite and peridotite, granite, etc. Lunar basalts exhibit remarkable diversity of isotopic ratios (e. g.,
Systematic geochemical differences have been observed even among superficially similar rock types from different Apollo sites (Warren et al., 1983a; Shervais and Taylor, 1986; Laul, 1986). Orbital spectrometry has been used to gage the composition of roughly 20% of the lunar surface (e.g., Metzger et al., 1977, 1979), but only for a few elements, only with moderate sensitivity and precision, and only within two narrow, near-equatorial bands. Direct sampling has been limited to a total of 9 sites. Of these, only 6 sites (the Apollo sites) supplied "rock" samples. The unmanned Soviet "Luna" landers only acquired a few tens of grams of soil, with little or no material >1 cm across. To make matters worse, all of these sites are clustered over a small portion of the Moon's central nearside hemisphere (Fig. 1). Of course, this situation is mitigated by the "gardened" nature of the lunar crust, owing to its early exposure to intense meteoritic bombardment. However, in the absence of more comprehensive sampling, models for the composition, petrology, and origin of the lunar crust might be seriously flawed.

In recent years, this need for augmented sampling of the Moon has been met, by the roundabout method of harvesting meteorites from Antarctica. The total number of Antarctic lunar meteorites now stands at 11, although this tally drops to 8 if corrected for obvious pairings, namely MacAlpine Hills 88104 with MAC-88105, and Yamato-82192 with Y-82193 and Y-86032 (Lindstrom, 1990). Henceforward in this paper, these meteorites will be designated (except where specific subsamples are characterized) as MAC88104/5 and Y-82192/86032; and the "total" number of lunar meteorites will be considered to be eight (Table 1).

We report here new compositional data, mainly from INAA, for five lunar meteorites. Most importantly, we report comprehensive bulk-compositional data for Yamato-793274, the first lunar meteorite found to consist of subequal proportions of highlands and mare materials; and for an extraordinary clast separated from Y-791197. Also, for bulk-rock samples of four lunar meteorites (ALHA81005, EET-87521, Y-791197, and Y-82192/86032), we augment our previous studies (Kallemeyn and Warren, 1983; Warren and Kallemeyn, 1986, 1987; Warren et al., 1989) with further data for a few key, previously-undetermined elements. For completeness,
Table 1. Summary of known lunar meteorites, as of mid-1990.

<table>
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<th>Meteorite</th>
<th>Mass (g)</th>
<th>Pairing</th>
<th>Rock type</th>
<th>Season collected</th>
<th>Year publicized as lunar</th>
<th>Cons.-style as lunar allocations?</th>
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<td>Y8X</td>
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<td></td>
<td></td>
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<td>'90</td>
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<td>'90</td>
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</tr>
</tbody>
</table>

References: mostly LINDSTROM's (1990) review, but the characterizations of Asuka-31 and Y-793169 are entirely due to YANAI and KOJIMA (1990).

1 Cons.-style: consortium-style, 2 RB: regolith breccia.

3 Y-82192/6032 is so immature, it is arguably best classified as an "ordinary" fragmental breccia.

4 Asuka-31 is a tentative name (YANAI and KOJIMA, 1990).
we include recently acquired bulk-rock data for MAC88104/5, described in greater detail by Warren and Kallemeyn (1990). We interpret these new data, and all of the available data for lunar meteorites collectively, as constraints on the nature of the lunar crust, its origin and evolution, and on the bulk composition and origin of the Moon.

A note regarding nomenclature: In the rather confusing nomenclature of mare basalts, the category "low-Ti mare basalt" is generally understood in a narrow sense, which excludes very-low-Ti (VLT) as well as high-Ti varieties. The low-Ti/VLT distinction is important in connection with several of the lunar meteorites. According to the authoritative BVSP (1981), the low-Ti/VLT division falls at 1.5 wt% TiO₂. However, among Apollo and Luna mare rock samples (excluding the mare volcanic glasses, many of which are ultramafic), wt% TiO₂ is almost never <1.8 wt%, except among the Apollo 17 and Luna 24 VLT mare basalts (BVSP, 1981; Taylor, 1982), which occasionally have TiO₂ as high as 1.44 wt% (Ryder and Marvin, 1978). Yanai (1990) shows that the Asuka-31 lunar meteorite resembles previously known VLT mare basalts more closely than it does low-Ti mare basalts, even though the TiO₂ content of Asuka-31 is 1.66 wt%.

2. Analytical Procedures

All samples were first studied by INAA, using a procedure modified from that of Kallemeyn et al. (1989). Bulk-rock samples were powdered in an agate mortar prior to INAA (MAC88105, 41 is an aliquot of a widely-distributed powder prepared from a 20-g sample of matrix by Eugene Jarosewich). After INAA, these samples were split into two aliquots, one 5-10× larger than the other. The smaller aliquot was used for determination of major elements and Ti with an electron probe, after conversion of the aliquot into a fused bead (we call this technique microprobe fused bead analysis, or MFBA). The larger aliquot was used for RNAA, following our standard procedure (Warren et al., 1986), to determine siderophile elements Au, Ge, Ir, Ni, Os and Re, along with volatile elements Cd and Zn. The clast from Y-791197 was studied by INAA, but not by RNAA or MFBA.

