

PROCRUSTEAN SCIENCE: INDIGENOUS SIDEROPHILES IN THE LUNAR HIGHLANDS, ACCORDING TO DELANO AND RINGWOOD

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(Received 20 July, 1978)

Abstract. The best estimate of indigenous lunar siderophiles comes from 29 pristine lunar rocks, characterized by low siderophile abundances, plutonic textures, and high age. Delano and Ringwood's blanket rejection of these rocks, on the contention that they are impact melts, is not justified by the petrologic evidence. Contrary to their claims, gold in highland breccias is largely meteoritic and is unaffected by fumarolic volcanism, as shown by its correlation with Ir and noncorrelation with fumarolic Tl ($r = 0.896$ and 0.272). Delano and Ringwood's approach, involving subtraction of an H-chondritic meteoritic component from highland breccias, ignores the variation of Ir/Au ratios in modern and ancient meteorites, and hence leads to spurious excesses of Au, Ni, and volatiles, and in some cases to physically meaningless, negative residuals. Their 'excess volatiles' in highland crust relative to mare basalts disappear when the highland composition is based on pristine lunar rocks rather than under-corrected breccias. Contrary to claims by Delano and Ringwood, the Ni/Co trend in Apollo 16 samples cannot be explained by an indigenous component rich in Ni (150–200 ppm) and Co (30–45 ppm); mixing lines show that much lower Ni and Co contents are required (e.g., ~ 7 ppm each).

Chondrites and lunar highland breccias show essentially parallel fractionation trends for the siderophile-element ratios Re/Ir, Au/Ir, Ni/Ir, Ni/Pd, and Os/Ir. Because the chondritic ratios were established in the solar nebula, it appears that the lunar ratios also reflect nebular processes, and have not been modified by planetary processes.

Properly derived abundances for the lunar highlands show large, systematic depletions relative to terrestrial oceanic tholeiites, by the following factors: Ge 270, Re 230, Sb 170, Zn 150, Au 60, Tl 50, Ag 48, Ni 42, Se 12. It would seem that the resemblance to the Earth's mantle is not quite as 'striking' as claimed by Delano and Ringwood.

1. Introduction

Delano and Ringwood (1978a, b, c) contend that the siderophiles in the lunar highlands are mainly of indigenous rather than meteoritic origin. I shall endeavor to show that they reach this conclusion by Procrustean methods, stretching or chopping the evidence to fit a pre-conceived mold.

2. Pristine Highland Rocks

The most direct way to determine the abundance of siderophiles and volatiles in the lunar highlands is to find pristine rocks that have escaped contamination by meteoritic debris. Over the years, 29 such meteorite-free rocks and clasts were identified at Chicago on the basis of their low siderophile element abundances (see Hertogen *et al.*, 1977, for a list), and additional samples were found by other workers. Warren and Wasson (1977),

and Warren *et al.* (1978) have developed systematic criteria for the pristine nature of such samples, and have compiled a list of 25 pristine rocks greater than 5 g in weight. Other authors who have discussed pristine highland rocks include Bickel and Warner (1977, 1978), Dixon and Papike (1975), and Warner and Bickel (1978).

Delano and Ringwood (1978a, b) made no mention of any of these data, and instead calculated an indigenous component (PLC, for parental lunar crustal magma) by subtracting an assumed meteoritic component from the bulk breccia analyses. In response to criticism at the 9th Lunar Science Conference, they later acknowledged the existence of meteorite-free rocks (Delano and Ringwood, 1978c, d) but contended that they were secondary differentiates from large impact melt pools that had frozen so slowly as to allow settling of metal. A similar point of view, but with unspecified melting events, was adopted by Wänke *et al.* (1978b). Delano and Ringwood (1978c) then developed this argument in detail for Apollo 15 KREEP basalt 15382, representing a single case among Warren and Wasson's (1977) 25 samples and 10 rock types.

There are many objections to this view, only a few of which will be stated here.

(1) There is no evidence that the combination of complete melting and slow freezing, needed for settling of metal, is ever attained in lunar impacts. Though impact melted rocks are common in the highlands (Irving, 1975; Simonds *et al.*, 1974) they generally still have their meteoritic metal and hence must have been chilled too fast (by entrained clasts, Simonds *et al.*, 1976) to lose their meteoritic metal. Even in those rare, pseudoigneous rocks where relict clasts are absent, implying complete melting (14310, 68415, etc.), meteoritic metal has been retained (James 1973). Since the basaltic texture of 15382 is essentially identical to that of pseudoigneous rocks such as 14310 (Dowty *et al.*, 1976), the freezing time cannot have been longer, and so there is no reason to expect settling of metal. The scale of the melting event does not change the picture; even in the 2 km melt sheets of Apollo 17, quenching by cold clasts led to rapid freezing (Simonds *et al.*, 1974, 1976) and retention of siderophiles (Higuchi and Morgan, 1975; Morgan *et al.*, 1975a).

(2) Some of the meteorite-free rocks are clasts within, and hence older than the siderophile-rich breccias that Delano and Ringwood consider more pristine. Many show isotopic evidence for greater age, i.e., Rb–Sr ages greater than 4.2 AE or very low initial $\text{Sr}^{87}/\text{Sr}^{86}$ ratios (see Table 1 of Warren and Wasson, 1977). KREEP basalt 15272, in particular, contains Pu^{244} fission tracks (Haines *et al.*, 1975), and hence must be older than the KREEP breccias that lack them. Because radiometric clocks can be reset by impact and metamorphism, it matters little that *some* pristine rocks have low ages. Others have higher ages, all the way to the venerable 76535 troctolite, dated at 4.55 AE.

(3) Many pristine rocks have textures indicating a plutonic origin, e.g., 76535; the 'marble cake' clast of 72255; spinel cataclastites (Herzberg and Wood, 1978); and all cataclastic anorthosites. There is not a scintilla of evidence that such textures can be duplicated by impact melts in even the largest basins.

