COMMENT

Geochemistry of grain-size fractions of Luna 24 soil: Comments on "The lunar regolith: Chemistry and petrology of Luna 24 grain size fractions" by J. C. Laul, O. D. Rode, S. B. Simon, and J. J. Papike

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A CHARACTERISTIC FEATURE of lunar regolith from mare sites is the enrichment of the very finest material (usually <10 or <20 μm sieve fractions) in elements associated with plagioclase (Al) and a corresponding depletion in elements associated with mafic mineral phases (Fe, Sc) compared to coarser fractions (DUNCAN et al., 1974; KOROTEV, 1976; LAUL and PAPIKE, 1980b). The finest material is also usually enriched in ITEs (incompatible trace elements), particularly the light REEs (rare earth elements). In a recent paper by LAUL et al. (1987), compositional data for grain-size fractions as small as <1 μm are presented for the first time. The data, which are for two samples of regolith returned by the Soviet Luna 24 mission to Mare Crisium, show that ultrafine material has the same pattern of enrichment and depletion observed in other soils from the maria but to an even greater extent.

Two mechanisms, differential comminution and mixing, have been offered to explain why lithophile element concentrations in the lunar regolith vary with grain size (EVENSEN, 1973; KOROTEV, 1976; DEVINE et al., 1982). A stated conclusion of LAUL et al. (1987) is that their data "provide direct evidence for differential comminution." LAUL et al. (1987) do not, however, exclude or even address the mixing mechanism, which also is qualitatively supported by their data. A preference for the differential comminution mechanism has been stated in other papers by some of these authors (PAPIKE et al., 1981, 1982; LAUL et al., 1982; SMITH et al., 1985), but in none of these papers have the two mechanisms been tested against each other and the mixing mechanism shown to be unimportant. In the large number of papers on the chemical composition of grain-size fractions of lunar soils produced by J. C. Laul, J. J. Papike, and coworkers (including a comprehensive review paper; PAPIKE et al., 1982), the authors leave the impression that the cause of the compositional variation with grain size is well understood and that differential comminution is largely responsible. In this paper I argue that this impression is inaccurate because, in the particular case of soils composed predominantly of mare basalt such as those at Luna 24, both mechanisms account qualitatively for the observed trends and data do not exist to justify dismissing either mechanism as unimportant.

I also suggest that conclusions regarding meteoritic material in LAUL et al. (1987) based on data for Ni (and possibly other siderophile elements) may be invalid because some of the samples are contaminated with a Ni-rich material.

LITHOPHILE ELEMENTS: DIFFERENTIAL COMMINUTION OR MIXING?

In the lunar literature, the term "differential comminution" (which was first used in this context by EVENSEN et al., 1973) is used to describe the effect that when a crystalline rock such as a mare basalt is reduced to fine-grained material by impact, the resulting grain-size distributions of the various minerals are not all identical. This is a consequence of differences in mean grain size of the various minerals in the target rock as well as differences in mechanical properties of the minerals. In the comminuted rock, a variation in modal abundance of the minerals and mesostasis with grain size will lead to variation in chemical composition with grain size. That differential comminution actually occurs during comminution of lunar rocks by meteorite impact has never been unambiguously proven, but laboratory simulations suggest that for a target of mare basalt it is a likely consequence (HASKIN and KOROTEV, 1977; HÖRZ et al., 1984; KOROTEV et al., 1986). In these experiments the finest grain-size fractions are enriched in ITEs, presumably as a result of preferential concentration of intergranular mesostasis. They are also enriched in Al and depleted in Fe compared to the whole rock as a result of preferential accumulation of plagioclase. The observed variation in chemical composition with grain size is qualitatively similar to that observed in soils from mare sites, suggesting that differential comminution plays an important role in lunar soils.