3. Evidence of Lunar Origin

The evidence for lunar origin of these meteorites is overwhelming. In most cases, an overall mineralogical/compositional similarity to lunar highlands-anorthositic materials was obvious to the initial describers (e.g., Yanai and Kojima, 1985; Score et al., 1989). Low-Ti mare basalts are not so easily distinguished from other types of meteorites (especially eucrites), but diagnostic ratios such as Fe/Mn, Ga/Al, Na/Ca, Co/Cr, etc. (e.g., Warren and Kallemeyn, 1989; Delaney, 1989), can be used to rule out kinship with any known nonlunar type of meteorite. Most useful are the oxygen isotopic ratios, which for lunar meteorites (and no other type of differentiated meteorites except the obviously nonlunar aubrites) fall along the Earth-Moon fractionation line (e.g., Mayeda et al., 1987; Score et al., 1989). Some lunar meteorites bear clear signs of a regolith history at an actively gardened, high solar-wind (i.e.,
inner solar system) location. These samples have high contents of noble gases (Fig. 2). The most mature regolith breccias are also distinctively rich in swirly-brown, agglutinitic glasses (e.g., Warren et al., 1983b). Detailed textural features also help to distinguish mare cumulates from cumulate eucrites: exsolution in cumulate-eucritic pyroxenes is generally far more extensive than in EET87521 or Asuka-31

![Fig. 2. Regolith maturity of the five highlands (or largely highlands) lunar meteorites, as gauged by their contents of solar-wind-implanted noble gases, exemplified by $^{36}\text{Ar}$. Data are from Bogard and Johnson (1983), Bischoff et al. (1987), Takaoka (1987), Eugster (1990a, b), and Takaoka and Yoshida (1990). Shaded bar at right indicates range observed for regolith breccias from Apollo 16 (Eugster, 1989).](image)

![Fig. 3. The HPF clast, prior to chipping to extract it from Y-791197, 121. Width of view ~ 2.0 mm.](image)
pyroxenes of similar composition (Takeda, 1979; Warren and Kallemeyn, 1989; Yanai, 1990). In addition, the “4π” cosmic-ray exposure ages of most lunar meteorites (e.g., Eugster, 1990a; Nishizumi et al., 1990) are extraordinarily young compared to most nonlunar meteorites.

4. The Y-791197 “HPF” Clast

This clast rich in hedenbergite, plagioclase, and fayalite was discovered during a thin-section study by Takeda et al. (1986). It consists of iron-rich pyroxene (zoned from Ca-rich pigeonite or pyroxferroite to hedenbergite), minor plagioclase, fayalite, and small amounts of olivine.

Table 2. Concentrations of 40 elements in bulk-rock and

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<th>Mass</th>
<th>Mg/mg/g</th>
<th>Si/mg/g</th>
<th>Al/mg/g</th>
<th>Ca/mg/g</th>
<th>Sc/mg/g</th>
<th>Ti/mg/g</th>
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<td>2.34</td>
<td>35</td>
<td>141</td>
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<td>232</td>
<td>110</td>
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<td>220</td>
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<td>87</td>
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<tr>
<th>Element</th>
<th>Nd/µg/g</th>
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<th>Eu/µg/g</th>
<th>Tb/µg/g</th>
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</table>

* Normal uncertainty limits (% relative) for 70% confidence.
**1 Uncertainty limits 1.1–2.0 x greater than normal.
**2 Uncertainty limits 2.1–4.0 x greater than normal.
**3 Uncertainty limits 4.1–6.0 x greater than normal.
**4 MAC88105, 41 is an aliquot from a powder sample produced using 20 g of matrix by E.
and a dark mesostasis with fayalite, silica and ilmenite. The clast was only studied by TAKEDA et al. (1986) as a small, oblong outcrop, about 1 mm across. With important assistance from Dr. TAKEDA, the NIPR loaned us a 169-mg sample of Y-791197, on which the remainder of the HPF clast was found, outcropping over an arrowhead-shaped area 1.5 mm in maximum dimension (Fig. 3). The clast was identified based on the unusual brownish color of its mafic silicates. It consisted of roughly 1/2 crystalline mafic silicate (brown grains), 1/3 plagioclase (white, commonly lath-shaped grains up to 0.7 mm long), plus a variety of much finer-grained, dark brown to black materials. After carefully cleaning the matrix away from the clast, we estimated that the resultant sample “Y-791197,121c” was at least 99%-pure clast.

clast (c) samples from lunar meteorites.

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<th>Mn</th>
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<th>Co</th>
<th>Ni</th>
<th>Zn</th>
<th>Ga</th>
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</tr>
<tr>
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<td>2.62</td>
<td>0.29</td>
<td>50*1</td>
<td>0.64</td>
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<td>0.22</td>
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<td>62.4</td>
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<td>0.135</td>
<td>0.73</td>
<td>0.078*1</td>
<td>450</td>
<td>8.2</td>
<td>6.4</td>
<td>0.6*3</td>
<td>0.28</td>
<td>0.079*2</td>
<td>64.3</td>
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<td>0.058*2</td>
<td>62.7</td>
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<td>0.42</td>
<td>0.057*1</td>
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<td>0.047*2</td>
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<td>2.2</td>
<td>0.169</td>
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<td>0.47</td>
<td>0.061*1</td>
<td>580</td>
<td>6.7</td>
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<td>0.53</td>
<td>1.24*2</td>
<td>&lt;1.5</td>
<td>— —</td>
<td>&lt;40 — &lt;1.9 &lt;1.3</td>
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<tr>
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<td>4</td>
<td>5</td>
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<td>10</td>
<td>8</td>
<td>5</td>
<td>6</td>
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</tbody>
</table>