Other arguments for the pristine nature of meteorite-free rocks have been presented by

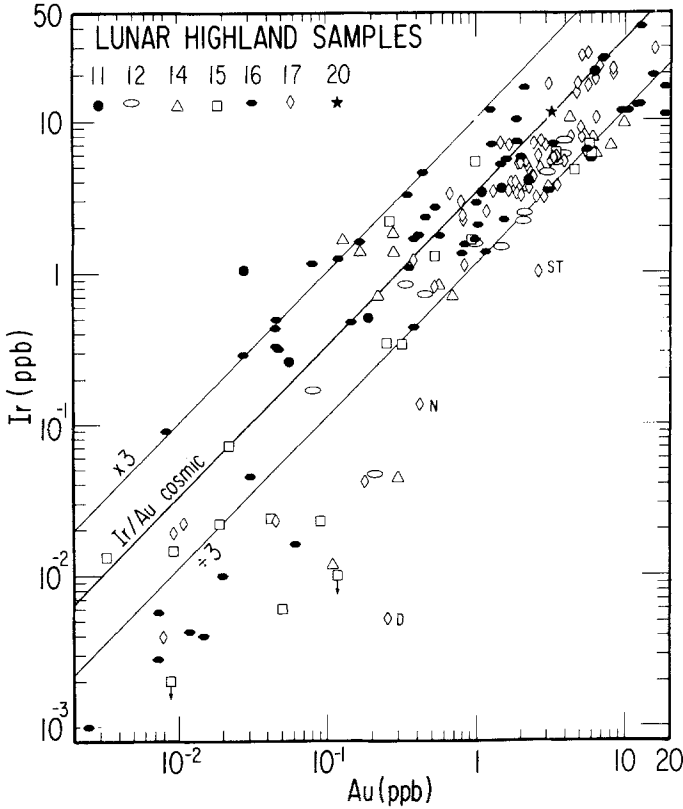


Fig. 1. Among rocks (mostly breccias) containing more than 0.1 ppb Ir, Ir and Au correlate within a factor of 3 of the cosmic ratio, and hence must be mainly of the same, meteoritic origin. The correlation begins to break down below 0.05 ppb Ir, but the apparent excesses, attributable to an indigenous Au component, lie mainly between 0.01–0.1 ppb Au. Most authors regard these, mainly plutonic, rocks as pristine.

Data from Janssens *et al.* (1978), Hertogen *et al.* (1977), and earlier Chicago papers cited therein; they include only rocks (except from Apollo 11, 12 and Luna 20, where the scarcity of highland rocks made it necessary to use soil separates). Three rocks of unusually high Au content are identified by letters: ST = spinel troctolite 73215, 9007, 170; N = norite 78235, 31a; D = dunite 72415, 10.

Warren and Wasson (1977). It will take more than a fiat by Delano and Ringwood to demote these rocks to impact melts.

3. Indigenous Siderophiles in the Lunar Highlands

3.1. INDIGENOUS COMPONENT FROM BRECCIAS (DELANO AND RINGWOOD)

Delano and Ringwood (1978a, b, c) reject pristine lunar rocks, and use an altogether different approach: they assume that all Ir in highland samples is meteoritic, and then calculate an indigenous component by subtracting an H-chondrite meteoritic component,

scaled to Ir, from the gross abundances in the breccias. This approach would be perfectly valid (though inaccurate, as it involves the difference of two large numbers) if H-chondrites were the only meteorites that had fallen on the lunar highlands. But this is manifestly not so. H-chondrites comprise only 37% of the present meteorite influx on Earth, and since the bodies responsible for the highland bombardment differed strikingly from present-day meteorites in half-life, flux, and size distribution (basins!), it is rash to assume that the composition was the same. But this idea, whatever its *a priori* plausibility, can be tested against observation, now that analyses of highland breccias are available. These breccias, like chondrites, show substantial variations in the ratio of the refractory siderophiles Ir, Re, Os (which condense ahead of Fe in the solar nebula) to the more volatile siderophiles Au, Ni, Pd (which condense along with Fe). As shown in Figure 1, very few samples have Ir/Au ratios near the H-chondrite value (0.98 the Cl chondrite value); some are higher but most are lower. Thus a meteoritic correction based on the Ir/Au ratio of H-chondrites will leave large excesses of Au for the samples below the line, and physically meaningless, negative Au concentrations for the samples above the line.

Delano and Ringwood attribute the scatter to volatilization of Au. As I shall show in Section 4, there is much evidence contradicting this view, but suggesting instead that the siderophile element pattern in the breccias has remained undisturbed in most cases. Let me therefore illustrate the consequences of the Delano–Ringwood approach in terms of the Chicago model, which subtracts an indigenous component (based on the rocks in the lower left of Figure 1) to find the net meteoritic pattern. Figure 2 shows the mean abundance patterns of 8 ancient meteoritic groups found in this way (Gros *et al.*, 1976), along with a pattern for H-chondrites (Hertogen *et al.*, 1977).

The first 3 groups, comprising the majority of samples in Figure 1, are deficient in Ir, not only relative to Au but also relative to other siderophiles. H-chondrites, on the other hand, show an essentially flat trend across the siderophiles, and 4 to 10-fold lower abundances of volatiles. Subtraction of an H-chondrite pattern *scaled to Ir*, as practiced by Delano and Ringwood, thus guarantees large positive residuals of all these elements for Groups 1 to 3, but negative residuals for Groups 6 and 7.

Delano and Ringwood assigned the positive residuals to their PLC component, but had the discretion not to mention the embarrassing negative residuals. In the best Procrustean tradition, they further tried to minimize the embarrassment by failing to use the data of Hertogen *et al.* (1977), which include 10 new members of Group 7, and by excluding rocks of more than 30% Al_2O_3 , many of which belong to Group 7 or are meteorite-free. There is little merit to their argument that samples containing more than 30% Al_2O_3 had to be excluded because the large ($> 3\times$) corrections for plagioclase dilution would have “introduced dispersions that were largely artifacts of the high Al_2O_3 in the sample”. Most Al_2O_3 -rich samples contain such small amounts of siderophiles (0.01–0.1 ppb), that even a large error in the plagioclase correction would cause only a small absolute error in the net siderophile abundance. Indeed, since many anorthositic rocks have (Ir, Os, Re/Au, Ni) ratios above the H-chondrite value, they would have yielded negative Au, Ni residuals by the Delano–Ringwood method – a clear warning of the fallacy of their approach.

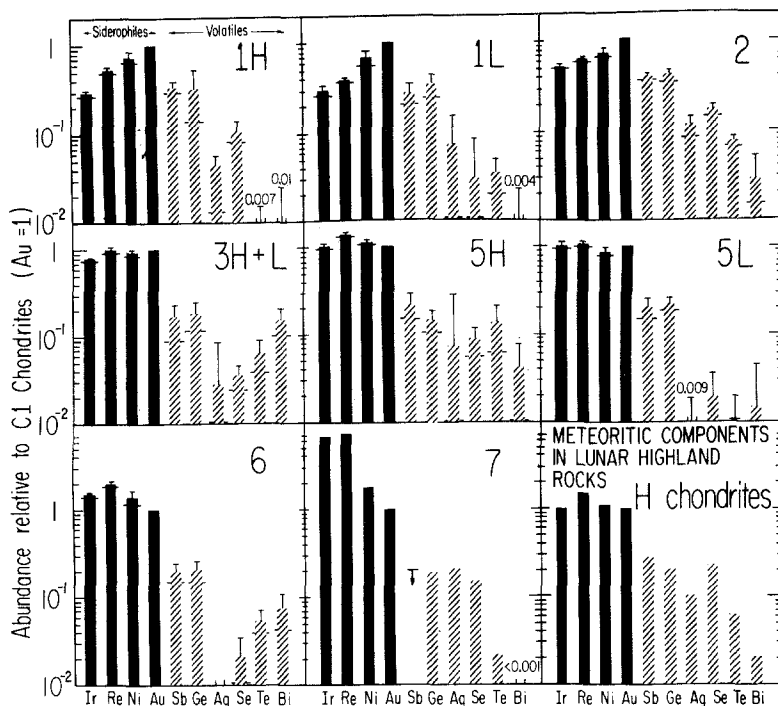


Fig. 2. Abundance patterns of 8 lunar meteoritic groups and H-chondrites (Gros *et al.*, 1976). The first 4 groups are depleted in Ir relative to H-chondrites, and so subtraction of an H-chondrite pattern scaled to Ir, as practiced by Delano and Ringwood, ensures large excesses at other elements. But when applied to Groups 6 and 7, this procedure gives physically meaningless, *negative* residuals at several elements.