Quantitative evaluation of the importance of differential comminution in lunar regolith is difficult, however, because the variations observed can also be explained qualitatively by another mechanism. All returned soil from mare areas contain some material from the highlands (RHODES, 1977; PAPIKE et al., 1982), and the chemical signature of this nonmare material is also an enrichment in Al and ITEs, particularly the light REEs. Typical highlands material has two to three times the Al concentration of mare basalt. Both the Apollo 15 and 17 sites, where the grain-size effect is well

documented, were at the interface of the maria and highlands, and soils of both mare and highlands affinities were collected at these sites. The highlands soils are richer in Al and ITEs than the mare soils. However, chemical mass balance models show that even the most Fe-rich soils from these sites contain some nonmare material (14% at Apollo 15 and about 20% at Apollo 17; KOROTEV, 1987; RHODES et al., 1974). Even soils from the Apollo 11 site, which, like the Luna 24 site, is distant from the nearest outcrop of highlands material, contain more than 25% nonmare material (LAUL and PAPIKE, 1980b). Fragments of anorthosite and glasses of KREEP composition have been identified in the Apollo 11 regolith (e.g., LAUL et al., 1983). If the nonmare component of these soils has a finer grain-size distribution than the mare component, then the finer grain-size fractions will have higher concentrations of ITEs and Al than will the coarser fractions. Thus, both proposed mechanisms, differential comminution and mixing, can account qualitatively for the variation in chemical composition with grain size that is observed in mare soils. (The "exotic component" concept of EVENSEN et al., 1973, 1974, and DEVINE et al., 1982, in which there is a moon-wide, fine-grained component rich in ITEs and radiogenic Sr, is just a special case of the mixing mechanism.)

Mixing is a necessary consequence of impact and its effects cannot be ignored. Any given sample of lunar regolith is a mixture of various lithologies, many of which are compositionally distinct. There is no reason to expect that the various lithologies all have the same grain-size distributions. Three examples in which they do not are discussed below.

1) At Apollo 17, the orange glass soil (sample 74220) consists almost entirely of pyroclastic glass beads with a narrow peak in grain-size distribution at about 40 μm, which is smaller than the mean grain size of most other Apollo 17 soils (MCKAY et al., 1974). This same type of glass is a significant component of all other basaltic soils at the site (10–25%, RHODES et al., 1974). A characteristic feature of the orange glass is its high concentration of Cr compared to other soil components. As a consequence, Cr concentrations are systematically greater in the 20–90 μm grain-size fractions of Apollo 17 mare soils than in the <20 μm or >90 μm fractions. Thus, Cr concentrations vary with grain size as a result of mixing of components with different grain-size distributions. This variation is reflected in the results of mixing models which show a maximum in orange glass component in the 20–90 μm size fraction (LAUL et al., 1979; LAUL and PAPIKE, 1980a).

2) At Apollo 15, the soils from station 2 on the Apennine Front consist predominantly of fine-grained highlands material. Other soils at Apollo 15 appear to be mixtures of soil such as at station 2 with various lithologies found at the site (mare basalt, KREEP basalt, and green glass). Station 6 soils are compositionally similar to station 2 soils, but have higher concentrations of ITEs. The composition of the <1 mm soils from station 6 can be modeled well as a mixture of about 70% station-2 soil, 10% mare material, and 20% KREEP basalt. The 1–2 mm grain-size fraction of station 6 soil 15270 is enriched by 20–30% in ITEs compared to the <1 mm fraction. This variation in composition with grain size results because the KREEP basalt component of the soil, which has

high ITE concentrations, is coarser grained than the "station 2" component. Evidence for this is (1) station 6 soils have larger mean grain sizes than station 2 soils (MORRIS et al., 1983) and (2) about 30% of the 1–2 mm particles examined in 15270 are lithic or glass fragments with KREEP basalt compositions. (Data and modeling for this example are from KOROTEV, 1987.)

The <20 μm fractions of both the stations 2 and 6 soils are enriched in ITEs and Al and depleted in Fe and Sc compared to the 20–1000 μm fractions. This pattern may result from differential comminution of highlands material or be some kind of mixing effect. Although LAUL and PAPIKE (1980b) attribute the pattern to mixing (specifically, to a greater fraction of anorthosite and KREEP in the <20 μm fraction), they attribute the same pattern of enrichment and depletion at Luna 24 to differential comminution, with no mention of why mixing is not also operative there.

