

## Evidence for meteoritic sulfur in the lunar regolith\*

J. F. KERRIDGE, I. R. KAPLAN, and C. PETROWSKI

Institute of Geophysics and Planetary Physics, University of California, Los Angeles,  
California 90024

**Abstract**—Soils contain sulfur of both indigenous and meteoritic origin. A proportion of this total sulfur is lost by an isotopically competitive mechanism during regolith maturation leading to an exposure-related enrichment in  $^{34}\text{S}$ . The loss mechanism may be impact vaporization, solar-wind sputtering, proton stripping, or a combination of these. The meteoritic component, which probably resembles indigenous sulfur isotopically, correlates in magnitude with inferred surface exposure times and quantitatively agrees with estimates of the recent meteorite influence based upon volatile trace-element abundances.

### INTRODUCTION

ALTHOUGH DEPLETED in the moon relative to its cosmic abundance, sulfur is present in virtually all lunar rocks, mainly as troilite, FeS. Mare basalts are substantially richer in S than are highland rocks, values up to 3000 ppm being found in the former and down to 20 ppm in the latter. Formation of regolith by comminution of rocks results in transfer of indigenous S to the soil and much of the interest in S has focused on explaining the distribution of S and  $\delta^{34}\text{S}$  within the regolith. The pertinent observations of lunar S systematics to date may be summarized as follows.

Soils are neither depleted nor enriched significantly in S with respect to their associated rocks (Moore *et al.*, 1972; Gibson and Moore, 1974; Kerridge *et al.*, 1975). An apparent depletion in soils at Apollo 11 (Kaplan *et al.*, 1970) may be due to incomplete sampling. Isotopic analysis of S in soils shows that they are enriched in  $^{34}\text{S}$  relative to rocks by amounts ranging up to about 1% (Kaplan *et al.*, 1970; Petrowski *et al.*, 1974; Rees and Thode, 1974). These enrichments correlate with surface exposure age (Rees and Thode, 1974; Petrowski *et al.*, 1974), and also show a tendency, at least at the Apollo 16 site, to increase with S content (Kerridge *et al.*, 1975). Too few soil breccias have been studied to permit generalizations, but they appear to resemble soils in abundance and isotopic composition (Kaplan *et al.*, 1970; Petrowski, 1974).

Abundance patterns of S in soils have been interpreted in terms of simple mixing models involving contributions from various identifiable major rock types (e.g. Moore *et al.*, 1972; Gibson and Moore, 1974). These authors concluded that there was no evidence for an extralunar S component, though Moore *et al.* (1974) have subsequently suggested that some surface-correlated S may be of meteoritic

---

\*Presented at the Conference on Origin and Evolution of the Lunar Regolith, Houston. November 1974.

origin. Simple mixing models of this type cannot explain the isotopic fractionation observed between rocks and soils.

Clayton *et al.* (1974) proposed that enrichment of soils in  $^{34}\text{S}$  is a result of Rayleigh fractionation of material either vaporized during meteorite impact or sputtered from vapor deposited layers. They suggested that  $\sim 30\%$  S loss from the regolith could result in isotopic enrichments of the magnitude observed, but they did not demonstrate that such losses are actually compatible with observed abundance data. The tendency for  $\delta^{34}\text{S}$  values of soil samples at the Apollo 16 site to increase with both S abundance and surface exposure age, taken in conjunction with the abundance patterns of regionally associated rocks and soils, led us to propose a complex S cycle on the lunar surface involving vaporization and recondensation (Kerridge *et al.*, 1975). We suggested that recondensation of S might be partly controlled by the availability of metallic iron surfaces. Subsequently, we noted that simple Rayleigh fractionation, such as proposed by Clayton *et al.* (1974), would lead to a negative correlation between  $\delta^{34}\text{S}$  and S content, rather than the positive correlation observed (Petrowski *et al.*, 1974).

In order to quantify some of the trends described above and to evaluate the foregoing interpretations, we present here abundance and isotopic data for a suite of 19 soils from the Apollo 16 mission. Although variations in major-element distributions at this site are not great, sufficient variety exists to enable mixing processes to be discerned, and the wide spectrum of surface exposure ages observed aids in recognition of time-dependent effects in soils (Kerridge *et al.*, 1974, 1975).

## EXPERIMENTAL

Two alternative methods are commonly employed for the extraction of S from lunar samples. These are acid hydrolysis and combustion in  $\text{O}_2$ . During earlier work, we used the former technique with results which generally compared well with those of other investigators. For example, abundance values for soils from our first Apollo 16 allocation showed a 4% systematic discrepancy below existing literature data (Kerridge *et al.*, 1975). Results from the Apollo 17 samples, however, revealed a much larger systematic discrepancy in which data obtained by hydrolysis procedures (Petrowski *et al.*, 1974; Rees and Thode, 1974) averaged 30% below those obtained by combustion (Gibson and Moore, 1974; Moore *et al.*, 1974). Although the cause of this discrepancy is still not established, the possibility was considered that a part of the lunar S may be in a form not accessible to acid hydrolysis, the efficiency of which is critically dependent upon the nature of the S-bearing species. If a non-hydrolysable species is present in lunar samples, any isotopic fractionation of S between this species and the hydrolysable troilite would perturb the measured isotopic composition away from the true value. For this reason we believe that caution should be exercised in the interpretation of results obtained by hydrolysis, until the nature of the discrepancy is resolved, and that hydrolysis data should not be combined with those obtained by other means. Consequently, the discussion in this paper is based solely upon results obtained during the present study by combustion in  $\text{O}_2$  and does not include data from our earlier hydrolysis of Apollo 16 soils (Kerridge *et al.*, 1975). We emphasize that this is an interim decision and that work is continuing in order to solve this problem. In fact, the conclusions reached in this paper may also be derived, with only slightly different quantitative results but with poorer statistical accuracy, using our previously published hydrolysis data.