Because the residuals depend so critically on the Ir/Au ratio, the following statement by Delano and Ringwood (1978b, c) is patently false: “*However, it must be emphasized that the general conclusions discussed in this study do not depend strongly on the assumption that the meteoritic component was precisely of H-group composition (their italics)*”.

3.2. INDIGENOUS COMPONENT FROM PRISTINE HIGHLAND ROCKS (CHICAGO GROUP)

Given a suite of meteorite-free highland rocks, it is easy to obtain representative trace element abundances for the highland crust. Of 16 trace elements measured at Chicago, 12 correlate with Rb and Cs at the 95% confidence level, and so their abundances can be represented by logarithmic regressions against Rb or Cs content (Gros *et al.*, 1976). Four others (Ir, Au, Ni, and Te) show only weak correlations of small slope, and for them it matters little whether the abundance is based on a regression (Figure 3) or a straight average. Using the regressions given by Gros *et al.* (1976), I have calculated a mean composition for the pristine highland crust, on the assumption that it contains 0.72 ppm Rb (the same as the estimated Rb content of the PLC component, based on

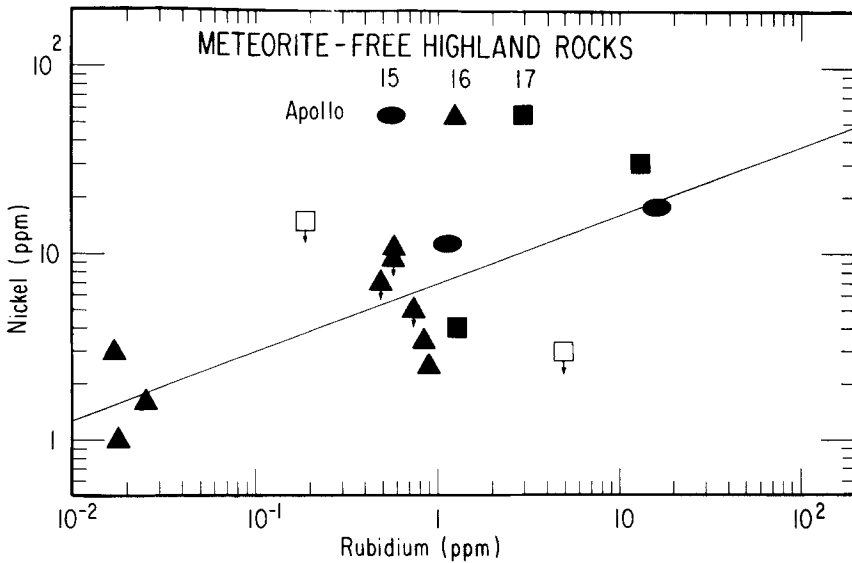


Fig. 3. Correlation of Ni with Rb in pristine highland rocks (data from Hertogen *et al.*, 1977, and earlier Chicago papers). The two samples represented by open symbols were not used in calculating the regression line.

$K = 250$ ppm and $K/Rb = 320$). It is not quite equivalent to the PLC component, which was corrected for plagioclase dilution on the (oversimplified) assumption that $D(\text{plag/liq}) = 0$.

These abundances are compared in Table I with those of terrestrial oceanic tholeiites and the PLC component of Delano and Ringwood (1978b, c). Both the PLC component and tholeiites are consistently enriched over pristine highland rocks, by factors of 6–70 and 12–270. The PLC component matches tholeiites for the first 3 elements,* but not for the next 4, and even less so for the last 2, Re and Tl. Delano and Ringwood thoughtfully omitted Re and Tl, although good data are available for both, but when this oversight is corrected, these elements, too, are greatly overabundant in tholeiites relative to the PLC component.

* The match for Ni seems to have been achieved by Procrustean techniques. Ringwood and Kesson (1977) first discard all Apollo 11 and 17 basalts, which happen to have very low Ni contents (< 10 ppm). They then plot 33 low-Ti basalts from Apollo 12 and 15, averaging about 50 ppm Ni, and contend that “the most primitive low-Ti basalts contain 150–170 ppm Ni (Figure 5)”. A value of 150 ppm Ni thereafter appears as the ‘mean abundance’ in Table 1 of Ringwood and Kesson (1977), and again in Delano and Ringwood (1978a, b, c). But a check of Figure 5 of Ringwood and Kesson (1977) shows the following Ni contents for the 5 most primitive basalts, in the order of decreasing primitiveness (= $Mg/Mg + Fe$): 45, 90, 60, 140, and 65 ppm. Only 3 of the 33 basalts have Ni contents above 100 ppm, and none exceeds 140. (Only green glass does, at 170 ppm, but this unique sample of controversial origin is not a true basalt). It is not clear how a range of 150–170 ppm, let alone a mean of 150 ppm, can be obtained from these data by conventional arithmetic.

TABLE I
Siderophiles and volatiles in lunar highlands and terrestrial oceanic tholeiites

	Earth Tholeiites ^a	Moon			PLC	Tholeiites
		Pristine rocks ^b	PLC ^c	Mare basalts ^a	Pristine	Pristine
Ni ppm	250	6	200	150	33	42
Au ppb	~1	0.017	~0.7	0.03	~40	~60
Se ppb	170	14	180	140	13	12
Ag ppb	30	0.62	4	1	6	48
Sb ppb	29	0.17	2	0.06	12	170
Ge ppb	1500	5.5	400	7	73	270
Zn ppm	100	0.67	11	1	16	150
Re ppb	1.05	0.0045	~0	0.0043	~0	230
Tl ppb	13	0.26	~0.3	0.60	1	50

^a Delano and Ringwood (1978b, c) except Tl and Re (Gros *et al.*, 1976; de Albuquerque *et al.*, 1972; Kimura *et al.*, 1974, and J. Hertogen *et al.*, unpublished work).

^b Regression analysis of 13-22 pristine highland rocks (Gros *et al.*, 1976), for Rb = 0.72 ppm. The Ni value is based on later, more complete data (Figure 3).

^c PLC (parental lunar crustal) magma, obtained by subtracting an H-chondrite component from lunar breccias and soils (Delano and Ringwood, 1978a, b, c). Though they quote no values for Re and Tl, abundances for these elements can be readily estimated on their model, since their H-chondrite component accounts for essentially all the Re but none of the Tl in the breccias.