JAROSEWICH.
Based on their thin-section observations, Takeda et al. (1986) inferred that the HPF clast might be either an extremely evolved extension of the ferroan anorthositic suite (FAS) of pristine nonmare rocks; or else a poorly representative sample of a low-Ti mare basalt. Our data (Table 2) tend to suggest that the clast is a piece of VLT mare basalt. Although REE contents are about 2× higher in the HPF clast than in average Apollo-17 VLT mare basalt, the pattern (heavy REE enrichment, faint negative Eu anomaly) is similar (Fig. 4). Compared to average Luna 24 VLT mare basalt, the HPF has similar light REE concentrations, but higher heavy REE. The HPF has lower light REE by a factor of 2 than an average low-Ti mare basalt. The incompatible element contents of the HPF are also superficially similar to some of the most REE-rich FAS samples (which have light REE at roughly 3–10×CI (Warren et al., 1991a)). However, considering the great disparity in plagioclase An ratio and olivine and pyroxene 100Mg/(Mg+Fe) ratio (hereafter abbreviated as mg) between the HPF and even the most REE-rich FAS sample, along with the observation (Takeda et al., 1986) that much of the HPF consists of fine-grained mesostasis material (FAS rocks, where not thoroughly brecciated, invariably have accumulate textures), if the HPF were an evolved variety of FAS material, it should probably have much higher REE, especially light REE, than any "normal" FAS sample. For a lunar rock, the HPF clast has extraordinarily high Sc/Sm and Mn/Sm and Mn/Sm ratios (Fig. 5). Several FAS rocks have comparable Sc/Sm and Mn/Sm, but among non-FAS nonmare rocks only the magnesian dunite 72417 (Laul and Schmitt, 1975), which is obviously not a close relative of HPF, has higher Sc/Sm. The HPF's Sc/Sm and Mn/Sm ratios place it squarely along the positive correlation trend of mare basalts, at the VLT (high-Sc/Sm) end of the trend. Similarly,
the Eu/Al ratio of the HPF is low, especially for a low-mg lunar rock (Fig. 6). This composition is equally consistent with an extremely evolved variety of FAS material, or a relatively low-mg variety of VLT mare basalt (on a plot such as Fig. 6, crystal
fractionation tends to drive residual melts upward and to the left). If the mare basalt interpretation is correct, the low Eu/Al is another indication of VLT affinity. Rigidly interpreted, the bulk-rock TiO$_2$ content, 2.0 wt%, is high enough to classify the sample as a low-Ti mare basalt. However, the analyzed sample was too small to be highly representative. Like our REE data (Figs. 4-6), the data of TAKEDA et al. (1986) for minor elements in HPF pyroxenes favor a VLT affinity (Fig. 7).

Conceivably, the two petrogenetic models mentioned by TAKEDA et al. (1986) are both partly correct. The FAS is probably a mass of flotation cumulates from a primordial magma ocean (or magmasphere) (e.g., WARREN, 1986). If so, the mare basalts were probably produced by remelting of basal cumulates (or more complex "paracumulates" (WARREN, 1989)) formed relatively late in the crystallization of the same magmasphere. The most extremely evolved FAS materials are thus expected to show some similarities to mare basalts. However, the same model implies that the mare basalt source regions are probably stratified with Ti content generally increasing upwards (e.g., BVSP, 1981; TAYLOR, 1982; SHERVAIS and TAYLOR, 1986), and VLT-source materials buried hundreds of kilometers below the surface. Our preferred interpretation is that the HPF clast is a poorly representative piece of a low-Ti, and most likely VLT, mare basalt.

5. Y-793274: A Regolith Breccia of Mixed Mare/Highlands Derivation

Our data for Y-793274 (Table 2) show typical lunar values for diagnostic ratios such as Fe/Mn (72.4), Ga/Al (5.2 x 10$^{-5}$), etc., and thus confirm the inference of YANAI and KOJIMA (1987) that this meteorite is of lunar origin. Like ALHA81005
Fig. 8. Bulk-rock Al$_2$O$_3$ vs. TiO$_2$ for lowest-Ti mare basalts and highlands regolith samples, including the four mainly-highlands lunar meteorites (shown as open diamonds) and Y-793274. Mixing-deconvolution implies that the mare component of Y-793274 must be a VLT variety. The data of Yanai (1990) for Asuka-31 and Y-793169 indicate that they are also uncommonly low in Ti.

Fig. 9. Bulk-rock Al$_2$O$_3$ vs. Sm for VLT mare basalts and highlands regolith samples, including the four mainly-highlands lunar meteorites (shown as open diamonds) and Y-793274. Mixing deconvolution implies that the highlands component of Y-793274 probably has a Sm content comparable to average Apollo 16 regolith. Regarding EET87521, see text.
and Y-791197, Y-793274 is a relatively mature regolith breccia, as indicated by petrographic observations (e.g., Koeberl et al., 1991; Takeda et al., 1990), and by its contents of solar-wind derived noble gases (Fig. 2). Geochemically, however, Y-793274 is unique. It was originally described as an anorthositic (highlands) regolith breccia (Yanai and Kojima, 1987), but a substantial component of mare material is also present (Takeda et al., 1990; Koeberl et al., 1991). Like many of the regolith samples from Apollo 15 and Apollo 17, Y-793274 apparently formed as a mixture of roughly equal proportions of mare and highlands material. A plot of Al$_2$O$_3$ vs. TiO$_2$ (Fig. 8) shows that for any reasonable model regarding the Al$_2$O$_3$ content of the mare component and the Al$_2$O$_3$ and TiO$_2$ contents of the highlands component, the TiO$_2$ content of the mare component must be <1.2 wt%—which implies that the mare component is clearly VLT. In this sense, Y-793274 is distinct from the two purely-igneous mare meteorites, Asuka-31 and Y-793169. Although our sample has an Al$_2$O$_3$ content of 17.4 wt%, samples analyzed elsewhere (Fukukoka, 1990; Koeberl et al., 1991; Lindstrom et al., 1991) are generally less aluminous, implying that the overall mare: highlands ratio in Y-793274 is probably roughly 2:1.

A plot of Al$_2$O$_3$ vs. Sm (Fig. 9) helps to constrain the nature of the highlands component. Assuming that the Sm content of the mare component is roughly similar to the Sm contents of other VLT materials, mixing-deconvolution implies that the Sm content of the highlands component is probably similar to that of average Apollo 16 regolith. Note that EET87521 cannot be regarded as a pure VLT-mare sample for this comparison, because EET87521 is polymict. In fact, if the same mixing-deconvolution model is applied to EET87521, its highlands component is implied to be unusually Sm-rich (comparable to Apollo 16 regolith). This result might not be significant, however, because although EET87521 is polymict, it is nearly pure VLT-mare matter, and its highlands component might not be representative of the general crustal environment where it formed. In contrast, Y-793274 is a regolith breccia, and thus its highlands component is presumably a blend of many different rocks from its crustal source region. Judging from our data and the results of Fukukoka (1990), Koeberl et al. (1991), and Lindstrom et al. (1991), the mare component in Y-793274 has average mg$_2$ intermediate between Luna 24 and Apollo 17 VLT basalts (which have average mg$_2$ ~ 39 and 54, respectively). This moderate mg$_2$ ratio inferred for the mare component of Y-793274 further distinguishes Y-793274 from EET87521 (mg$_2$ = 39; Table 2).