3) Apollo 16 is the only Apollo site where the regolith may be nearly devoid of mare basalt. Compositional variation among Apollo 16 soils results mainly from mixing of fine-grained mature soil, which is relatively mafic and rich in ITEs, with coarse-grained immature soil, which is felsic and poor in ITEs (KOROTEV, 1981, 1989). Most soils at the surface of the site contain a large fraction of the fine-grained mafic endmember. The coarse-grained felsic soils are found mainly around North Ray crater and in the 60009/10 core. The mafic soils (which contain a large component of mafic impact melt rocks) overlay more felsic lithologies, but local cratering events have brought some felsic material to the surface (STÖFFLER et al., 1985). For many soils, compositional differences between coarse and fine grain-size fractions are usually not large (BOYNTON et al., 1976; BLANCHARD et al., 1976; KOROTEV, 1981). Soils in which a large compositional variation with grain size is observed are usually those with a large component of coarse-grained felsic material. This is demonstrated by data for soils from the 60009/10 core (Fig. 1). Concentrations of Sc and Sm remain more constant in the finer fractions than in the coarser fractions with increasing mean grain size of the soil because the finer fraction consists mainly of the mafic ITE-rich soil. As the amount of coarse-grained felsic material in the soil increases, the mean grain size increases and Sc and Sm concentrations decrease, but relatively more so in the coarser fraction.

Although they do not provide any argument against the simple mixing explanation, PAPIKE et al. (1982) prefer to explain the Apollo 16 data with a differential comminution mechanism involving concentration of mesostasis from melt rocks and interstitial pyroxene from anorthositic rocks into the finer fractions. Such a process may occur, but it would be difficult to assess the importance of this effect in soils where the mixing effect is so strong.

It is reasonable to expect that the nonmare components of soils formed on the maria are fine grained. Fine-grained regolith existed about 4 Ga ago at the Apollo 16 site (MCKAY et al., 1986). If this was generally true for the early lunar surface, it is likely that most maria are underlain by an old surface of fine-grained, feldspathic regolith. RHODES (1977) has argued that most of the nonmare material in mare regolith located distant from the mare-highlands boundary (e.g.,
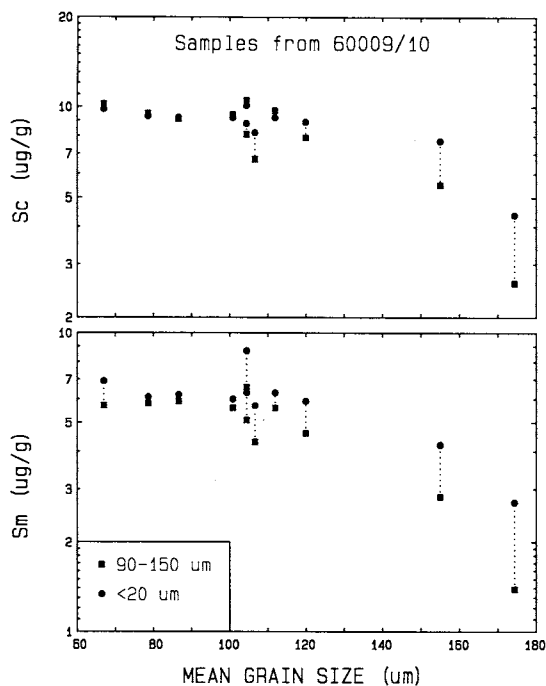
FIG. 1. Sc and Sm concentrations in two grain-size fractions (BLANCHARD et al., 1976, 1977) as a function of mean grain size (sub-centimeter data of McKAY et al., 1976, 1977) for 11 soils from the 60009/10 core.

Apollo 11) derives not from lateral mixing but from vertical mixing by impacts that penetrate the thin basalt flows. Thus, the highlands components of the mare soils may always have been relatively fine-grained.

If differential comminution and mixing of chemically distinct components with different grain-size distributions both lead to the same pattern of compositional variation with grain size, how do we determine whether one of the processes is the major cause of the variation? EVENSEN et al. (1973) first noted that compositional variation with grain size could be explained by either process, but argued that the variation in Rb and Sr isotopes strongly suggested that two-component mixing was the dominant process in Apollo 15 soil 15531. The only work specifically addressing the question quantitatively is that of KOROTEV (1977) for mare soils from Apollo 17. In that study, grain-size fractions were modeled as mixtures of local rock types as well as mineral phases (plus mesostasis) separated from a mare basalt. KOROTEV (1977) concluded that most of the compositional differences between the <20 μm and 90-150 μm fractions resulted from a greater proportion of highlands material in the <20 μm fraction (typically 30%) compared to the 90-150 μm fraction (typically 10%). However, the least-squares fit for the <20 μm fraction was not as good as for the 90-150 μm fraction, suggesting that some process other than mixing was also important. A large improvement in the fit of the model was obtained by varying the mineral proportions of the basalt component with grain size. The best fit was obtained when the basalt component of the <20 μm fraction contained 28% plagioclase (by weight) compared to 18% in the 90-150 μm fraction. These results led KOROTEV (1977) to conclude that mixing

was the predominant cause of the variation but that differential comminution was an important secondary cause.