Delano and Ringwood (1978b, c) find it necessary to explain why certain elements (Zn, Ge, As, Ag, Sb, and Au) are enriched in the PLC component relative to mare basalts, and invoke fumarolic volcanism on the lunar surface (see Section 4 for criticism of this suggestion). However, these troubles are entirely of their own making. Pristine highland rocks, in contrast to the PLC component, show no significant enrichment in these elements (Table I) or in other volatiles, relative to mare basalts (see Table 3 of Gros *et al.*, 1976, for data on additional volatiles).

Delano and Ringwood (1978b, c) contend that tholeiites, mare basalts, and the PLC component agree closely in their siderophile element patterns, and since this pattern allegedly shows the unique signature of the Earth, they conclude that the Moon comes from the Earth's mantle. However, to fit the data into this Procrustean bed, they find it necessary to redefine the geochemical character of certain elements. Thus Se, S, Cu, and Ga are redefined as 'siderophile, non-volatile', whereas Au, As, and Sb are demoted to 'volatile', without regard to the nebular condensation sequence (Grossman and Larimer, 1974; Wai and Wasson, 1977; Takahashi *et al.*, 1978), or to the ample evidence that S is volatile under lunar conditions (Sato *et al.*, 1973; Brett, 1976; Gibson *et al.*, 1977). One wonders why they omitted Re, a siderophile, non-volatile element *par excellence*, which nonetheless is 200-fold overabundant in terrestrial rocks (Table I). With a Mohs hardness of 6, it may have been too hard for Procrustes' axe.

4. Volatility of Meteoritic Elements in Lunar Rocks

Early in their paper, Delano and Ringwood (1978b, c) casually dismiss our entire effort on meteoritic components in the highlands, arguing that because *some*, mainly *volatile*, elements (Rb, K, Na, In, Cd, Zn, Ge; and allegedly also Fe and Si) occasionally fractionate upon impact, *none* of the *siderophile* elements we use for our classification (Ir, Re, Os, Ni, Pd, Au) can be trusted. A few pages later, they suspend their qualms long enough to argue that the low abundance of volatile Bi and Te (relative to siderophile Ir) proves that the projectiles were of chondritic composition. Apparently the two processes that play havoc with volatiles throughout their paper – impact volatilization and fumarolic volcanism – can be ignored whenever the data fit.

There is no question that post-impact volatilization is a potentially important process. But the unwary reader of Delano and Ringwood would never guess that this process has been of abiding concern to us since 1971 (Laul *et al.*, 1971; Morgan *et al.*, 1972; Ganapathy *et al.*, 1973, etc.). Because we measure 12–14 other, mainly volatile elements along with the siderophiles used for classification, we have good internal checks on any volatilization losses, and enough redundancy among our diagnostic siderophiles to expose any anomalies.

Until 1974, we also used Ge and Sb as diagnostic elements but as we came to realize that these elements, though reliable for lunar soils, gave erratic results for some 20% of the breccias (Ganapathy *et al.*, 1973; Morgan *et al.*, 1974), we abandoned them in favor of less volatile elements (Higuchi and Morgan, 1975; Gros *et al.*, 1976; Hertogen *et al.*, 1977). This led to the demise of Group 4 (low Ge and Sb), which in its heyday consisted of 3 samples. Over the years, we have applied a significant part of our effort to critical reexamination of our methods, and to improvements in our criteria for contamination, volatilization, and other errors. Because we had no trouble recognizing a few isolated instances of volatilization or redistribution of Au (76295,49 and –52; 79215,34; Morgan *et al.*, 1974; Higuchi and Morgan, 1975; Hertogen *et al.*, 1977), we may not be quite the simpletons Delano and Ringwood accuse us of being. Nonetheless, it is instructive to examine their claims for the mobility of gold, a pivotal element in our classification.

4.1. MAXWELL'S DEMON AT WORK

Delano and Ringwood (1978b, c) contend that Au has been fractionated from Ir in the lunar highlands by fumarolic volcanism or certain sequences of igneous and hydrothermal processes. If Ir and Au were indeed decoupled from each other to such an extent, then one must wonder what Maxwellian demon kept them so nicely correlated in Figure 1. Rocks containing between 0.3 and 30 ppb Ir nearly always have an Au content within a factor of 3 of the cosmic ratio; only at lower Ir contents do excesses of Au begin to show up. But these excesses are small in an absolute sense; less than 10^{-1} or even 10^{-2} ppb. In contrast, the rocks at the upper end of the graph would require 1 to 10 ppb Au to displace them below the cosmic (or H-chondrite) line to the observed degree. I marvel at the parsimony of Maxwell's demon, who carefully limited his gifts of gold to within

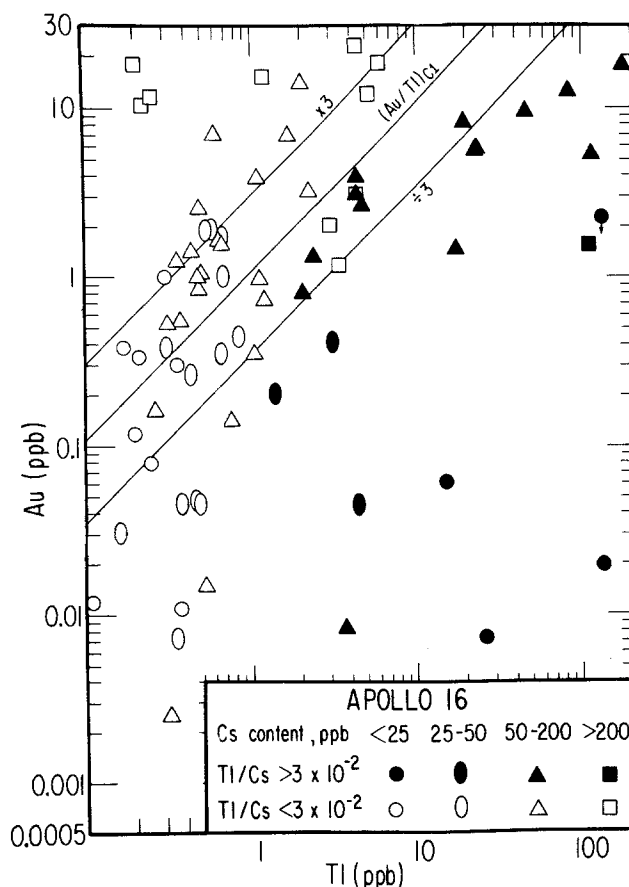


Fig. 4. If gold were of fumarolic origin, as proposed by Delano and Ringwood (1978b, c), then it should correlate with fumarolic Tl. However, rocks containing excess Tl (filled symbols) range all the way from very low to very high Au contents. There is no apparent correlation between Au and excess Tl. Data from Krähenbühl *et al.* (1973), Ganapathy *et al.* (1974b), and Hertogen *et al.* (1977); they include rocks and soil separates but no bulk soils.

a factor of 3 of the cosmic value at the high end of the graph, and allowed himself flings of generosity only at the low end. Yet his charity was well controlled even there, never exceeding 0.5 ppb.