The sample is outgassed at  $150^\circ\text{C}$  under  $10^{-3}$  mmHg overnight. Oxygen, generated by heating  $\text{CuO}$ , at a partial pressure of about 10 cmHg is circulated over the sample which is heated to  $1220^\circ\text{C}$ . Evolved

gases are separated into condensible and non-condensable fractions in liquid N<sub>2</sub>, the condensable fraction being further separated by distillation from pentane ice at -120°C into CO<sub>2</sub> and SO<sub>2</sub> fractions. SO<sub>2</sub> is measured manometrically, collected and analyzed mass spectrometrically against the Canyon Diablo Troilite standard. Results are given in Table 1. Uncertainty limits in the table and in Figs. 1 and 2 are based on replicate analyses. System sensitivity and precision equal 3 μgS, the line blank being always less than this value. Average reproducibility of abundance values is ±4% based on replicate analyses. Accuracy of abundance measurements and reliability of the isotopic determinations are considered in greater detail below.

Since we reported our earlier results for Apollo 16 soils a considerable quantity of data has been published about these samples. Comparison of our combustion data with literature values reveals the following trends. Our abundance values average about 8% below the mean of all other investigators, employing a variety of techniques. However, significant structure exists within the comparative data. Thus, our values agree very well with those of Gibson and Moore (1973) (combustion-infrared absorption), but are systematically less than those of Moore and Cripe (1974) and Cripe and Moore (1975) (combustion-titration) and greater than those of Rees and Thode (1974) and Chang *et al.* (1975) (both using acid hydrolysis). Clearly, the discrepancy with the mean is due to the systematic difference between our data and those of the Arizona group and to the statistical weight carried by the preponderance of their analyses. This is not to imply that their data are necessarily in error; however, on the basis of present evidence we see no reason to regard our abundance values as unreliable, and in this paper they are treated as representing the true S contents of the samples.

As for isotopic results, the comparative data base is much more restricted, only two other groups, both employing acid hydrolysis procedures, having produced any data. Our δ<sup>34</sup>S values lie systematically 2–3‰ above those of Rees and Thode (1974) and Chang *et al.* (1975). We are currently working to resolve this discrepancy, but exchange of standards and samples between laboratories has so far failed to identify its cause. Although the possible failure of acid hydrolysis to extract all the S from a lunar sample *may* be responsible for the discrepancy, this point has not been demonstrated and we do not feel justified in treating our isotopic data as inherently more reliable than other investigators'.

Table 1. Sulfur abundances and isotopic compositions for Apollo 16 soils using combustion procedure.

Sample	S <sub>ppm</sub>	±	δ <sup>34</sup> S <sub>CDT</sub>	‰ ±
60051,31	520	6	+ 11.0	0.2
61141,33	330	17	+ 9.2	1.1
61161,27	570	10	+ 12.0	0.2
61220,29	470		+ 6.1	
61281,26	480	30	+ 9.9	1.5
62240,6	534	8	+ 10.8	0.5
64421,60	650	14	+ 13.5	0.3
64501,63	600	19	+ 11.7	0.1
64801,52	660	68	+ 13.3	0.3
64811,2	600	8	+ 11.9	0.5
65701,42	790	14	+ 13.2	0.1
65901,66	770	25	+ 13.0	0.1
66041,66	810	18	+ 12.3	
67460,3	320	23	+ 7.3	0.5
67480,23	330	5	+ 8.3	0.2
67601,5	360		+ 10.0	
67941,10	420	26	+ 6.6	0.0
68821,45	530	70	+ 11.1	0.2
68841,7	750	6	+ 11.1	0.0

Possible sources of error in our isotope measurements fall into three categories. These are, incomplete extraction of S from the sample, interference by some other species at mass 66, and variation in the  $^{18}\text{O}$  content of the  $\text{SO}_2$ . In view of the fact that our yields are systematically *greater* than those of the groups reporting *lower*  $\delta^{34}\text{S}$  values, and in the absence of compelling evidence that our abundance data are erroneously low, we believe that the first possibility is unlikely.

Although a number of species capable of interfering at mass 66 may be hypothesized (e.g.  $^{28}\text{Si}^{19}\text{F}_2^+$ ) none of these would pass through our separation procedure or are abundant enough to account for the observed discrepancy. Similarly, although lunar soils are known to be enriched in  $^{18}\text{O}$  (e.g. Taylor and Epstein, 1973), the excess of  $\text{O}_2$  supplied by the combustion line should swamp any contribution due to lunar oxygen. However, we are attempting to check these possibilities by investigating the effect of further purification of the  $\text{SO}_2$ . In one such experiment, an  $\text{SO}_2$  sample was reacted with an acid solution of stannous chloride, precipitating stannic sulfide. This was then combusted in  $\text{O}_2$  of standard  $^{18}\text{O}$  content to produce  $\text{SO}_2$  which was collected, measured and analyzed in the usual way. Unfortunately, the process was not quantitative, with a yield of 56%, so that the observed reduction in  $\delta^{34}\text{S}$  of 1.2‰ was inconclusive, being due possibly to fractionation in the stannous chloride treatment. Some confirmation for this alternative came from a repeat of the stannous chloride process on the same sample. A yield of 72% for the second reaction was accompanied by a further reduction in  $\delta^{34}\text{S}$  of 0.6‰, so that the apparent fractionation was proportional to the S loss during the reaction. Further experiments along these lines are clearly required.

For present purposes of interpretation a self-consistent set of data is required. Consequently we choose to employ our abundance and isotopic data which have been generated by a single technique, namely sample combustion. If these data are systematically in error by amounts comparable to the observed interlaboratory discrepancies, the effect will be to yield erroneous fractionation factors for any modelled lunar processes. However, as will become apparent below, for the model we present a small linear shift in  $\delta^{34}\text{S}$  which should not influence our interpretation.