Thus, the geochemical uniqueness of Y-793274 is not merely superficial. It seems almost certain that Y-793274 is not paired with any other lunar meteorite. At least, it is not paired in the conventional sense of pairing, which would imply that the "pair" once formed a single fragment that became disrupted by interaction with the Earth's atmosphere and/or solid surface. Given the nondescript shock history of Y-793274 (Takeda et al., 1990; Koeberl et al., 1991), it could hardly have left the Moon as part of any single fragment more than a few meters in diameter—a scale at which lunar regolith tends to be nearly homogeneous.
6. Discussion

6.1. How many lunar meteorite launch sites?

The data for trapped solar-wind-derived noble gases (Fig. 2) indicate that all of the five highlands-derived lunar meteorites (including Y-793274) contain at least minor regolith components. Y-791197 and ALHA81005, and to a lesser extent Y-793274, are mature regolith samples, while at the other extreme Y-82192/86032 is so immature as to be arguably better classified as an ordinary fragmental breccia (e.g., Bischoff et al., 1987). MAC88105 is intermediate. Regolith breccias are products of thorough gardening of the upper few meters of the Moon, and consequently even a few tens of milligrams of sample may be representative for salient compositional characteristics. In cases where two mature regolith samples show a pronounced contrast vs. one another, the contrast cannot be dismissed as a fluke of the sampling process. If the lunar meteorites were polymict breccias of the “ordinary” fragmental type, or impact-melt breccias, or worse still, if they were pristine rocks, we would have far less assurance for the meteorites being compositionally representative of their source regions.

The great value of the lunar meteorites stems from the possibility that they represent otherwise unsampled regions of the Moon, but the number of such regions represented depends on the possibility that each of the lunar meteorites represents a separate source crater. Two meteorites might reach the Earth at separate locations, and still be products of a single lunar source crater. Even assuming that the proportion of ejecta from lunar craters eventually reaching the Earth is independent of crater size (in all likelihood there is a correlation with crater size), the mass of material sent to Earth should scale as the crater volume, i.e., in proportion to $D^3$, where $D$ is the crater diameter. The number of lunar craters in any given size range scales roughly as $D^{-1.8}$ (BVSP, 1981). Thus, of the total number of lunar meteorites of any given mass, the proportion derived from lunar craters of any given diameter should, by this model, scale as $D^{1.2}$. In practical terms, this implies that if we could find many thousands of lunar meteorites, the total proportion derived from craters $y$ km in diameter is likely to be only $0.43 \times$ the proportion derived from craters with $D=2y$, and only $0.18 \times$ the proportion derived from craters with $D=4y$. Of course, with only 8 distinct lunar meteorites in the total population, quantum effects greatly distort these yield ratios. There is no reason to conclude, a priori, that the number of separate source craters must be less than eight. Nevertheless, the potential for source-crater pairing among the eight lunar meteorites is clearly high.

Cosmic-ray exposure age constraints are available for all four of the preponderantly-nonmare lunar meteorites (Table 3). The relevant age is the sum of the $4\pi$ (Moon–Earth) exposure age plus the “terrestrial” age. The observation that 3 of the 4 dated lunar meteorites have “blast-off” ages $\ll 1$ Ma is hardly surprising. Arnold (1965) and Wetherill (1968) used Monte Carlo computer modeling to simulate accretion to the Earth of objects ejected from the Moon into heliocentric orbits. The Moon’s escape velocity is $1.65 \times$ the velocity for escape from the Earth’s gravity at the lunar distance, so most of ejecta that escapes from the Moon probably passes through a heliocentric phase before eventually colliding with the Earth. Even ignoring
objects traveling from the Moon to the Earth without first going heliocentric, Wetherill (1968) finds that 52% of the Moon–Earth transits take less than 1 Ma; and 1/4 of the transits take less than 0.1 Ma. From the data in Table 3, Y-82192/86032 left the Moon 10 Ma before any of the others. MAC88104/5 probably left the Moon after Y-791197 and ALHA81005, but the data are not conclusive. As argued by Warren and Kallemeyn (1986), ALHA81005 is compositionally distinctive, with a far higher $mg$ than Y-791197, or any other lunar meteorite (Table 2), a circumstance that tends to favor the hypothesis that ALHA81005 represents a separate source crater. In summary, it seems likely that four separate source craters are represented by the four preponderantly-nonnmare lunar meteorites, but the number of separate sources could easily be as small as three (Y-82192/86032 plus two others), and might be just two (Y-82192/86032 plus one other).

The number of separate source craters represented by the other four (mare or mainly mare) lunar meteorites is still conjectural. The coincidence that all four contain VLT, or borderline low-Ti/VLT, types of mare materials is clearly suggestive of source-crater pairing (or even sensu stricto pairing). However, studies of the lunar highlands meteorites have revealed many clasts of apparent mare derivation, and most of these clasts have modes, textures, and pyroxene Ti/(Ti+Cr) ratios indicative of VLT or at least low-Ti affinities (Treiman and Drake, 1983; Lindstrom et al., 1986; Goodrich and Keil, 1987). Given the evidence against source-crater pairing of all of the highlands meteorites, the preponderance of VLT and borderline low-Ti varieties among these samples is evidently not merely a manifestation of pairing. The lunar meteorites, collectively, seem to be revealing that VLT (and borderline low-Ti) varieties of mare basalt constitute a higher proportion of the total mare component of the lunar crust than suggested by the limited sampling achieved at the nine Apollo/Luna sites.