4.2. IS GOLD A FUMAROLIC ELEMENT?

There is more direct evidence against the fumarolic hypothesis. Thallium shows the largest fumarolic enrichment, by up to 3 orders of magnitude (Krähenbühl *et al.*, 1973) whereas other volatiles such as Cd and Br show smaller but correlated enrichments (Figure 3 of Ganapathy *et al.*, 1974a; Figure 4 of Ganapathy *et al.*, 1974b). But Au shows no correlation with Tl. Ganapathy *et al.* (1973) showed by a statistical study of 150 lunar samples that Au correlates with Ir but not Tl, and hence is meteoritic, not hydrothermal

(correlation coefficients of 0.896 and 0.272). But since this work has been ignored by Delano and Ringwood, the point will be made pictorially (Figure 4).

Of the rocks containing excess Tl (defined as having Tl/Cs at least 3 times the average lunar ratio of 1×10^{-2} ; Krähenbühl *et al.*, 1973) some, in the upper right, have up to 20 ppb Au, whereas others, in the lower right, have less than 0.1 or even 0.01 ppb Au. Conversely, some of the rocks highest in Au are among those lowest in Tl (upper left). This chaotic trend cannot be blamed on the smaller volatility and hence earlier deposition of Au in the fumarole, because then high Au should never be associated with high Tl. Moreover, Tl enrichments suggestive of fumarolic volcanism have been detected only at Apollo 16, not at other sites where low Ir/Au ratios are equally common (Figure 1).

Figure 4 nicely illustrates the random pattern of two unrelated elements in the absence of a Maxwellian demon, which we looked for in vain in Figure 1. Unfortunately for Delano and Ringwood, their model predicts exactly the reverse trend for these two figures.

4.3. GREEN GLASS

As further proof of the volatility of Au, Delano and Ringwood (1978b, c) cite the fact that 93% of the Au was removed from a handpicked sample of 15421 green glass when 15% of the sample was dissolved in HCl and HF (Chou *et al.*, 1975). But so was 34% of the Ir, and since all parties agree that Ir is not very volatile, leachability alone is not proof of volatility. A clue is provided by an earlier leaching experiment on soil 10084 (Laul *et al.*, 1971), which showed that about 20–30% of the meteoritic elements Ir, Au, Ni, etc., is removed when only ~3% of the soil is dissolved. Apparently these elements reside in a surface coating of recondensed meteoritic material.

Moreover, the leaching experiment of Chou *et al.* (1975) had no way of distinguishing a genuine surface component from contamination. No material balance data were obtained in that experiment, and since their green glass sample (Table II) had twice as high an Au/Ir ratio as another sample measured by Ganapathy *et al.* (1973), contamination is a distinct possibility. Indeed, since the 'non-green-glass' sample from the same soil was an order of magnitude richer in Au, it, too, must be considered as a possible source of excess, acid-leachable Au.

Even if future work should establish the presence of a surficial Au component, this would not prove that Au is volatile in lunar volcanic processes, as opposed to impact processes. A fine-grained green glass fraction from breccia 15426 was uniformly enriched

TABLE II
Siderophiles in Apollo 15 green glass

Sample	Ir ppb	Au ppb	Au/Ir	Reference
15426, 35, 1 Green Glass	0.22	0.19	0.86	Ganapathy <i>et al.</i> (1973)
15421, 27 (> 105 μ m) Green glass	0.35	0.59	1.69	Chou <i>et al.</i> (1975)
15421, 27 (> 105 μ m) Non-green-glass	5.1	5.3	1.04	Chou <i>et al.</i> (1975)
15431, 33 Soil	4.5	1.63	0.36	Ganapathy <i>et al.</i> (1973)

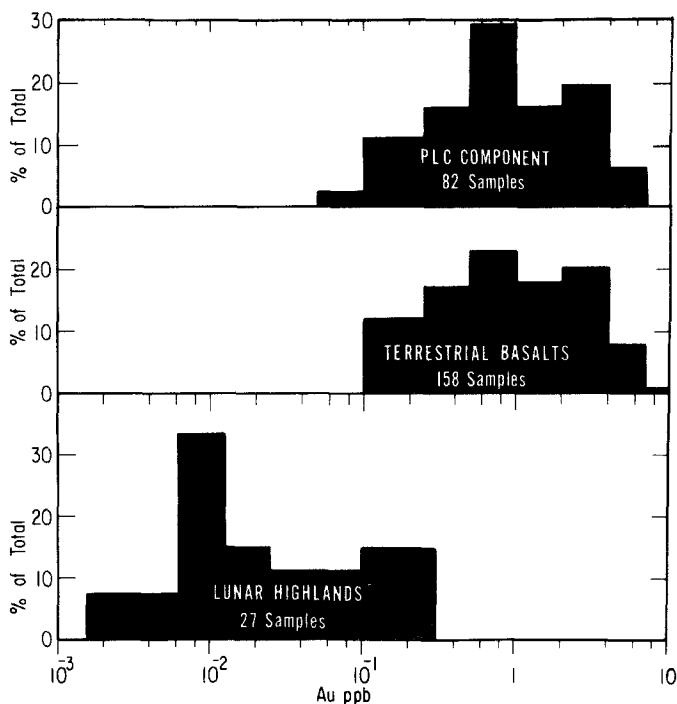


Fig. 5. Delano and Ringwood's (1978a, c) PLC (parental lunar crustal) component matches terrestrial oceanic basalts, but only because it contains spurious excesses of Au resulting from under-correction for the meteoritic component. In contrast, pristine highland rocks (bottom panel) lie 2 orders of magnitude lower and hence do not match terrestrial basalts.

relative to a coarse fraction in 12 trace elements, ranging from highly volatile Bi, Tl, Br, In, Cd, Se to refractory Ir and Re (Figure 5 of Ganapathy *et al.*, 1973). As these authors point out, covariance of elements of such diverse geochemical character is not likely to result from magmatic processes, but presumably reflects non-selective vapor deposition in an impact. But since even very refractory elements such as Ir and Re vaporize fully in a superheated impact cloud, *non-selective*, surface enrichment of Au in impacts does not imply volatility in volcanic processes. On the other hand, selective, large enrichments in non-cosmic proportions, as found for Zn, Cd, In (Chou *et al.*, 1975), can definitely be attributed to volcanism or other endogenous processes, especially when confirmed by direct measurements on the ion microprobe (Meyer *et al.*, 1975).

