

tions, in general, than those measured. At the same time it is also probable that the differences in absolute trace-element concentration between the whole rocks and the separated phases are in part due to the presence of interstitial, accessory phases, such as phosphates, alkali feldspar, or glass, that are rich in trace elements. That our separated pyroxene and feldspar may represent early formed material is supported to some extent by the fact that the trace element ratios between these minerals and their respective whole rocks are close to those expected on the basis of measured trace-element partition coefficients (5, 9) between such minerals and the liquids from which they are crystallizing. The consistency of the mineral to whole-rock trace element ratios, in spite of the differences between the whole-rock concentrations themselves, tends to indicate that the separates were fairly pure. In the case of the ilmenite separates the consistency is problematic; we did not expect such high trace element concentrations in this phase, particularly the high concentration of the large Rb cation. Perhaps Rb is substituting for oxygen, or possibly the high trace element concentrations reflect late crystallization of the separated ilmenites. Perhaps the consistency is fortuitous and we are seeing contamination by late phases, a possibility favored to some extent by the complementary nature of the ilmenite and feldspar Sr, Rb, and K concentrations, for example. These questions cannot be answered at the present time. A matter on which the mineral data do throw some light is the redox state of the lunar samples. The partitioning of Eu (an element that differs from the other trivalent rare-earths in showing significant natural stability in the divalent state) between two phases, is a function of a number of parameters, including redox conditions (10). Measured partition coefficients (9) indicate that augite will contain more trivalent Eu than coexisting calcic plagioclase, but that the plagioclase will contain considerably more divalent Eu than the augite. Hence, the ratio of total Eu between feldspar and pyroxene will be a function of the divalent to trivalent Eu ratio of the system which in turn is a function of the redox conditions. The ratio of Eu between plagioclase and pyroxene in the lunar rocks is higher than between comparable terrestrial feldspar and pyroxene, and this indicates a higher divalent to trivalent Eu ratio than on earth, reflecting more reducing condi-

tions. Finally, it is interesting that the relative trace-element concentrations of 44 plagioclase, and of 44 pyroxene, are very similar to those of pyroxene and plagioclase separated from the Juvinas achondrite (4).

The overall similarity in the trace-element characteristics of the breccias, the soil, and two of our igneous rocks, in particular, is apparent from Table 1 and Fig. 1. It appears that the process, probably meteorite impact, that produced the fragmental rocks, did not greatly alter their trace element concentrations from those in their presumably igneous parent rocks. The differences that exist between the two similar igneous rocks, the breccias and the soil, most noticeably the differences in absolute concentrations and the small differences in the ratios of Sr and of Eu to the other elements, are not those expected for a volatilization process (the similarity between the < 400 mesh fraction and the bulk soil is pertinent in this respect) but rather are just those observed for igneous differentiation involving feldspar (8). It appears that these lunar rocks might well represent liquids produced by fractional crystallization or varying degrees of partial fusion. If these rocks were derived from material with chondritic trace elements, then some process must be invoked to produce enrichment of the other trace elements relative to Rb and K. An igneous process does not seem likely; perhaps volatilization is required. In contrast, the trace element characteristics of these lunar rocks, including Rb/Sr, K/Rb, K/Ba, and the negative Eu anomaly, when considered in the light of appropriate partition co-

efficient data, appear to be consistent with these rocks being the result of limited partial fusion of source material similar to the brecciated eucrites. It has been suggested that these meteorites are of lunar origin (11); the lunar whole rock major element analyses (1, 12) and our trace element analyses seem not inconsistent with this hypothesis.

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Total Carbon and Nitrogen Abundances in Lunar Samples

Abstract. *Total carbon and nitrogen abundances were determined by using combustion-gas chromatographic techniques. Weighted mean analyses for bulk fines were 225 and 140 ppm total carbon and 150 and 100 ppm total nitrogen. Total carbon in fine breccia was 230 ppm; in coarse breccia, 100 ppm; in fine-grained rock, 70 ppm; and in medium-grained rock, 64 ppm. The finest sieved fraction had the highest carbon content. Total nitrogen in fine breccia was 125 ppm; in coarse breccia, 100 ppm; in fine-grained rock, 115 ppm; and in medium-grained rock, 30 ppm. The total carbon and nitrogen fines appear to be mixtures of indigenous lunar material together with meteoritic and solar wind components.*

Total carbon and nitrogen contents were determined in Apollo 11 lunar samples with analytical techniques that were developed for these elements in meteorites (1). These elements are important because of their crucial role as organogenic elements. Their abun-

dances give an indication of the oxidation-reduction conditions and, owing to their volatility, of thermal conditions near the lunar surface.