6.2. Have we sampled the far side?

The answer to this question probably is yes, we have. Statistical theory for binomial distributions (in which there are only two mutually exclusive data types or “outcomes”) implies that the relevant probability function is:

$$P = \frac{n!}{(n-x)!x!}p^x(1-p)^{n-x},$$

where $n$ is the total number of outcomes (or “trials”), $x$ is the number of outcomes of a given type (the number of random “successes”), and $p$ is the probability for
Geochemical Investigation of Five Lunar Meteorites

“success” in each individual trial (each trial is assumed to have the same \( p \)). Assuming that we have lunar highlands meteorites from four separate craters (i.e., ignoring the partly-mare Y-793274), eq. (1) implies that the probability for all four to be from the nearside is \( 0.5^4 = 0.0625 \). Even if we assume that only two separate source sites are represented, the probability for both to be from the nearside is only \( 0.5^2 = 0.25 \). These calculations assume that the probability \( (p) \) for each lunar highlands meteorite to be from the nearside is 50%. The cratering rate on the farside is probably the same, to within <1%, as on the nearside (BANDERMANN and Singer, 1973). Maria cover ~32% of the nearside, but only ~2% of the farside (e.g., Taylor, 1982), which implies that an otherwise random lunar location known to be essentially pure highlands is \( \approx 1.4 \times \) more likely to occur on the farside than on the nearside. However, the meteorites in question all contain a tiny proportion of mare clasts (Treiman and Drake, 1983; Lindstrom et al., 1986; Goodrich and Keil, 1987). The levels of incompatible trace elements for all four of these meteorites are far below those found among central nearside regolith samples, and far below the levels found over much of the nearside by orbital spectrometry (Metzger et al., 1977). Thus, the probability for each individual Th-poor meteorite to be from the nearside is \( \ll 50\% \). On balance, 50% seems a conservatively high value to assume for \( p \), and the probability \( P \) that none of the four discrete lunar highlands meteorites came from the farside is very low.

As the Moon orbits, meteoroids have to “catch up” with the trailing hemisphere. As a result, Bandermann and Singer (1973) estimate that cratering is \( \approx 1.2 \times \) more intense on the leading (western) hemisphere than on the trailing hemisphere. Also, leading-hemisphere impacts tend to occur at higher velocity. Thus, our collection of lunar meteorites is probably biased towards longitudes close to 90°W, and against longitudes close to 90°E. This bias may help to explain why the ratio of mare vs. highlands lunar meteorites is so high, 3.5/4.5 (counting Y-793274 as half mare), compared to the 1/5 ratio of mare to highlands surfaces over the whole Moon. The western (Procellarum) hemisphere has a higher percentage of mare surface than the eastern hemisphere.

6.3. Comparison with central nearside regoliths

6.3.1. Major elements

Table 4 shows the major-element compositions of the lunar highlands meteorites compared with average regolith compositions for the four central nearside sites where essentially mare-free regoliths were sampled. The lunar meteorites tend to be more anorthositic (richer in \( \text{Al}_2\text{O}_3 + \text{CaO} \), poorer in \( \text{MgO} + \text{FeO} \) than regolith samples from the nine Apollo/Luna central-nearside sampling sites. However, geochemists have always assumed that the Luna 20 and Apollo 16 sites (especially the latter, which was more extensively sampled) are more representative of the overall highlands than the Apollo 17 and Apollo 14 sites. The lunar meteorites show that the Apollo 16 site is the most representative for major elements of all the Apollo/Luna sites. In fact, instead of \( 25.2 \pm 2.9 \) wt% \( \text{Al}_2\text{O}_3 \) (the mean and standard deviation for the data base of Apollo 16 + Luna 20), the average \( \text{Al}_2\text{O}_3 \) content for the upper nonmare crust is now constrained to be \( 26.4 \pm 2.0 \) wt% (based on the first 6 columns of Table 4).
Table 4. Compositional data for four lunar highlands meteorites compared to other highlands regolith samples.

<table>
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<tr>
<th></th>
<th>Y-82192/86032</th>
<th>ALHA81005</th>
<th>Y-791197</th>
<th>MAC88105 new data</th>
<th>Luna 20 Avg. Soil</th>
<th>Wpollo 16 Avg. Soil</th>
<th>Wpollo 17 Al-rich Soil</th>
<th>Apollo 14 Avg. Soil</th>
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<tr>
<td>Na₂O</td>
<td>wt%</td>
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<td>MgO</td>
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<td>8.2</td>
<td>6.1</td>
<td>4.0</td>
<td>9.3</td>
<td>6.0</td>
<td>9.7</td>
</tr>
<tr>
<td>Al₂O₃</td>
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<td>25.6</td>
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<td>27.2</td>
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<td>15.0</td>
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<td>0.25</td>
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<td>0.23</td>
<td>0.47</td>
<td>0.54</td>
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<tr>
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<td>MnO</td>
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<td>0.075</td>
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<td>0.110</td>
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<tr>
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<td>7.2</td>
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<td>0.532</td>
<td>0.559</td>
<td>0.514</td>
<td>0.553</td>
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</table>

Samples are arranged in order of Th concentration.

Data sources: The composition shown for MAC88105 is a weighted mean of our data for MAC88105.41 and MAC88105.34.
For other lunar meteorites, see KoEBERL et al. (1989) or WARREN et al. (1989).

1 Y-82192/86032 has an extremely low regolith maturity, and is arguably best classified as an "ordinary" polymict breccia.
2 The Apollo 17 composition is a literature average for 73141 (references in WARREN and KALLEMEYN, 1986), the most Al-rich soil from that site.
This new, relatively precise estimate strengthens one of the prime arguments in favor of the magma ocean (or magmasphere) hypothesis, which holds that the upper crust became enriched in Al\(_2\)O\(_3\) by flotation of plagioclase over a primordial intrusion of global breadth. The alternative to the magmasphere hypothesis, formation of the crust entirely by piecemeal, "serial" magmatism (Walker, 1983), has difficulty in accounting for a concentration of Al\(_2\)O\(_3\) far in excess of the maximum expected for an aggregation of internally generated partial melt, which is constrained by phase equilibria (Walker et al., 1973) to be \(\sim 19\) wt% (assuming conservatively SiO\(_2\)-poor, MgO-rich melts). It is now clear that the upper crust has a far higher average Al\(_2\)O\(_3\) content than this. An aggregation of partial melts with \(19\) wt% Al\(_2\)O\(_3\) might generate a global upper crust with a slightly higher average Al\(_2\)O\(_3\) content as a result of localized plagioclase flotation and possibly also crystal mush diapirism (Walker, 1983; Longhi and Ashwal, 1985), but the plagioclase-enrichment process is clearly expected to be far more efficient over a global magmasphere.