## RESULTS

Figure 1 illustrates the relationship between S content and  $\delta^{34}\text{S}$  value and reveals the positive, albeit weak, correlation, referred to earlier. Such a trend might be expected to result from a mixture of lunar S with some extralunar component enriched in  $^{34}\text{S}$ . However, the lack of any obvious evidence for an excess of S in soils with respect to rocks (Moore *et al.*, 1972) and the fact that meteoritic S is characterized by  $\delta^{34}\text{S}$  values in the range 0–0.5‰ (Kaplan and Hulston, 1966) argue against this interpretation. The solar abundance of S is too low for implanted solar-wind atoms to be a significant contribution (Moore *et al.*, 1971). It follows that the trend in the isotopic data is more likely to reflect a fractionation process than a mixture, and this is borne out by Rees and Thode's (1972) observation that enrichments of  $^{33}\text{S}$  and  $^{36}\text{S}$  are respectively half and twice that of  $^{34}\text{S}$ . These data also indicate that nuclear processes have not affected the isotopic composition of regolith S (Clayton *et al.*, 1974). Enrichment of soils in  $^{34}\text{S}$  correlates strongly with nitrogen content (see Fig. 2). Solar-wind-implanted N is very efficiently retained (Müller, 1974; DesMarais *et al.*, 1974; Kerridge, 1975) and thus may be used as a monitor for soil maturity. We conclude that fractionation of S isotopes is an ongoing process on the lunar surface.

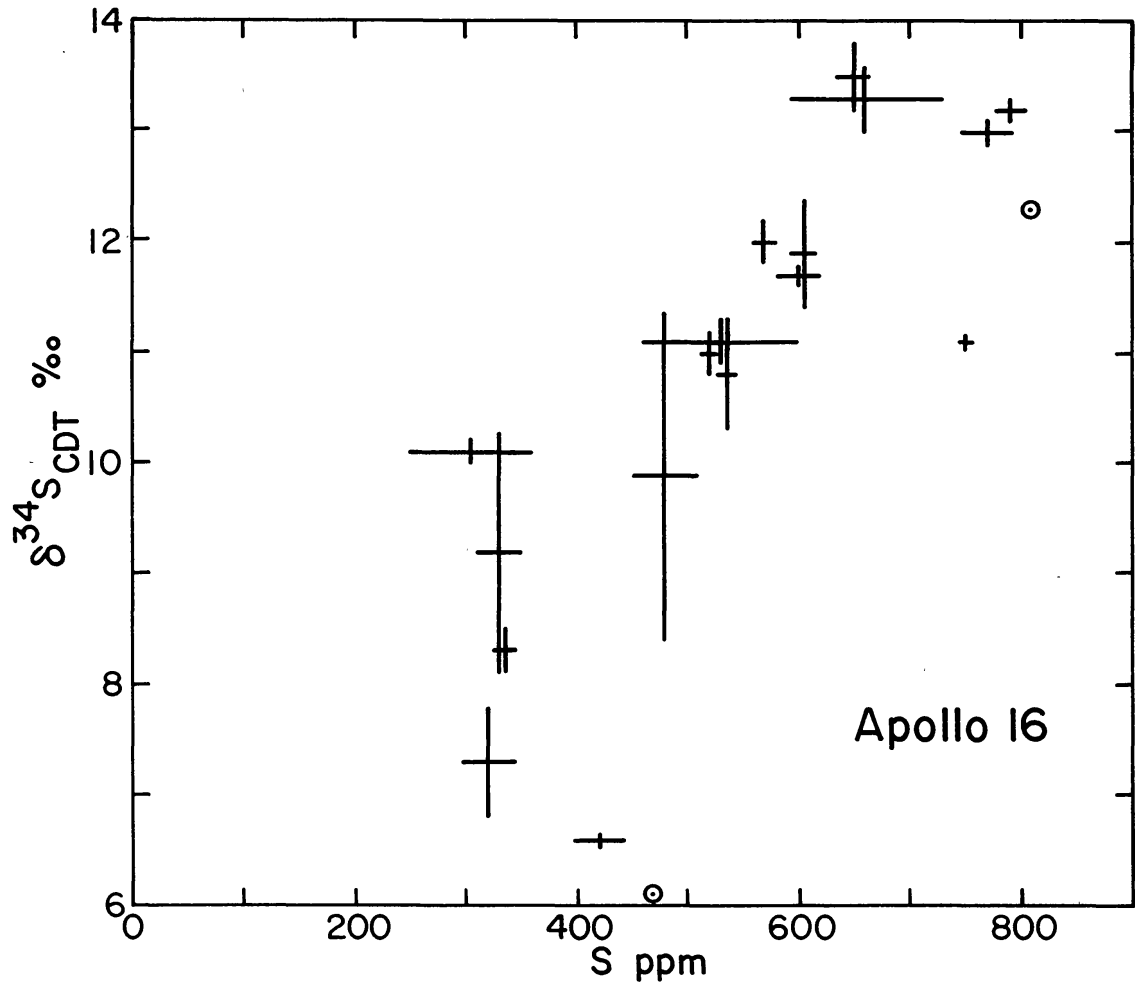


Fig. 1. Sulfur abundances and  $\delta^{34}\text{S}$  values for Apollo 16 fines show a weak positive correlation.

#### DISCUSSION

Clayton *et al.* (1974) proposed that the observed enrichments in  $^{34}\text{S}$  for soils implied loss from the moon of substantial amounts of  $^{32}\text{S}$ -enriched S. Assuming that the mass dependence of the loss mechanism was governed by the inverse square root of the isotope mass, they calculated that a mature soil could have lost about 30% of its original complement of S. It is important to note that there is no way to obtain the fractionations observed without losing a large fraction of material. This suggests that the apparent agreement between soil and rock abundances is fortuitous and may not support a simple mixing hypothesis. In what follows, we explore the possibility that a hypothetical meteoritic component,  $S_M$ , may explain the discrepancy between the amount which might reasonably have been supplied by indigenous rocks,  $S_R$ , and the quantity of S implied by Clayton's calculations. These calculations indicate that an amount of S, which we put equal to  $S_L$ , has been lost from soils. We define a quantity,  $S_o$ , equal to the sum of  $S_L$