4.4. GOLD IN BASALTS

The very low abundance of Au in lunar mare basalts was one of the first chemical differences between Earth and Moon established by Apollo 11 (Ganapathy *et al.*, 1970), and has persisted as data became available from other mare sites (Kimura *et al.*, 1974) and pristine highland rocks (Gros *et al.*, 1976). Delano and Ringwood (1978a, b),

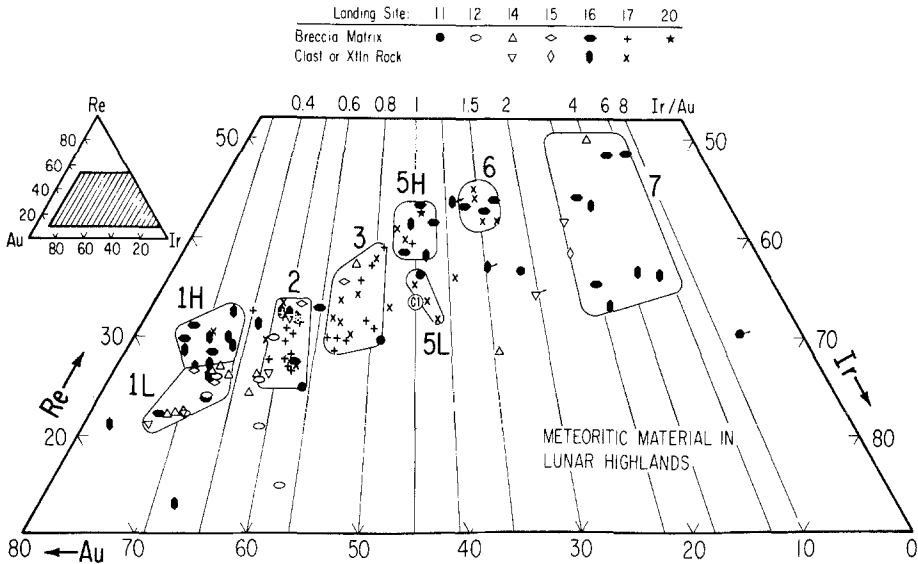


Fig. 6. Ancient lunar meteorite groups (Janssens *et al.*, 1978) do not form a single continuum, as argued by Ringwood, for the following reasons.

(a) On an AuReIr diagram they are better resolved from each other than are individual chondrite classes, which are known to be discrete (Figure 7).

(b) Different groups are not evenly distributed over landing sites, as expected if the siderophiles were controlled by post-impact processes on the lunar surface. Thus Apollo 17 samples occur mainly in Groups 2 and 3, and Apollo 16 samples in 1H and 5–7. Any redistribution can have occurred only to a limited extent, because few Apollo 16 samples lie on a mixing line between Groups 1H and 5–7, and few Apollo 17 samples lie between Groups 2 and 3.

faithful to the Procrustean approach, omit pristine highland rocks, plot only their PLC component based on undercorrected breccias (Figure 5), and state:

The crucial point is that the siderophile element abundances in Earth's upper mantle are uniquely terrestrial; there is no way in which they can be the product of cosmochemical metal-silicate fractionations in the solar nebula prior to accretion. The similarity between lunar and terrestrial siderophile element abundance patterns is thus of profound genetic importance, clearly implying that the moon was derived from the Earth's upper mantle after the Earth's core had segregated.

It is instructive to reinstate pristine highland rocks (bottom panel). This figure speaks for itself.

5. Reality of Ancient Meteoritic Groups

With commendable candor, Delano and Ringwood (1978b, c) state that their inference of a large, indigenous siderophile component, "if verified, would not only invalidate any previous claims for identification of distinct meteoritic projectiles, but would also have important implications for the origin of the Moon." There is no need to launch into a

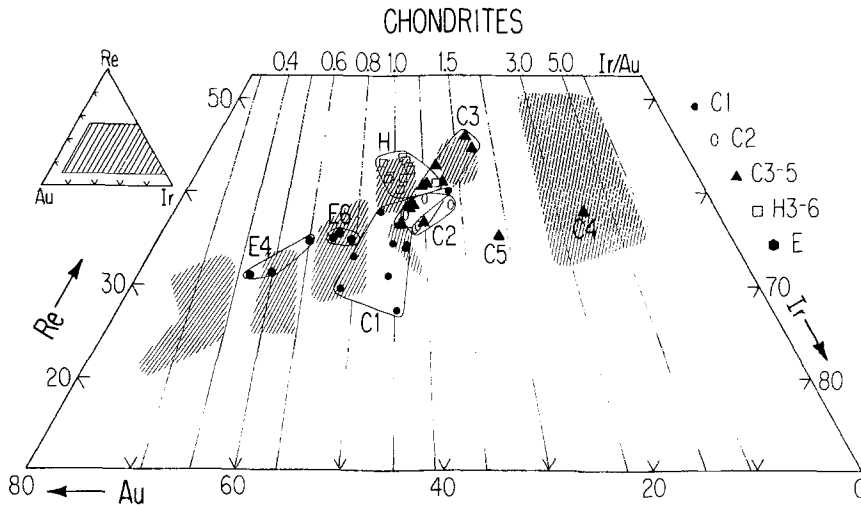


Fig. 7. Chondrites (Hertogen *et al.*, 1977) show substantial variations in Ir/Au ratio, though over a smaller range than ancient meteoritic groups, and so a meteoritic correction based on a single Ir/Au ratio (Delano and Ringwood, 1978b, c) is not justified. E-chondrites, in particular, have Ir/Au ratios as low as some of the lunar groups, and it therefore seems unnecessary to invoke ad hoc processes on the lunar surface to explain these low ratios. Note that chondrite classes are less well resolved, and often no less distended, than are lunar meteoritic groups.

full-fledged defense of our ancient meteoritic groups, because verification of the Delano–Ringwood hypothesis does not seem imminent. Nonetheless, it may be useful to cite a few relevant facts.

(1) Our method of characterizing meteoritic projectiles from their chemical signature in crater materials has been tested against ‘ground truth’ at Meteor and Wabar craters, where the projectile is known (Morgan *et al.*, 1975b). Further confirmation was obtained at East Clearwater crater, where our identification of a C1 chondrite (Palme *et al.*, 1978) was subsequently confirmed by a mixing model for the impact melt (Grieve, 1978).

(2) The reality of the groups recognized on ternary diagrams has been confirmed by 3 objective statistical methods: cluster, factor, and discriminant analysis (Higuchi and Morgan, 1975; Gros *et al.*, 1976).

(3) The ancient meteoritic groups tend to cluster at specific landing sites; thus Apollo 16 samples dominate in Groups 1H and 7, and Apollo 17 samples, in Groups 2 and 3 (Figure 6). This would not be so if the abundance pattern were governed by indigenous siderophiles or by redistributed meteoritic siderophiles, as contended by Delano and Ringwood (1978b, c).