Our data for Si substantially increase the total data base for this important element in lunar meteorites. We find that the Si contents of the highlands samples are monotonously close to 210 mg/g (44.9 wt% SiO\(_2\)). The only previous analyses for Si in ALHA81005 (217.2 mg/g: Palme et al., 1990) and Y-791197 (202.8 mg/g, after adjustment for terrestrial H\(_2\)O: Yanai and Kojima, 1984) indicated a wide disparity. Our data indicate no disparity. The mass-weighted mean of our data for Y-82192/86032 is similarly 210.9 mg/g. Koeberl et al. (1989) reported slightly lower results for Y-86032 (201 and 204.1 mg/g, determined by M. Lindstrom and H. Haramura, respectively). Our results for MAC88104/5 are near the middle of a narrow range defined by the results from Jarosewich (1990), Lindstrom et al. (1990), and Palme et al. (1990). Even for the mainly mare Y-793274 meteorite, our bulk-rock result of 220 mg/g suggests that the Si concentration of the highlands component is probably within the range 210–220 mg/g.

Another important parameter for the composition of the lunar crust is the Ca/Al ratio, which is sensitive to the proportion of high-Ca pyroxene among the mafic silicates. Earth-based spectroscopic studies (Lucey and Hawke, 1988; Pieters, 1989) suggest that large regions of the southern and western nearside highlands have high-Ca pyroxene as the dominant mafic silicate. If high-Ca pyroxene is abundant, the molar Ca/Al ratio should be much greater than 0.48, the average ratio of nonmare lunar feldspar (nonmare feldspar seldom extends beyond the range An\(_{88-97}\), which corresponds to a range for Ca/Al of 0.468–0.492). Among the lunar highlands meteorites (Table 4), molar Ca/Al is confined to the range 0.517–0.537, which is slightly higher than the average Ca/Al for Apollo 16 regolith (0.514), but low compared to the Ca/Al ratios of the regoliths at the three other central nearside highlands sites (0.553–0.557). The Ca/Al ratios of the highlands meteorites might be closer to that of the Apollo 16 regolith if the meteorites did not contain possibly significant proportions (spotted as clasts among a small number of thin sections) of mare materials. The Luna 20, Apollo 14, and Apollo 17 regoliths are similarly "impure." However, taken at face value, the data in Table 4 imply that there is among lunar highlands materials an inverse correlation between Al\(_2\)O\(_3\) (i.e., plagioclase) and Ca/Al (i.e., high-Ca pyroxene); and also a weaker positive correlation between Th (and associated
incompatible elements) and Ca/Al.

6.3.2. Incompatible trace elements: Implications for bulk-Moon K, Th, U, etc.

The lunar meteorites have confirmed the basic representativeness of previous (central nearside) sampling for major elements, but the situation for incompatible trace elements (epitomized by Th: Table 4) is just the opposite. The pattern of element-to-element rations for incompatible elements is basically KREEPlike in all of these samples (Warren and Kallemeyn, 1990). However, the lunar meteorites consistently have lower levels of incompatible elements than their central nearside counterparts. Part of this disparity may be attributed to a general inverse correlation between incompatible elements and Al (Fig. 9). Even compared to the comparably Al-rich Apollo 16 regolith, however, all of the lunar highlands meteorites have far lower incompatible element levels.

From Fig. 9, it appears that the highlands components in both EET87521 and Y-793274 (which, as a regolith breccia, is far more significant) are richer in Sm and associated incompatible elements (Th, U, etc.) than any of the nearly-pure-highlands meteorites. This result is not too surprising, because orbital spectrometry data (e.g., Metzger et al., 1977) indicate a rough (1000-km-scale) correlation between the abundance of incompatible elements and the abundance of mare basalts—both are concentrated over the west-central nearside. Metzger et al. (1977) used their orbital spectrometry results to derive estimates of 550 and 0.80 µg/g for the bulk-crustal concentrations of K and Th, respectively. These and similar estimates have been widely interpreted as evidence in favor of a major enrichment in Th, U, and associated refractory lithophile elements in the bulk-Moon vs. the bulk silicate portion of the Earth. For example, Drake (1986) argued that mass balance for K and Th implies that the bulk-Moon U content (the bulk-Moon K/U and Th/U ratios are indubitably within a few percent of 2500 and 3.7, respectively) must be 30–32 ng/g, or higher if any K or Th has been retained in the lunar mantle. Taylor (e.g., 1986) uses similar reasoning, stressing K, to argue for a bulk-Moon U content of at least 34 ng/g. One problem common to both of these arguments is that they introduce a systematic error of a factor of 1.18, because they inexplicably assume that the crust constitutes 12 wt% of the Moon, when in fact the crust constitutes only 12 vol% (based on the conservative assumption that the center-of-mass/center-of-figure offset is almost entirely due to thicker crust on the farside—the directly measured thickness of the nearside crust, extrapolated globally, would imply ~9 vol%, see, e.g., Taylor, 1982, p. 181). Corrected for the far lower density of the crust, 12 vol% translates into 10.2 wt%. It is also hardly a proven fact that the upper crust measured by Metzger et al. (1977) is compositionally representative of the entire 73–75 km thickness (following Taylor and Drake) of the underlying crust. But for the sake of argument, we will assume it is.