Although Laul, Papike, and coworkers have subsequently produced a large amount of compositional data on soils from many different landing sites, they have never disputed the conclusion of KOROTEV (1977) with quantitative arguments in which one explanation was tested against the other. In their earlier papers, the grain-size effect is clearly attributed to mixing (LAUL et al., 1978a, 1979; LAUL and PAPIKE, 1980a; KEMPA et al., 1980), whereas in later papers differential comminution is stated as the more likely (or only) cause of the effect (PAPIKE et al., 1981, 1982; LAUL et al., 1982, 1987; SMITH et al., 1985). In transitional papers both explanations are offered (LAUL and PAPIKE, 1980b; LAUL et al., 1981; DEVINE et al., 1982). No data or arguments are presented in this series of papers to justify the shift in preference. A petrographical study of <10 μm fractions of soils from four missions was unable to eliminate mixing as the cause of the effect (DEVINE et al., 1982). The application of compositional mass-balance models ("mixing models") to grain size fractions using local rock types as components implicitly assumes that mixing is the cause of variation in composition with grain size. In every case in which the compositions of mare soils have been modeled in this way, results have shown that the variation in composition with grain size is consistent with a smaller fraction of mare material and a larger fraction of nonmare material in the finest grain-size fractions (KOROTEV, 1976; LAUL et al., 1979, 1981; LAUL and PAPIKE, 1980a,b; PAPIKE et al., 1982; SMITH et al., 1985). Curiously, even though the models show that mixing can explain the observed effect, a paper in which mixing models are presented concludes instead with a statement that differential comminution is the cause (SMITH et al., 1985).

It is particularly difficult at Luna 24 to separate quantitatively the effects of differential comminution from those of mixing because the quantity of material available for study is so small and the rock types of the Luna 24 site are poorly known. Nevertheless, soils from Luna 24 are important in our efforts to resolve the relative importance of these two processes because the site geology and data are unique in several respects. Among soils from mare sites, the Luna 24 soils have the lowest ITE concentrations. However, ITE-rich, noritic breccias from the highlands are a component of the soils (BLANCHARD et al., 1978). Both McKAY et al. (1978) and LAUL et al. (1987) found that the grain-size distribution of Luna 24 soils was bimodal and attributed this to mixing of a fine-grained soil and a coarser one. Thus, qualitatively, the Luna 24 data can be explained by a component of ITE-rich, highlands breccia that is relatively more abundant in the finer grain-size fractions (BLANCHARD et al., 1978). Quantitative modeling must await a better characterization of the rock types of the Luna 24 regolith.

Two special arguments in favor of differential comminution at Luna 24 can be made, although neither was noted by LAUL et al. (1987). First, the fraction of highlands material in Luna 24 soils is considerably lower than that of soils from other mare sites (<5%, BLANCHARD et al., 1978). We might, therefore, expect the effects of mixing of highland material to be minimal there, yet the relative enrichment of ITEs in the finest grain-size fractions of Luna 24 soil is considerably

greater than that of any other soil from the maria (e.g., Fig. 16, PAPIKE et al., 1982). A better argument that some process other than simple mixing must be involved is given by BLANCHARD et al. (1978), namely, that on a Rb-Sr isochron plot (NYQUIST et al., 1978) the <20 μm fraction does not fall on a mixing line between the bulk soil and ITE-rich components such as noritic melt rocks from Apollo 17.

Thus, although differential comminution may contribute significantly to the variation in chemical composition with grain size observed in the Luna 24 regolith, the statement of LAUL et al. (1987) that their data "provide evidence in favor of comminution . . ." is not valid because such a conclusion requires a compelling argument that mixing effects are inconsequential and no such argument is given. To the contrary, LAUL et al. (1987) show that the Luna 24 soil is a mixture of coarse-grained and fine-grained material. This is likely to result in variations in composition with grain size such as those observed. They provide no evidence that differential comminution, such as that observed in laboratory experiments, is the principal cause of the grain-size effect in Luna 24 soil.