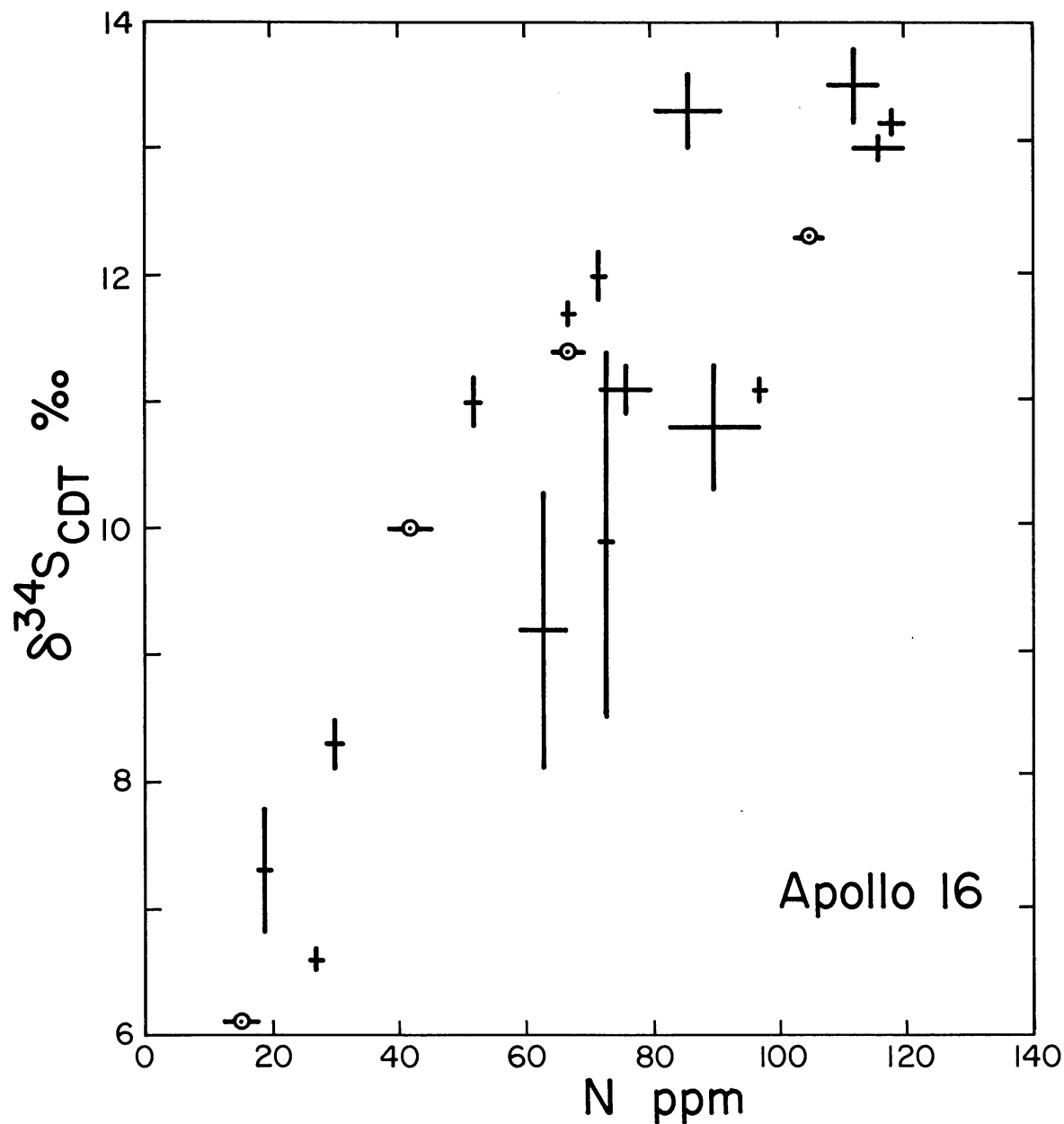


Fig. 2.  $\delta^{34}\text{S}$  values for Apollo 16 soils increase with increasing maturity as measured by agglutinate contents or, as here, by total N contents.

and the presently observed S content of the soil,  $S_P$ . Therefore:

$$S_O = S_P + S_L \quad (1)$$

and

$$S_M = S_O - S_R. \quad (2)$$

Following Clayton *et al.* (1974) we assume that the fractionation which accompanies reduction of the S content from  $S_O$  to the presently observed amount,  $S_P$ ,



may be approximated mathematically by the Rayleigh equation:

$$\ln ({}^{34}\text{S}/{}^{32}\text{S})_O - \ln ({}^{34}\text{S}/{}^{32}\text{S})_P = \left(1 - \frac{1}{\alpha}\right) \ln (S_P/S_O), \quad (3)$$

where  $\alpha$  is the fractionation factor. Although we cannot at this stage specify the actual loss process with any certainty, the different mechanisms proposed, impact vaporization, sputtering and proton stripping, rely upon diffusive separation so that in the optimum case, each would be characterized by a Rayleigh fractionation equation. Note that, regardless of isotopic considerations, impact vaporization is a necessary part of the lunar S cycle in order to account for the high degree of surface correlation found by Rees and Thode (1974). Both of the other processes affect only the surface of grains and require vaporization and recondensation as a preliminary stage in order to make indigenous S accessible to their attack.