Metzger et al. (1977) derived their bulk-crustal estimates using an algorithm that reduces to

\[ C = 0.288 c_{\text{nsmb}} + 0.712 c_{\text{rs1}} \quad (2) \]

where \( C \) is the average crustal concentration, \( c_{\text{nsmb}} \) is the estimated average concentration for the nearside and mare-border highlands, and \( c_{\text{rs1}} \) is the estimated average
concentration for the farside and limb highlands. We will not quarrel with the values of $c_{\text{namb}}$ (720 µg/g K and 1.47 µg/g Th) favored by METZGER et al. (1977). However, their preferred values for $c_{\text{fsl}}$, 480 µg/g K and 0.53 µg/g Th, are far higher than any of the concentrations of these elements in the highlands meteorites, which average 205 µg/g K and 0.32 µg/g Th (Table 4). Clearly, the METZGER et al. (1977) estimates for $c_{\text{fsl}}$ should be revised. WARREN and KALLEMEYN (1990) show that many of the lowest K results used by METZGER et al. (1977) are probably erroneously high, but we will assume that their mean is still basically accurate for the roughly 20% of the farside mapped. Based on the results of METZGER et al. (1977), factor-of-two variations in surface K and Th concentration seldom occur over distances of less than ~1000 km. Assuming the highlands meteorites represent four random farside and/or limb zones, 500–1000 km in radius, they would represent roughly 14–55% of the total area of farside and limb highlands. Assuming 20%, for the sake of argument, implies that our revised estimate should be simply the average of the METZGER et al. $c_{\text{fsl}}$ and the average highlands-meteorites concentration. Thus, we estimate $c_{\text{fsl}}=342$ µg/g for K and 0.43 µg/g Th. Using eq. (2), the implied values for C are then 451 µg/g for K and 0.73 µg/g Th. Assuming that nearly all of the bulk Moon’s K and Th occur, at these concentrations, within its 10 wt% crust, the implied bulk-Moon K and Th contents are roughly 50 and 0.08 µg/g, respectively. Translated into an estimate for bulk-Moon U concentration (again using the ratios 2500 and 3.7), these results imply roughly 21 ng/g. This estimated bulk-Moon U content is far lower than the analogous estimates (32–34 ng/g) of DRAKE (1986) and TAYLOR (1986), but accords well with inferences based on lunar heat flow, allowing for effects of insulation by the Moon’s thick megaregolith (WARREN and RASMUSSEN, 1987).

6.3.3. Other trace elements

Our new data for highly siderophile elements (Ni, Ge, Re, Os, Ir and Au) in Y-86032,69 are similar to our previous Y-82192/86032 data (WARREN et al., 1989). For MAC88105, we find basically the same siderophile pattern as for the other lunar meteorites, except for a slightly higher Ge/(Re+Os+Ir) ratio (see WARREN and KALLEMEYN, 1990, for further discussion). In general, these new data only strengthen the conclusions of WARREN et al. (1989) concerning the nonrepresentative chemistry, for siderophile elements, of the megaregolith in the relatively small region associated with Apollo 16 and the other central nearside highlands sites. Our RNAA of Y-793274 is not yet complete, but the INAA results (Ni, Ir, Au) already show the sort of moderate-high concentrations that typify regolith breccias, and especially those of largely highlands derivation.

Our results for the “labile” metals Zn, Br and Cd, especially the latter, indicate considerable heterogeneity within individual meteorites. We confirm the finding of KACZARAL et al. (1986) that the Y-791197 meteorite is uncommonly rich in this class of elements. However, we find in our sample of Y-791197 only a slight enrichment in Br, and Zn and Cd are 0.39× the mass-weighted mean concentrations found by KACZARAL et al. (1986). KACZARAL et al. (1986) also found extraordinarily high Au in Y-791197 (mass-weighted mean: 21 ng/g), but we find that Au is uncommonly low in our Y-791197 sample. A small (4 mg), volatile-rich (Zn=361 µg/g) portion of Y-791197 analyzed by BISCHOFF and PALME (1986) has a relatively moderate Au
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content, 2.2 ng/g. Koeberl (1988) found a relatively high Au content (6.6 ng/g) in a Y-791197 sample with uncommonly low Br (<80 ng/g). Clearly, despite its high maturity by regolith breccia standards (Fig. 2), Y-791197 is heterogeneous for Au and the “labile” trace elements. Probably the most plausible mechanism to engender these heterogeneous enrichments is by deposition from fumarolic fluids (Kaczaral et al., 1986).

6.4. Abundance of regolith breccias

As samples from the archetypical nomare Apollo site, the Apollo 16 rocks are commonly assumed to be highly representative of the upper lunar crust, chemically and petrologically. However, the small proportion (9%) of the Apollo 16 rocks classified as regolith breccias by Ryder and Norman (1980) stands in stark contrast with the prevalence of regolith breccias among the randomly-derived lunar highlands meteorites: three clear-cut regolith breccias, one partly-mare sample that is also a clear-cut regolith breccia (Y-793274), and one borderline regolith/fragmental breccia (Y-82792/86032). Consider the results if eq. (1) is applied to the probability for having 3 out of 3 randomly-derived highlands rocks be regolith breccias. If the individual probability were truly (as suggested by the Apollo 16 data) 9%, the probability for 3 out of 3 regolith breccias would be $0.09^3=0.00073$. Even with the individual probability trebled to 27%, and assuming that the trio of Y-791197, ALHA81005, and MAC88104/5 is actually (due to some form of pairing) a duo, the probability for 2 out of 2 regolith breccias would be a none-too-likely $0.27^2=0.073$. Based on these calculations, we conclude that either (a) some lunar meteorites are transformed from incoherent soils into regolith breccias by the same impacts that blast them Earthward (Warren and Kallemeyn, 1986; Ostertag et al., 1986), or (b) regolith breccias are underrepresented among the Apollo 16 rocks. In fact, the proportion of regolith breccias is much higher (roughly 50%) among rocks from the one other “pure highlands” Apollo site, Apollo 14 (Fruland, 1983). Also, Spudis (1984) has argued that sampling procedures on the Apollo 16 mission biased the collection in favor of tough, crystalline rocks, and against “clastic debris” (i.e., regolith breccias).