Ringwood argued at the 9th Lunar Science Conference that the distribution in Figure 6 actually is a continuous mixture of 2 components, rich in Ir–Re and Au–Ni, respectively; and that our groups therefore are artifacts. I agree that the groups are not well separated in a ternary diagram, especially if hybrid samples such as glasses and soil separates are included. But neither are chondrites (Figure 7), and by Ringwood’s logic, one

APOLLO 16 HIGHLANDS

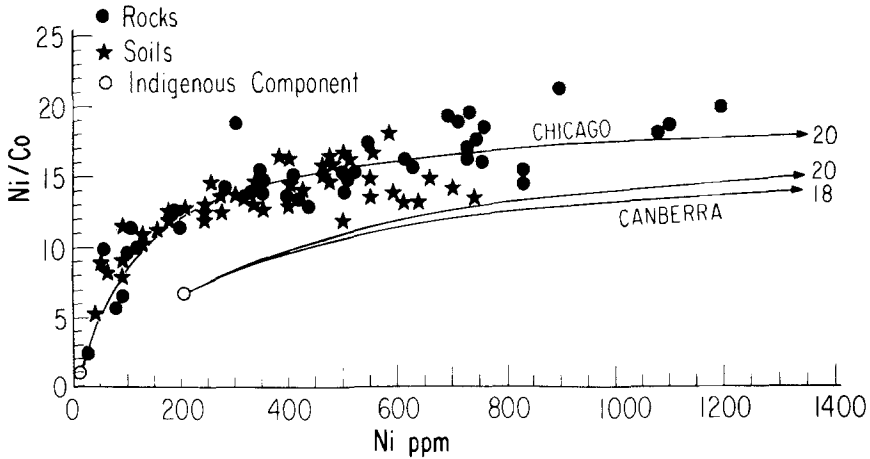


Fig. 8. Test of the claim of Delano and Ringwood (1978a, b, c) that Apollo 16 samples are binary mixtures of a meteoritic component of high Ni content (> 800 ppm) and $\text{Ni/Co} = 18\text{--}20$, and an indigenous component similar to mare basalts ($\text{Ni} = 150$, $\text{Co} = 45$ ppm) or their PLC component ($\text{Ni} = 200$ ppm, $\text{Co} = 30$ ppm). None of the experimental points fall on the 'Canberra' mixing lines between PLC and meteoritic components of $\text{Ni/Co} = 18$ or 20 . On the other hand, it appears that the 'Chicago' mixing line, with the same meteoritic component but much lower indigenous siderophiles ($\text{Ni} = 7$ ppm, $\text{Co} = 7$ ppm) gives a slightly better fit to the data. The Apollo 16 data were replotted without change from Delano and Ringwood (1978a, b, c).

would thus have to conclude that all chondrites are a single, continuous class. Actually, the proper lesson to be learned from these figures is that Au, Re, and Ir are not very good discriminants even for manifestly different meteorite classes, and that additional criteria (such as 4–6 dimensional cluster analysis) are needed to resolve overlapping groups. This has been an abiding concern in our lunar work, as our papers attest.

6. Ni/Co Systematics

Delano and Ringwood (1978a, b, c) conclude from their Ni/Co vs. Ni plot (Figure 8) that highland breccias consist of two components: a meteoritic component of high Ni content (> 800 ppm) and $\text{Ni/Co} = 18\text{--}20$, and an indigenous component of 'much lower' Ni abundance and a Ni/Co ratio between 5 and 10. They neglect, however, to test this proposition by constructing a mixing line between the two end members, and I have therefore done so (Figure 8).*

The lower of the two 'Canberra' lines uses as endmembers the PLC component ($\text{Ni} = 200$ ppm, $\text{Co} = 30$ ppm) and the Delano–Ringwood 'primordial' meteoritic component

* It would have been preferable to replot the graph as Co/Ni vs. $1/\text{Ni}$, because a mixing line would then be a straight line. However, it seemed best to keep manipulation to a minimum.

of Ni/Co = 18. Inexplicably, they prefer this arbitrary ratio to the H-chondrite or CI chondrite ratios of 20 and 21, and I have therefore calculated a second Canberra line for a ratio of 20. The 'Chicago' line, on the other hand, uses 7 ppm Ni and 7 ppm Co for the indigenous component, and Ni/Co = 20 for the ancient meteoritic component. (For consistency with Delano and Ringwood, I have resolved the data in terms of two fixed endmembers, although both the meteoritic and indigenous components are variable. Because Co is not measured at Chicago, the Co value was found by trial and error.)

This figure, too, speaks for itself.

7. The Tungsten Paradox

Dreibus *et al.* (1977) and Rammensee and Wänke (1977) have drawn attention to the curious fact that W is depleted by the same factor of 19 (relative to chondrites) in the Earth, Moon, and eucrite parent body. Laboratory data show that the distribution coefficient of W ($= D_W$) between Ni-Fe and two terrestrial rocks (tholeiite and olivine-nephelinite) is ≤ 40 at temperatures of 1300° , indicating that a nickel-iron core of some 26% would be needed to produce the observed 19-fold depletion. Accordingly, these authors conclude that the depletion took place within the Earth, and that the Moon and eucrite parent body were spun off the Earth after core formation.

Delano and Ringwood (1978b, c) approvingly cite the first two-thirds of this conclusion, but discreetly omit the last one-third: the eucrite parent body. Later on they argue, on the basis of oxygen isotopes, that the eucrite parent body must have a separate origin, but they do not explain why it, too, is depleted in W by the same factor of 19. They also have no qualms about using the 1-atm D 's for the Earth, though elsewhere in their paper they chastise Morgan *et al.* (1974) for applying 1-atm D 's for Au (Kimura *et al.*, 1974) to the Moon, and proclaim that the abundance pattern in the Earth's mantle (and the PLC magma) is due to:

(1) Large changes in metal/silicate partition coefficients caused by the high pressures deep within the Earth.

(2) The presence of large quantities of an element with a low atomic weight, such as oxygen, within the segregating core, which caused large changes in metal/silicate partition coefficients.

Apparently these factors need to be invoked only when the low-pressure data do not fit. Perhaps Delano and Ringwood confuse Procrustes' axe with Occam's Razor.

The identical depletion of W in at least 2 planets (Wänke *et al.*, 1973) is an important observation, and Rammensee and Wänke (1977) deserve much credit for providing both the basic data and a first-order analysis of the problem. However, other alternatives remain to be explored. Rammensee and Wänke (1977) assume that metal-silicate fractionation took place at solidus temperatures (1300°C), but since the process must have involved coalescence of metal particles, temperatures of $1500\text{--}1550^\circ$, near the liquidus, actually would be more realistic. At these temperatures, D_W would be 4–5 times higher (Rammensee and Wänke, 1977). Further differences might be expected from the presence

of P, C, and other minor elements in the metal, from a more exact thermodynamic treatment of the silicate phase, etc. There is no assurance that these factors will explain the constant depletion factor of 19 in 3 planets, but since this constancy itself is well established only for the Moon, Delano and Ringwood's exuberant pronouncement of a 'staggering dilemma' would seem slightly premature.