The fractionation factor,  $\alpha$ , is calculated assuming that the volatile species is  $\text{S}^0$ , probably arising from dissociation of  $\text{FeS}$ , so that  $\alpha$  is equal to  $\sqrt{34}/\sqrt{32}$ , i.e. 1.03. For the case of proton stripping, the appropriate species would be  $\text{H}_2\text{S}$ , involving only a trivial difference in  $\alpha$ . Thus Eq. (2) may be used to calculate  $S_O$  from the presently observed S content,  $S_P$ , and its isotopic composition provided the initial  ${}^{34}\text{S}/{}^{32}\text{S}$  ratio is known. Unfortunately, the isotopic composition of indigenous lunar S cannot be exactly defined at present. Of 20 rocks, from all sites, studied in our laboratory, 17 gave  $\delta^{34}\text{S}$  values which lay in the range 0.8–2.8‰ relative to Canyon Diablo Troilite. The three exceptions gave negative values (–5.0–0‰) which are not understood at this time. In order to make a model tractable, we shall assume that rock S is isotopically homogeneous, with  $\delta^{34}\text{S} = 1.8\text{‰}_{\text{CDT}}$ , but these exceptions should be borne in mind. Anders *et al.* (1973) have identified the major recent meteoritic component on the moon as having the composition of CI carbonaceous chondrites. Since the S in the meteorites is close to indigenous lunar S in its  ${}^{34}\text{S}$  content (Kaplan and Hulston, 1966), we assume that  $\delta^{34}\text{S}_O$  equals  $1.8\text{‰}_{\text{CDT}}$ . Consequently, in the case of a sample for which the S content and  $\delta^{34}\text{S}$  value are known,  $S_O$  may be calculated.

From values of  $S_O$ , it follows that knowledge of the indigenous component,  $S_R$ , would enable  $S_M$  to be calculated for each soil sample. The agreement, or otherwise, between these values and other estimates of meteoritic influence in the samples may then serve as a test of the role of meteoritic S in lunar regolith chemistry.

Even within a single sampling station, individual rocks show too much variation in their S contents to permit calculation of a reasonable value of  $S_R$  characteristic of that area. However, a plot of S content against total Fe content for Apollo 16 rocks, Fig. 3, reveals a strong correlation, suggesting that the Fe content of a soil may be used to calculate the magnitude of  $S_R$ . Values for total Fe content are available in the literature for most of our soil samples and these have been used to calculate values of  $S_R$  via Fig. 3. It may be argued that this method of assessing the role of indigenous lunar S fails to take into account the possible effect of lateral transport from distant regions of the moon. Such transport of mare material into the Apollo 16 site may be inferred from the observations by

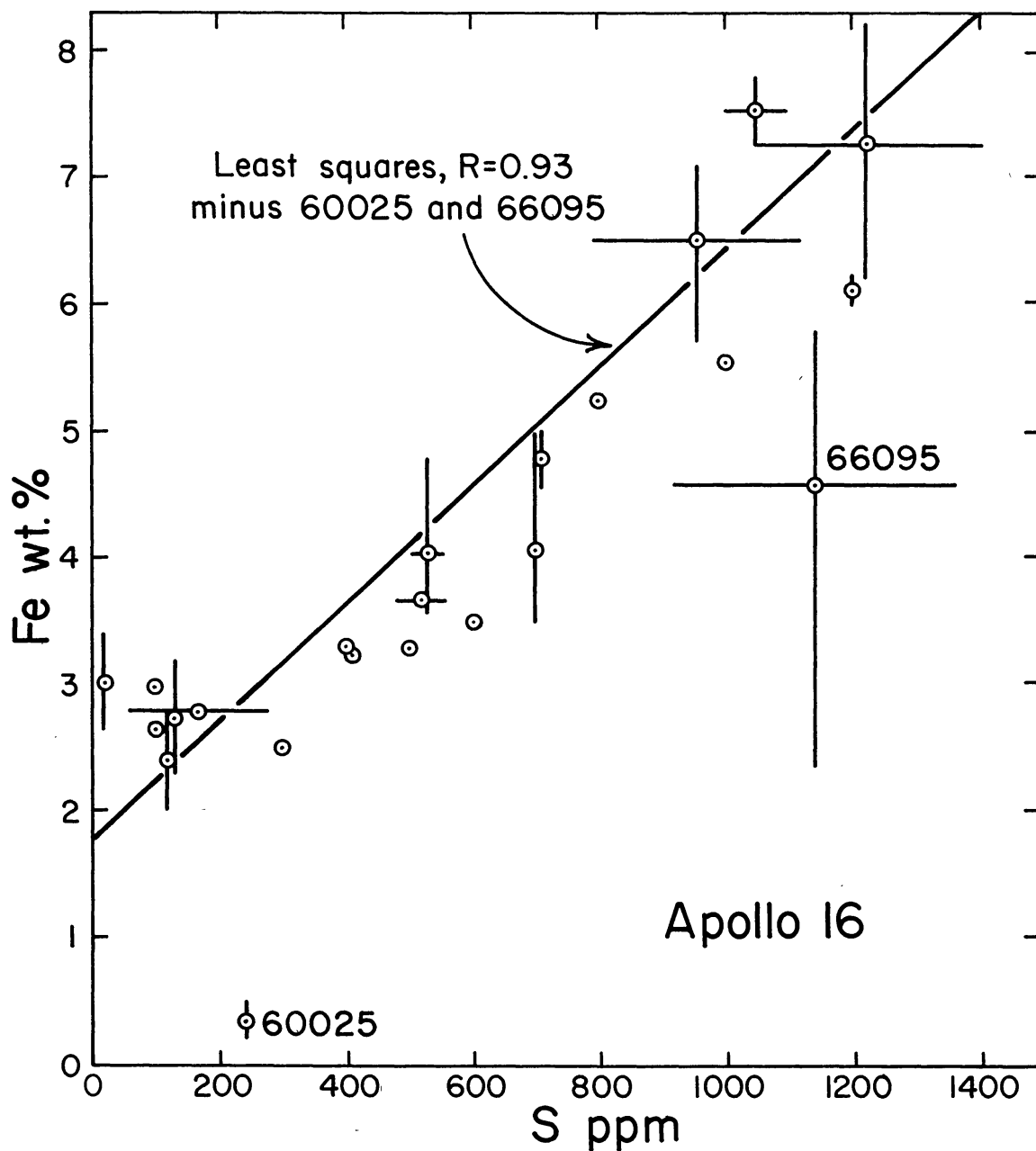


Fig. 3. Total Fe and total S contents for Apollo 16 rocks correlate strongly.  $S_R$  contents of soil samples were calculated from their Fe contents using the solid line which was computed without including points 60025 and 66095. Data from *Apollo 16 Lunar Sample Information Catalog and Proc. Fourth and Fifth Lunar Sci. Conferences.*