SIDEROPHILE ELEMENTS

In mare soils most of the Ni, Ir, and Au derive from impact of micrometeorites, which are primarily carbonaceous chondrites (ANDERS et al., 1973; WASSON et al., 1975). The variation of siderophile element concentrations with grain size is potentially important because it can tell us how meteoritic material is distributed during regolith evolution. LAUL et al. (1987) state that, "Since the comminution process involves impacts by numerous micrometeorites, finer soil fractions are expected to be enriched in meteoritic siderophile elements." Why should we expect Ni and other siderophile elements to be concentrated in the finest grain-size fractions? Enrichment of siderophile elements in coarser size fractions has been reported in Apollo 16 soil (BOYNTON et al., 1976). Also, as the authors note, micrometeorite impact is a constructional as well as destructional process because agglutinate particles are formed from fusion of finer-grained material during impact (MCKAY et al., 1974; PAPIKE et al., 1982; LAUL et al., 1987). Ni concentrations in the <250 μm fines at Luna 24 are about 140 μg/g, while agglutinate particles separated from soil have about 180 μg/g Ni (LAUL et al., 1978b; BLANCHARD et al., 1978), so it is unlikely that the finest grain-size fractions of the soil should have concentrations greatly in excess of this.

Thus, it is unexpected that LAUL et al. (1987) obtain extraordinary enrichment in Ni and moderate enrichment in Co, Ir, and Au in some of the <10 μm size fractions they analyzed. LAUL et al. (1987) attribute the high concentrations of siderophile elements to a large component of meteoritic material and imply that the micrometeorites which cause the comminution of the soil concentrate in the finest fraction. The Ni concentration reported for the <2 μm fraction of 24214 is 80% of the concentration in CI chondrites, implying an extremely large proportion of meteoritic material in this fraction, if the implication is valid.

In some previous studies in which high concentrations of Ni have been observed in the finest grain-size fractions, these enrichments have been dismissed as contamination from the nickel sieves used to obtain the fractions (LAUL et al., 1978a). In the study of LAUL et al. (1987), size fractions below 40 μm in diameter were obtained by sedimentation in acetone, so one might expect minimal problems with contamination, especially since the possibility of contamination is not discussed. However, several quantitative arguments suggest that a large fraction of the Ni in the finest size fractions is not meteoritic but probably results from contamination. (1) The concentrations of Ni in the Ni-rich fractions are much too large compared to those of Co, Ir, and Au for the Ni to be derived solely from carbonaceous chondrites or any other type of meteorite. (2) The mass-weighted sum concentrations of Ni in the two soils (<250 μm grain-size fractions) are 790 and 360 μg/g. These values are high compared to "bulk" samples of Luna 24 soil, i.e., material that has passed through a 250 μm sieve only. The mean value for 13 such samples is 143 μg/g Ni (range: 90–230; BLANCHARD et al., 1978; MA et al., 1978; LAUL et al., 1981, 1987). (3) The <2 μm fraction of 24214 is strongly enriched in Ni and the <2 μm fraction of 24176 is only slightly enriched. This is a reasonable result if particulate contamination is the source of the Ni but an unexpected one if some lunar process is responsible. (4) The difference in Fe concentration between the <2 μm fraction of 24214 (which has 0.87% Ni) and the <2 μm fraction of 24176 (which contains only 0.04% Ni) is 1.6% Fe (Table 3, LAUL et al., 1987). For the other size fractions the results for Fe are much more similar for the two samples. This suggests that the contaminant also contains Fe and that the Fe/Ni ratio is about 2 (=1.6/[0.87 − 0.04]). If the high Ni concentrations result from terrestrial contamination, then the Co, Au, and Ir data are also compromised because these elements are likely impurities in commercial Fe-Ni materials.

Thus, conclusion number 3 of LAUL et al. (1987)—"High concentrations of meteoritic indicator elements (Ni, Au, Ir) in the finer fractions are consistent with the comminution process by impacts with micrometeorites."—is misleading because the high concentrations are also "consistent with" contamination, a likely alternative which was not discussed. As in the case of the lithophile elements, two models are qualitatively supported by the data, one of which (meteoritic source) is favored by the authors even though the other (contamination) is not acceptably dismissed. Because the alternate model is not discussed, the impression is left that the data are adequately explained. For the siderophile elements, however, the favored explanation can be shown to be fallacious when subjected to quantitative scrutiny.

Editorial handling: R. A. Schmitt

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