8. Comments on the Reply

In their Reply, Delano and Ringwood (1978d) have provided useful clarifications of their ideas, especially on pristine lunar rocks. However, they have also made several questionable statements about the Chicago work.

Dispersion of siderophiles. We have never tried to hide either the large dispersion of the siderophiles or their poor correlation with Rb and Cs. But in our own plots of these data, we find a smaller range for the same elements, e.g., 12× rather than 200× for Re and 30× rather than 100× for Ni (Figure 3 of Gros *et al.*, 1976; Figure 3 of this paper).

'Average' abundance of Ir, Au, Ni, Te? Contrary to the Appendix' claim, Gros *et al.* calculated the abundances of these elements from the regressions in their Table 2, not from "their average value in 'pristine' highland rocks".

Ge-Rb, Cs correlation. Delano and Ringwood accuse Gros *et al.* (1976) of omitting 9 samples and plotting 5 spurious ones in their Ge-Cs diagram, and present a new figure showing a much broader spread (Figure 3 of the Reply). Let us compare this figure with an updated version of that from Gros *et al.* (1976), including 5 later Apollo 16 results from Hertogen *et al.* (1977) and one other (Figure 9).

The 5 'spurious' samples include 15256 and 15475, which were designated 'non-mare' basalts by LSAPT and LSPET, and have never been officially demoted to mare basalts. Whatever their true nature, they can be deleted with little harm to the correlation. The remaining 'spurious' samples are 3 of the 4 subsamples of 15405 in the upper right (Ge = 50–160 ppb). Only the topmost of these 4 subsamples technically qualified as meteorite-free (Ir = 0.006 ppb), but since the other three had low enough Ir contents to ensure that most of the Ge was indigenous (0.34, 1.64, 1.28 ppb), it seemed fair to include these samples, to improve statistics at the high-alkali end of the correlation. Yet the correlation does not hinge entirely on 15405; soil separate 15272,9,22 falls close to the correlation line, and if this composite sample is corrected for 60% mare basalt contamination (Morgan *et al.*, 1973), it moves virtually on top of the lowest of the 15405 samples.

Of the 9 omitted samples, we rejected 3 Tl-rich anorthosites for "extraneous enrichment in volatiles . . . , presumably by fumarolic volcanism" (p. 2407 of Gros *et al.*, 1976). I do not understand why anyone should seek to reinstate these samples, considering that Tl-enrichment is *prima facie* evidence for fumarolic volcanism. The 5 Tl-rich anorthosites [including 2 new ones from Hertogen *et al.* (1977)] are systematically higher in Ge than the 5 Tl-poor anorthosites: 2.3 to 82 ppb (mean 22 ppb) vs. 1.4 to 2.7 ppb.

“this specific claim has never before been made in any of Anders’ publications . . .”, there exist equivalent statements in the Chicago group’s earlier papers (Morgan *et al.*, 1972, p. 1387; Krähenbühl *et al.*, 1973, p. 1244; Morgan *et al.*, 1974, p. 1709; Higuchi and Morgan, 1975, pp. 1627–1641; etc.).

“*Anders has chosen not to plot their data from rock fragments separated from the lunar soils*”. Indeed he hasn’t. The ‘rock fragments’ were not single rocklets, but composites of some 10–20 particles hand-picked from 1–2 mm lunar soils according to color and texture, to keep us occupied during the long wait for rock samples from the LRL (Morgan *et al.*, 1972, 1973; Krähenbühl *et al.*, 1973; Ganapathy *et al.*, 1974 a, b). Despite the skill of our petrographer colleagues who did the picking, there always was a danger that these composites might be heterogeneous, as shown for one important case by Morgan *et al.* (1973). To express this lower reliability, we represented soil separates by smaller symbols on our graphs (Ganapathy *et al.*, 1973 and later papers). Starting in 1975 (Higuchi and Morgan, 1975), we deleted soil separates altogether, basing our classification only on rocks, but from time to time reexamined our older data on soil separates, to see how well they fit. In our most recent effort (Figure 2 of Hertogen *et al.*, 1977), we found that 12 out of 18 soil separates fell inside the group boundaries, whereas 6 others fell outside. But of the 12 ostensible matches, 9 could be fortuitous, because the samples lay near mixing lines joining the most extreme groups.

Lacking objective criteria for distinguishing fortuitous from true matches, we have chosen to reject soil separates altogether, except for (1) single rocklets, (2) composites from sites where large highland rocks are scarce (Apollo 11 and 12, and Luna 20) and (3) composites that are meteorite-free or have extreme Ir/Au ratios and hence cannot be mixtures of other groups (e.g., 67602,14-3 in Group 7). I am genuinely sorry that our extensive data on composites go unused under these criteria, but until someone comes up with an objective, after the fact criterion for distinguishing pure-bred from hybrid composites, I see no other choice. The story of the meteoritic component in the lunar highlands is hard enough to decipher in the rocks, without the additional complication of reassembling Humpty-Dumpty from soil fragments.

“*Genetically related subsamples of the same sample or boulder*”. Delano and Ringwood want to eliminate ‘redundancies’ by averaging all such samples of identical Ir/Re/Au/Ni unless petrographic or age data show that they are not cogenetic. But by this criterion, none should be so averaged. Because of the very labor-intensive nature of our analyses, we do not analyze multiple samples from the same rock *unless* petrographic evidence shows them to be different.

Rhenium. Delano and Ringwood compare Re abundances in lunar mare and terrestrial basalts in an indirect way, based on the *continental* basalt BCR-1. But Re analyses of two *oceanic* basalts have been published by Kimura *et al.* (1974): 1154 (1.13 ppb) and V25-1-T9 (1.79 ppb). They seem representative, being similar to additional unpublished analyses by Hertogen *et al.*

Basalt 1154 contains 5.59 ppm La, or 15× the abundance of the Leedey chondrite. Thus, if anything, its Re content should be scaled up by a factor 20/15 for comparison

with mare basalts. The abundance of Re in lunar mare basalts was given as 0.01 ppb by Ringwood and Kesson (1977), representing the mean of 6 Apollo 12 basalts (0.020 ppb; Lovering and Hughes, 1971) and 6 Apollo 15 basalts (0.0043 ± 0.0016 ppb; Ganapathy *et al.*, 1973). It seems likely that the lower value is representative for Apollo 12 as well, because a recent analysis of the mare basalt fragment 12033, 390 gave ≤ 0.003 ppb Re; Janssens *et al.*, 1978). Anyhow, the depletion of Re in lunar basalts thus is not merely $12\times$, but at least $1.13/0.01 = 110\times$, and probably as high as $1.05/0.0043 = 240\times$ (Table I).

Acknowledgement

I am indebted to Marie-Josée Janssens and Jane Crabb for some of the graphs, and to Jonathan Whipple for assistance in meeting editorial deadlines. This work was supported in part by NASA Grant NGL 14-001-167.

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