For the determination of total carbon, samples were burned in a flowing oxygen atmosphere at approximately

Table 1. Total carbon in Apollo 11 lunar rocks. The symbol Δ denotes results obtained by heating for 24 hours at 300°C in a flowing helium atmosphere.

Sample weight (g)	Total carbon ($\mu\text{g/g}$)	Weighted mean ($\mu\text{g/g}$)
<i>Fines, sample 86,A</i>		
0.3433	130 $\pm$ 4	...
0.3094	162 $\pm$ 6	...
0.0782 Δ	160 $\pm$ 25 Δ	...
0.1555 Δ	122 $\pm$ 13 Δ	...
(0.8864)	...	142 $\pm$ 10
<i>Fines, sample 86,B</i>		
0.1080	190 $\pm$ 20	...
0.2586	181 $\pm$ 8	...
0.2565	109 $\pm$ 8	...
0.3482	353 $\pm$ 6	...
(0.9713)	...	226 $\pm$ 10
<i>"C" breccia, sample 2,54</i>		
0.2831	228 $\pm$ 7	...
0.2874	231 $\pm$ 7	...
(0.5705)	...	230 $\pm$ 7
<i>"C-B" coarse breccia, sample 44,37</i>		
0.2093	108 $\pm$ 9	...
0.3677	76 $\pm$ 6	...
0.1058	180 $\pm$ 18	...
(0.6828)	...	102 $\pm$ 10
<i>"A" basalt, sample 49,23</i>		
0.2268	68 $\pm$ 9	...
0.3464	71 $\pm$ 6	...
(0.5732)	...	70 $\pm$ 8
<i>"B" basalt, sample 50,33</i>		
0.2394	152 $\pm$ 8	...
0.3234	15 $\pm$ 7	...
0.3007	47 $\pm$ 6	...
(0.8635)	...	64 $\pm$ 8

1800°C to form CO₂. After necessary purification and trapping of the effluent gases, the CO₂ was detected by a gas chromatographic technique in a LECO 589-400 low carbon analyzer. Lunar samples were heated to about 2400°C in a graphite crucible in a helium atmosphere to reduce all nitrogen compounds to N₂, which was detected in a LECO Nitrox-6 gas chromatographic analyzer. Differential thermal conductivity was used as the detection method in both systems. National Bureau of Standards low carbon and nitrogen steel standards were used to construct standard analytical curves for both determinations. The combustion-gas chromatographic detection technique determines total carbon and nitrogen but does not discriminate their chemical state in the lunar samples.

The results of the analyses are listed in Table 1 for total carbon and in Table 2 for total nitrogen. No attempt was made to homogenize individual samples, and thus the possibility of contamination was minimized and sample inhomogeneity was tested. Rock samples were crushed by one stroke in a clean diamond mortar. All the rock specimens were crushed quite easily to

a moderately fine material. To guard against contamination, they were not sieved or run through a mechanical splitter.

The precision (indicated as $\pm$ in Tables 1 and 2) is taken from the 90 percent confidence level on the line of regression for the reference standards. Analytical precision of the nitrogen analyses is superior to that for carbon because the difference between the thermal conductivities of nitrogen and helium is greater than that for carbon dioxide and helium.

The carbon and nitrogen abundances show some similarities. Both are highest in the fines samples 86,A and 86,B and in the fine-grained breccia 2,54. The coarse-grained basalt 50,33 contains the lowest concentrations of both carbon and nitrogen. With respect to carbon, the fine-grained basalt 49,23 is similar to the coarse-grained basalt, but its nitrogen content is relatively high. The analyzed sample of rock 44,37 had the macroscopic characteristics of a coarse-grained basalt but was catalogued as a coarse-grained breccia. In terms of both carbon and nitrogen, it is intermediate to the fine-grained breccia 2,54 and the coarse-grained basalt 50,33.