Rhodes *et al.* (1975) of time-dependent enrichments of soils in elements such as Fe, Mn, and Ti. However, the S/Fe ratio at all other regions of the moon sampled to date is *lower* than that characteristic of rocks at the Apollo 16 site, so that our method of calculating the magnitude of the indigenous component will actually overestimate slightly the effect of S transported in particulate form from distant regions. Calculation of the amount of lunar S transported via the vapor phase and reimplanted from the lunar atmosphere is more difficult and requires data which



are not yet available. Qualitatively, however, it is easy to show that this effect is small by means of the following argument. Since  $\delta^{34}\text{S}$  of mare soils also increases with maturity (Petrowski *et al.*, 1974), it follows that S vaporized from these regions must be isotopically light. Implantation of such S in highland soils will increase their S content but will tend to reduce their  $\delta^{34}\text{S}$  values. The fact that these delta values actually increase with time of exposure therefore requires even greater fractionation, i.e. loss, of S from the highlands, which is not supported by the evidence. We shall return to this point later, meanwhile we feel justified in taking our calculated values of  $S_R$  as realistic measures of the indigenous component. Subtraction of these values from those calculated for  $S_O$  leads to estimates of  $S_M$  for each sample and these are plotted in Fig. 4 against N content as a measure of soil maturity. This reveals a strong correlation, which is clear evidence for a time-dependent component, as expected for meteoritic material.

Figure 4 does not indicate whether or not the magnitude of  $S_M$  is compatible with other estimates of the meteoritic influence. Therefore in Fig. 5, values of  $S_M$ , as calculated above, are plotted against values calculated from estimates of the

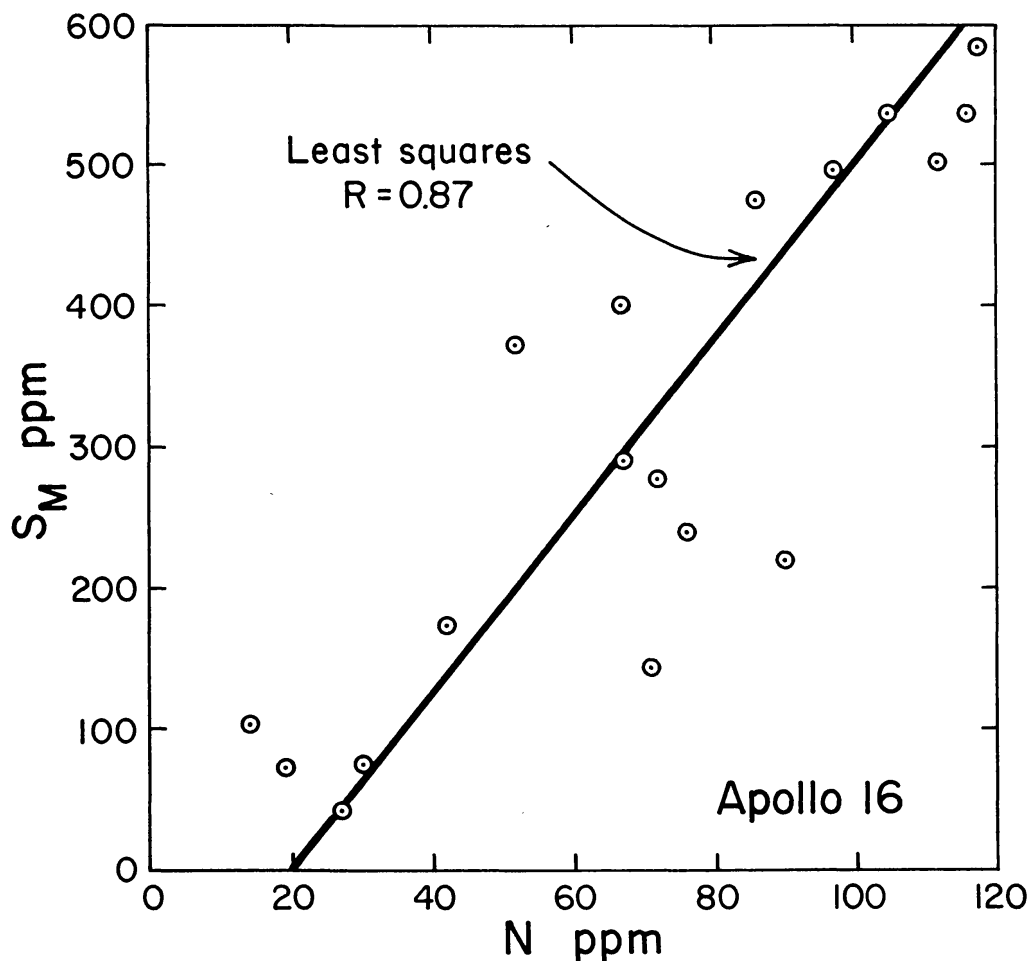


Fig. 4. Magnitude of the calculated meteoritic S component correlates with total N content as a measure of soil maturity.