If the proportion of regolith breccias in the lunar megaregolith really is much higher than implied by the Apollo 16 rock collection, one of the implications relates to interpretation of lunar thermal evolution. Interpretation of lunar heat flow data is strongly dependent on the mean thermal conductivity assumed for the megaregolith (Conel and Morton, 1975; Warren and Rasmussen, 1987). Warren et al. (1991b) show that megaregolith insulation also can lead to substantial (10–100 Ma) time lags between igneous crystallization and isotopic closure for Sm-Nd and Rb-Sr ages, for rocks formed many kilometers deep in the lunar crust. Regolith breccias tend to have high porosity, generally 15–37% (Wentworth and McKay, 1984). Among lunar materials, thermal conductivity varies in proportion to $e^{-0.13q}$, where $q$ is the porosity in percent (Langseth et al., 1976; Warren and Rasmussen, 1987). Warren and Rasmussen (1987) deliberately excluded many regolith breccias from the database that they used to estimate the overall megaregolith porosity. If, as an example, the mean porosity of the megaregolith is 20%, instead of 17% as assumed by Warren and Rasmussen (1987), its conductivity should be roughly 68% of that assumed, and
its insulating should be $1.5 \times$ greater than those calculated, by Warren and Rasmussen (1987) and Warren et al. (1991b).

6.5. The mare/nonmare petrologic dichotomy of lunar magmatism

As discussed earlier, the prevalence of VLT varieties among mare meteorites (and mare clasts in highlands meteorites) suggests that VLT types are more common than previously assumed. Remote-sensing data are not very helpful in mapping the relative proportions of VLT vs. "normal" mare basalts, because neither the Apollo gamma-ray spectrometry (Metzger et al., 1979) nor Earth-based spectral reflectance (Pieters, 1978) are sensitive enough to yield precise measurements for TiO$_2$ contents of <2 wt%. The correlation between results from the two techniques (Metzger et al., 1979, Fig. 4) is feeble, even for much higher TiO$_2$ contents. If VLT types are common, then the fundamental dichotomy between mare and nonmare products of lunar magmatism becomes slightly blurred. The dichotomy has seldom been defined in any precise way, because it is generally obvious whether an Apollo rock of Luna rocklet is mare or nonmare. Wood (1975) suggested that mare materials be defined as those having molar Ca/Al > 0.786 - 0.229 log$_{10}$ (TiO$_2$), where TiO$_2$ is in wt%. Using this criterion, EET78521, Apollo 17 VLT, and Luna 24 VLT plot within the mare range, but close to the proposed dividing line; and our analysis (Table 2) of the Y-793274 highlands-mare mixture plots on the highlands side of the dividing line.

Spectral reflectance studies (Lucey and Hawke, 1988; Pieters, 1989) suggest that large regions of the southern and western nearside, traditionally mapped as highlands, have high-Ca pyroxene as the dominant mafic silicate. These regions rich in high-Ca pyroxene may be dominated by rocks transitional between the VLT mare basalts and the nonmare Mg-suite gabbronorites of James and Flohr (1983). Two of the six Mg-gabbronorites identified by James and Flohr (1983), namely the 67915 sodic ferrogabbro of Marti et al. (1983) and one of the 76255 clasts described by Warner et al. (1976), are classified as mare by Wood's (1975) chemical criterion. However, the VLT-mare samples are distinctive texturally as well as compositionally. Compared to typical nonmare pristine rocks (sometimes known as nonmare "plutonic" rocks), the VLT samples, including lunar meteorites, have distinctly unequilibrated pyroxenes (e.g., Vaniman and Papike, 1977; Kurat and Kracher, 1981; Yanai and Kojima, 1990)—confirming that they cooled rapidly within lava flows or extremely shallow intrusions. In contrast, most of the Mg-gabbronorites have relatively well-equilibrated pyroxenes. Two separate Mg-gabbronoritic clasts described from 76255 by Warner et al. (1976), and the 61224 Mg-gabbro described by Marvin and Warren (1980), all have coarse high-Ca exsolution lamellae (within low-Ca pyroxenes), typically 25-40 µm across. Pyroxenes in the 73255 Mg-gabbronorite clast described by James and McGee (1979) contain exsolution lamellae up to 6 µm across. However, some of the pristine Mg-suite gabbronorites are far from "plutonic". The 67667 olivine gabbronorite has pyroxene exsolution lamellae only up to about 1 µm across, and is inferred to have formed in "a near-surface environment which allowed relatively rapid cooling" (Hansen et al., 1980). Pyroxenes in the 67915 sodic ferrogabbro are similarly unequilibrated, with submicron sized exsolution lamellae (Marti et al., 1983). Most of these rocks, even those with coarse exsolution lamellae, are relatively
fine-grained, by nonmare pristine rock standards. 

Warren (1986) argued that the slower cooling of deeply-buried intrusions enhances their potential for assimilation of preexisting plagioclase, which should lead to an inverse correlation between depth of origin and abundance of high-Ca pyroxene (which has relatively high Ca/Al, whereas plagioclase has relatively low Ca/Al). Lucey and Hawke (1988) noted that potential for assimilation might also have been limited, in the western nearside highlands, by a scarcity of preexisting anorthosite—which might have resulted in a relatively high proportion of gabbroic (high-Ca pyroxene-rich) nonmare rocks. Of course, the occurrence of gabbroic rocks in these nominally highlands regions might be explained more prosaically if these regions actually consist of highlands materials thoroughly blended with a thin veneer of uncommonly ancient (and thus, uncommonly impact-battered) mare basalts. The two Mg-gabbronorites that have been dated by the Sm-Nd method are relatively young, by pristine nonmare rock standards: 4.18±0.007 (Carlson and Lugmair, 1981b) and 4.23±0.05 Ga (Carlson and Lugmair, 1981a), for the 67667 and 73255 samples, respectively. A mare-basaltic clast from Apollo 14 has a Rb-Sr age of 4.23±0.05 Ga (Taylor et al., 1983). Thus, Mg-gabbronoritic magmatism and mare magmatism temporally overlapped. It seems likely that at least occasionally intermediate rock types were generated. Additional sampling of the Moon should eventually allow us to delineate more precisely the nature and significance of the fundamental mare/nonmare dichotomy.

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