The carbon and nitrogen in Apollo 11 lunar samples may be from four sources: (i) indigenous, (ii) meteorite or comet impact, (iii) solar wind, and (iv) contamination. Contamination may have resulted from the landing of the lunar module, from astronaut activity, or from subsequent laboratory handling. The elements may be indigenous to the moon as elemental carbon, as interstitial atoms of carbon or nitrogen, as organic material, or as inorganic compounds (carbonate, carbides, ammoniacal nitrogen, molecular nitrogen). If the elements were accumulated on the lunar surface by both meteorite impact, solar wind, or both, it may be difficult to distinguish which process was dominant. Both processes are rather catastrophic with respect to the components involved. Meteoritic material arrives with hypervelocity impacts, causing volatilization and atomization of the projectile and part of the target. Solar wind particles arrive as single atoms or parts of atoms.

The total carbon trends seem to support the idea that the values of 70 and 64 $\mu\text{g/g}$ (ppm) found in the basalts are primarily due to indigenous carbon. The close agreement for the two splits of the fine-grained basalt 49,23 indicates

Table 2. Total nitrogen in Apollo 11 lunar rocks. The symbol Δ denotes results obtained by heating for 24 hours at 300°C in a flowing helium atmosphere.

Sample weight (g)	Total nitrogen ($\mu\text{g/g}$)	Weighted mean ($\mu\text{g/g}$)
<i>Fines, sample 86,A</i>		
0.0971	142 $\pm$ 5	...
0.1822	87 $\pm$ 2	...
0.1239	100 $\pm$ 3	...
0.0749 Δ	107 $\pm$ 5 Δ	...
0.1192 Δ	92 $\pm$ 4 Δ	...
(0.5973)	...	102 $\pm$ 4
<i>Fines, sample 86,B</i>		
0.1969	150 $\pm$ 2	...
0.1299	164 $\pm$ 5	...
0.1024	144 $\pm$ 4	...
(0.4292)	...	153 $\pm$ 4
<i>"C" breccia, sample 2,54</i>		
0.0896	113 $\pm$ 5	...
0.1588	135 $\pm$ 4	...
0.1146	119 $\pm$ 4	...
(0.3630)	...	125 $\pm$ 4
<i>"C-B" coarse breccia, sample 44,37</i>		
0.1666	135 $\pm$ 4	...
0.1266	95 $\pm$ 4	...
0.1993	69 $\pm$ 3	...
(0.4925)	...	98 $\pm$ 4
<i>"A" basalt, sample 49,23</i>		
0.1510	85 $\pm$ 3	...
0.1293	35 $\pm$ 4	...
0.1159	247 $\pm$ 5	...
(0.3962)	...	116 $\pm$ 4
<i>"B" basalt, sample 50,33</i>		
0.1888	32 $\pm$ 2	...
0.2067	28 $\pm$ 2	...
(0.3955)	...	30 $\pm$ 2

some degree of homogeneity with respect to the analyzed sample, but the variation between the splits of the coarse-grained basalt 50,33 indicates a more sporadic distribution of carbon among or between the large crystalline phases.

The two fines samples 86,A and 86,B and the fine dark breccia 2,54 are significantly higher in total carbon than are the basalts. The fact that sample 86,A received less handling in the Lunar Receiving Laboratory may account for its lower carbon values. There is no evidence to support this assumption, and the overlap in the range of values in 86,A and 86,B, together with a constant carbon-nitrogen ratio, may indicate a normal sample variation. To investigate the distribution with respect to particle size, a split of sample 86,A was sieved and analyzed for carbon. Material from 60 to 140 mesh had 115 $\pm$ 20 $\mu\text{g/g}$ total carbon; from 140 to 300 mesh, 210 $\pm$ 10 $\mu\text{g/g}$; and minus 300 mesh, 500 $\pm$ 20 $\mu\text{g/g}$. Evidently the carbon is concentrated in the finest-sized material. Microscopic examination of the material larger than 60 mesh, and the 60- to 140-mesh fraction indi-

cated that it is made up primarily of fine-grained basalt, glassy impactite material, and some crystal fragments. Samples 86,A and 86,B may contain different proportions of coarse and fine fragments and, hence, have different total carbon abundances. The fine-grained breccia appears to consist primarily of compacted fines, and it apparently lost no carbon during lithification.