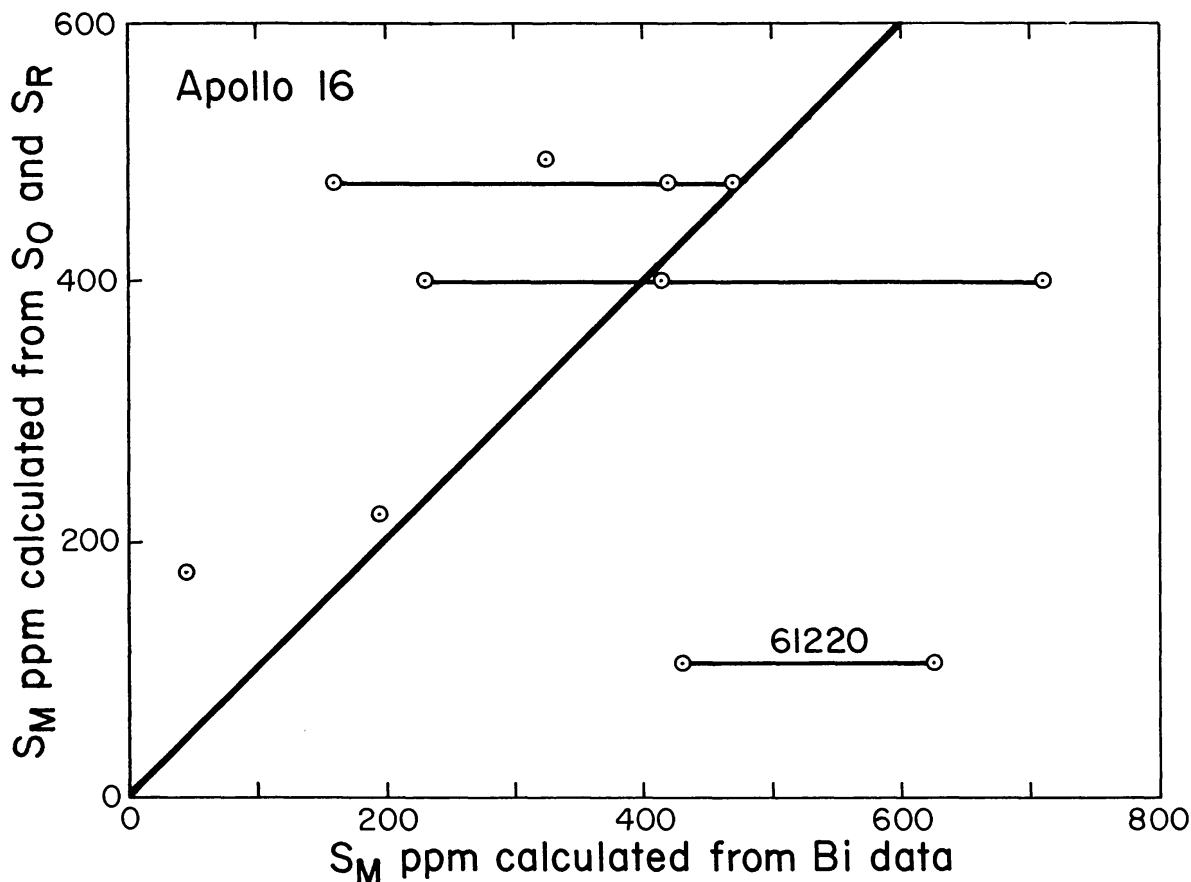


Fig. 5. Magnitude of the calculated meteoritic S component is compatible with estimates of the recent micrometeorite influence using the procedure of Krähenbühl *et al.* (1973) and measured Bi contents. Bi data from Allen *et al.* (1973), Hughes *et al.* (1973), and Krähenbühl *et al.* (1973). Solid line has 45° slope and is not a least-squares fit.

meteoritic component based on Bi contents, using the procedure of Krähenbühl *et al.* (1973), and assuming an S content of 5.5% in the meteoritic material (Mason, 1963; Kaplan and Hulston, 1966). Although the data are too scanty and their scatter is too great to permit a statistically useful correlation, they are clearly compatible with a meteoritic origin for the  $S_M$  component.

We conclude that the S systematics of the lunar regolith support a model in which S from a number of indigenous rock types is mixed with a time-dependent, extralunar component of probably meteoritic origin. This mixture is processed on the lunar surface by a time-dependent mechanism which removes  $^{32}\text{S}$ -enriched S from the moon, enriching the residue in  $^{34}\text{S}$ . The loss of S by this mechanism *approximately* equals the meteoritic input, producing the *apparent* coincidence between rock abundances and those in soils which has led to adoption of simplified mixing models. In reality, at least for the Apollo 16 site, addition of meteoritic S slightly outweighs loss of S from the moon, resulting in the positive trend in Fig. 1.

Unfortunately, although the fractionation process may be adequately modelled by means of the Rayleigh equation with a fractionation factor equal to the inverse

square root of the mass ratio, this does not uniquely specify the physical process involved. Clayton *et al.* (1974) have argued that parallel enrichments in the heavy isotopes for both S and K do not support a low-temperature stripping mechanism but rather require volatilization, presumably during meteorite impact, probably followed by partial recondensation and ion sputtering of the vapor deposited layer. We are in basic agreement with this view though, in light of the general tendency of fractionation factors to decrease with increasing temperature, we believe that sputtering may be the more important fractionation process.

Finally, although our model provides an acceptable interpretation of the S systematics at the Apollo 16 site, it remains to be shown whether or not it will be applicable at other sites. In particular, it is important to determine the actual extent of lateral transport of S between distant regions. Work is in progress on this issue and also on the question of the isotopic homogeneity of lunar S, referred to earlier.

*Acknowledgments*—We thank D. Winter for assistance, S. Chang and K. Marti for helpful discussions, S. Chang for permission to use unpublished results and LSAPT and the JSC Curatorial Staff for supplying so many useful samples. Work supported by NASA grant NGR-05-007-289.

*Note*—By measuring both the 66/64 mass ratio of the  $\text{SO}_2^+$  fragment and the 50/48 mass ratio of the  $\text{SO}^+$  fragment in the mass spectrometer it is possible to correct for variations in the  $^{18}\text{O}/^{16}\text{O}$  ratio (Holt and Engelkemeir, 1970). We have performed these measurements for two  $\text{SO}_2$  samples, produced by combustion of a basalt and a soil, and have obtained results which agree closely with those obtained solely from the 66/64 mass ratio. This confirms that neither interference nor variations in  $^{18}\text{O}/^{16}\text{O}$  ratio are perturbing isotope values obtained by combustion.

Holt B. D. and Engelkemeir A. G. (1970) Thermal decomposition of barium sulfate to sulfur dioxide for mass spectrometric analysis. *Anal. Chem.* 42, p. 1451–1453.