The rare gas analyses reported in the preliminary examination of lunar samples from Apollo 11 (2) also showed enrichment in the fines and breccia. Assuming that all the rare gas content is due to solar wind, some rough calculations may be made to indicate how much of the carbon may be attributed to solar wind. The major unknown in the calculation is a "sticking" or retention factor relating the relative retention on the lunar fines to each of the rare gases. A comparison of Apollo 11 rare gas abundances with Cameron's 1967 (3) table of abundances in the solar system indicates a fractionation in the lunar materials favoring the retention of higher atomic weight gases. If the solar carbon/neon is normalized to the neon in the lunar samples, the solar wind contribution of carbon to the lunar fines is 6 $\mu\text{g/g}$ or about 4 percent of the total carbon. If the carbon abundance is calculated from the krypton abundance, the solar wind contribution is about 50 $\mu\text{g/g}$, and, if it is calculated from xenon, the solar wind contribution would be 300 $\mu\text{g/g}$. Assuming a similar "sticking" factor, the solar wind contribution would be about 5 times lower for nitrogen than for carbon.

A general calculation may also be made to estimate the possible meteoritic and cometary contribution to the lunar fines. The major unknown in this case is the composition of the meteoritic influx. A useful start might be to take material with the composition of carbonaceous chondrites. Such a choice is strengthened by comparing the increase in nickel from the lunar igneous rocks with the lunar breccia and fines as reported in the preliminary examination of samples from Apollo 11 (2). Nickel and carbon both show significant increases, and nickel, like carbon, has a much higher concentration in carbonaceous chondrites than in lunar rocks.

Mason's (4) review of carbonaceous chondrites gives typical concentrations of 2 percent carbon and 1 percent nickel in these meteorites. Using these values to calculate the approximate

contribution of meteoritic extra-lunar material indicates that about 0.5 to 2 percent has been mixed with lunar material. The nitrogen contribution of meteorites is about 10 percent that of carbon and, again, this element does not give an apparent satisfactory material balance. Based on our simplifying assumptions, both solar wind and meteorite influx fail to account for the relatively high nitrogen abundances. They may be explained by the possibility that indigenous nitrogen is high in the lunar rocks, as indicated by the fine-grained basalt 49,23, or that solar wind or meteoritic nitrogen is retained to a greater degree than is carbon.

The possibilities for contamination from nitrogen in returned lunar samples appear to be greater than for carbon. Possible sources are the exhaust gases from the lunar module and the nitrogen atmosphere to which the fresh lunar rocks were exposed at the Lunar Receiving Laboratory.

To see if nitrogen or carbon could be easily desorbed from lunar fines, a split of sample 86,A was heated for 24 hours at 300°C in a flowing helium atmo-

sphere. No carbon or nitrogen appeared to be removed by this treatment. If contamination by atmospheric nitrogen has taken place, it is strongly bonded to the fines material.

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Instrumental Neutron Activation Analyses of Lunar Specimens

Abstract. *Ten Apollo 11 specimens were divided into 24 samples. Sodium contents of 8 diverse specimens cluster tightly about 0.3 percent. Plagioclase separated from sample 10044 contains about 1.09 percent Na; barium is not enriched in the plagioclase separate. Contents of the rare earths are strikingly high, and relative abundances resemble those of calcium-rich achondrites or abyssal basalts but are depleted in Eu by factors of 2 to 3 and in La by about 20 percent. The plagioclase separate is enriched in Eu and pyroxenes and opaque minerals are Eu-depleted. Fine fractions of 10044 are about 20 to 40 percent richer in most rare earths (50 percent for Eu) than coarse fractions, probably because of the presence of small grains in which rare earths are markedly concentrated. "Microgabbro" 10045 is impoverished, relative to the soil, in rare earths and Hf. Ratios by mass of Zr to Hf are comparatively low. Abundances of Mn, Co, Fe, Sc and Cr suggest systematic differences between igneous rocks on one hand and breccias and "soil" on the other. From the Co abundances, no more than about 3 percent of the present "soil" can consist of chondritic meteorite contamination.*

We have analyzed ten Apollo 11 specimens by techniques similar to those of Gordon *et al.* (1), with such modifications as recent experience dictates. Twenty-six elements were determined, but owing to space limitations we here report only salient observations and conclusions. A table of abundance and error estimates will be provided on request, and data and interpretations will be presented more fully in a subsequent paper (2).

Specimens were split where feasible to provide replicates. Two, 10044 and

10084, were treated more elaborately. The "microgabbro" powder, 10044, was split to yield a representative sample and then sieved with 100-mesh nylon screening. Fine material was split again, and the coarse material was hand-cobbed for separates of plagioclase, pyroxenes, and opaque minerals. The residue from the hand-cobbing, which remains representative of the coarse material to a good approximation, was split to give duplicate samples. The soil specimen, 10084, was sieved with 100-mesh and 325-mesh screening; splits of the origi-