## REFERENCES

- Allen R. O., Jovanovic S., Showalter D., and Reed G. W. (1974)  $^{204}\text{Pb}$ , Bi and Zn in Apollo 16 samples and inferences on the lunar geochemistry of  $^{204}\text{Pb}$  based on meteoritic and terrestrial sample studies (abstract). In *Lunar Science V*, p. 12–14. The Lunar Science Institute, Houston.
- Anders E., Ganapathy R., Krähenbühl U., and Morgan J. W. (1973) Meteoritic material on the moon. *The Moon* 8, 3–24.
- Chang S., Lennon K., and Walberg G. (1975) Sulfur in magnetic and non-magnetic particles of Apollo 16 soils. In press.
- Clayton R. N., Mayeda T. K., and Hurd J. M. (1974) Loss of oxygen, silicon, sulfur and potassium from the lunar regolith. *Proc. Lunar Sci. Conf. 5th*, p. 1801–1808.
- Cripe J. D. and Moore C. B. (1975) Total sulfur contents of Apollo 15, 16 and 17 samples (abstract). In *Lunar Science VI*, p. 167–168. The Lunar Science Institute, Houston.
- DesMarais D. J., Hayes J. M., and Meinschein W. G. (1974) Retention of solar wind-implemented elements in lunar soils (abstract). In *Lunar Science V*, p. 168–170. The Lunar Science Institute, Houston.
- Gibson E. K. and Moore G. W. (1973) Carbon and sulfur distributions and abundances in lunar fines. *Proc. Lunar Sci. Conf. 4th*, p. 1577–1586.
- Gibson E. K. and Moore G. W. (1974) Sulfur abundances and distributions in the valley of Taurus-Littrow. *Proc. Lunar Sci. Conf. 5th*, p. 1823–1837.
- Hughes T. C., Keays R. R., and Lovering J. F. (1973) Siderophile and volatile trace elements in Apollo 14, 15 and 16 rocks and fines: Evidence for extralunar component and Ti-, Au- and Ag-enriched

- rocks in the ancient lunar crust (abstract). In *Lunar Science IV*, p. 400–402. The Lunar Science Institute, Houston.
- Kaplan I. R. and Hulston J. R. (1966) The isotopic abundance and content of sulfur in meteorites. *Geochim. Cosmochim. Acta* **30**, 479–496.
- Kaplan I. R., Smith J. W., and Ruth E. (1970) Carbon and sulfur concentration and isotopic composition in Apollo 11 lunar samples. *Proc. Apollo 11 Lunar Sci. Conf.*, p. 1317–1329.
- Kerridge J. F. (1975) Solar nitrogen: Evidence for a secular increase in the ratio of nitrogen-15 to nitrogen-14. *Science* **188**, 162–164.
- Kerridge J. F., Kaplan I. R., and Lesley F. D. (1974) Accumulation and isotopic evolution of carbon on the lunar surface. *Proc. Lunar Sci. Conf. 5th*, p. 1855–1868.
- Kerridge J. F., Kaplan I. R., Petrowski C., and Chang S. (1975) Light element geochemistry of the Apollo 16 site. *Geochim. Cosmochim. Acta* **39**, 137–162.
- Krähenbühl U., Ganapathy R., Morgan J. W., and Anders E. (1973) Volatile elements in Apollo 16 samples: Implications for highland volcanism and accretion history of the moon. *Proc. Lunar Sci. Conf. 4th*, p. 1325–1348.
- Mason B. (1963) The carbonaceous chondrites. *Space Sci. Rev.* **1**, 621–646.
- Moore C. B. and Cripe J. D. (1974) Total sulfur contents of Apollo 15 and Apollo 16 lunar samples (abstract). In *Lunar Science V*, p. 523–525. The Lunar Science Institute, Houston.
- Moore C. B., Lewis C. F., Larimer J. W., Delles F. M., Gooley R., and Nichiporuk W. (1971) Total carbon and nitrogen abundances in Apollo 12 lunar samples. *Proc. Lunar Sci. Conf. 2nd*, p. 1343–1350.
- Moore C. B., Lewis C. F., Cripe J. D., Delles F. M., Kelly W. R., and Gibson E. K. (1972) Total carbon, nitrogen and sulfur in Apollo 14 lunar samples. *Proc. Lunar Sci. Conf. 3rd*, p. 2051–2058.
- Moore C. B., Lewis C. F., and Cripe J. D. (1974) Total carbon and sulfur contents of Apollo 17 lunar samples. *Proc. Lunar Sci. Conf. 5th*, p. 1897–1906.
- Müller O. (1974) Solar wind nitrogen and indigenous nitrogen in Apollo 17 lunar samples. *Proc. Lunar Sci. Conf. 5th*, p. 1907–1918.
- Petrowski C. (1974) Light element geochemistry at the Fra Mauro region of the moon. M. S. thesis, UCLA.
- Petrowski C., Kerridge J. F., and Kaplan I. R. (1974) Light element geochemistry of the Apollo 17 site. *Proc. Lunar Sci. Conf. 5th*, p. 1963–1973.
- Rees C. E. and Thode H. G. (1972) Sulphur concentrations and isotope ratios in lunar samples. *Proc. Lunar Sci. Conf. 3rd*, p. 1479–1485.
- Rees C. E. and Thode H. G. (1974) Sulfur concentrations and isotope ratios in Apollo 16 and 17 samples. *Proc. Lunar Sci. Conf. 5th*, p. 1963–1973.
- Rhodes J. M., Adams J. B., Charette M. B., and Rogers K. V. (1975) The chemistry of agglutinate fractions in lunar soils (abstract). In *Lunar Science VI*, p. 665–667. The Lunar Science Institute, Houston.
- Taylor H. P. and Epstein S. (1973)  $O^{18}/O^{16}$  and  $Si^{30}/Si^{28}$  studies of some Apollo 15, 16 and 17 samples. *Proc. Lunar Sci. Conf. 4th*, p. 1657–1679.