Loss of oxygen, silicon, sulfur, and potassium from the lunar regolith

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Abstract—The processes of formation and maturation of lunar soils lead to enrichments in the heavy stable isotopes of oxygen, silicon, sulfur, and potassium. The isotopic enrichment implies substantial losses of these elements from the moon. Vaporization by micrometeorite impact and by ion sputtering have removed at least 1% of the mass of the regolith. The losses of sulfur and potassium amount to at least 20–30% of their original abundance in the regolith.

INTRODUCTION

VARIATIONS IN ISOTOPIC COMPOSITIONS in lunar soils have been reported for several light elements: H, C, N, O, Si, S, and K. The elemental abundances and isotopic compositions of the first three—hydrogen, carbon, and nitrogen—are dominated by the addition of atoms of extralunar origin, predominantly from the solar wind. Detailed interpretation of their isotopic variations is difficult due to lack of knowledge of the isotopic composition of the incoming material, uncertainties in the fluxes of solar wind, meteoritic and cometary matter, and inadequate understanding of the mechanisms for trapping the extralunar matter in the regolith. Of the other four elements, oxygen and silicon are major elements, sulfur and potassium are minor elements, and all exhibit the same range of concentrations in the soil as in crystalline rocks. Their abundances are sufficiently high that contributions to the soil from extralunar sources cannot exceed a few percent of the indigenous complement of these elements. Thus the observed variations in isotopic composition are attributable to processes of formation and maturation of the regolith, and interpretation of the variations should enhance our understanding of these processes.

RESULTS AND DISCUSSION

Processes leading to variation in isotopic abundance may be broadly classified as either nuclear or chemical, the latter encompassing all mass-dependent phenomena, including diffusion. For elements with three or more stable isotopes, an experimental distinction between nuclear and chemical processes can be made by comparing the relative magnitudes of isotope ratio variations for two or more

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different isotope ratios. Rees and Thode (1972) measured S^{33}/S^{32}, S^{34}/S^{32}, and S^{36}/S^{32} in lunar rocks and soils, and found the variations to be in the proportions $1:2:4$ as expected for chemical processes. Epstein and Taylor (1971) reported that variations in Si^{29}/Si^{28} were one-half as great as the corresponding variations in Si^{30}/Si^{28}. A similar test can be made, in principle, for potassium, but the low abundance of K^{40} makes this difficult (Barnes *et al.*, 1973). Partial fluorination reactions similar to those first described by Epstein and Taylor (1971) have been carried out on soils 14163 and 76501, and on soil breccia 70019. The resulting δO^{17} and δO^{18} values are shown in Fig. 1. For each sample, the first-reacting oxygen, representing $<0.6\%$ of the whole sample, is greatly enriched in the heavy isotopes, and successive fractions approach the whole-soil value ($\delta^{18} \approx 6\%_o$) monotonically. The points conform well to a chemical fractionation line of slope $\frac{1}{2}$.

Isotopic compositions of oxygen, silicon, sulfur, and potassium are very uniform in lunar igneous rocks (Epstein and Taylor, 1972; Clayton *et al.*, 1973; Rees and Thode, 1972; Barnes *et al.*, 1973). It can therefore be concluded that the isotopic variations observed in lunar soils result from chemical fractionation processes associated with formation of soil from rocks, and with maturation of the soil on exposure at the surface.

The isotopic variations of the four elements under discussion have several features in common, which suggest a common origin:

(a) All show enrichments in the heavy isotopes in soils relative to rocks.
(b) The magnitudes of the heavy isotope enrichments in oxygen, silicon, and sulfur are correlated with measures of soil maturity, such as noble gas content, metallic iron, particle tracks, etc. (Epstein and Taylor, 1971, 1972; Taylor and Epstein, 1973; Rees and Thode, 1974).
(c) There is some evidence for surface correlation for the effects in oxygen and silicon (Epstein and Taylor, 1971; Epstein and Taylor, 1972; Taylor and Epstein, 1973; Clayton *et al.*, 1971) and sulfur (Rees and Thode, 1974).
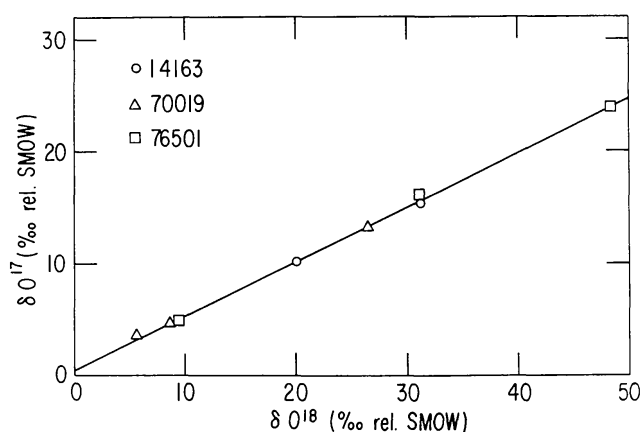

Fig. 1. Results of partial fluorination reactions of lunar soils (14163, 76501) and a soil breccia (70019). The data points fall along a line with slope $\frac{1}{2}$, showing that the isotopic variations are the result of mass-dependent processes.

Taken together, these observations imply that the heavy-isotope enrichment processes operate continuously or repeatedly within the regolith, and lead to a net enrichment of heavy isotopes relative to the source materials. No complementary heavy-isotope depleted materials are known among the lunar samples. Material balance thus requires that the heavy-isotope depleted component has been removed from the moon (Epstein and Taylor, 1971; Gibson et al., 1973).

Removal of matter from the moon may be visualized in two steps: (1) transport from the surface or interior of a solid grain to the lunar "atmosphere" and (2) loss from the atmosphere to interplanetary space. These may be parts of the same event, as in a hypervelocity micrometeorite impact (Gault et al., 1972), or may be two discrete processes, such as diffusive loss of volatile compounds from grains (Kaplan et al., 1970; Housley et al., 1974) followed by photoionization and removal by the solar wind (Manka and Michel, 1971). More complex models can be constructed, involving volatile transport, deposition on grain surfaces, and subsequent removal from the moon by sputtering due to solar ion bombardment (R. M. Housley, personal communication, 1974).

Although it is possible that different combinations of processes are predominant for different elements, the parallelism in behavior of the four elements under consideration suggests that a common mechanism may govern all four. This conclusion was also reached by Epstein and Taylor (1974). These elements are very disparate in their pertinent chemical properties, such as ionization potentials, volatility as elements, stability and volatility of hydrides, etc. In particular, potassium forms no compounds volatile at lunar surface temperatures, so that removal of a substantial fraction of potassium from the interiors of grains, as implied by the large isotope effects, requires volatilization at high temperatures, probably in excess of 1000°C (Gibson et al., 1973). Under these conditions, extensive decomposition of sulfides and loss of sulfur by vaporization also occurs (Gibson and Moore, 1973). Loss of oxygen and silicon by volatilization in micrometeorite impacts is also feasible, as shown by the vapor pressure measurements of DeMaria et al. (1971).

There is good evidence that brief exposure of liquid droplets of lunar magma to the interplanetary vacuum does not result in significant loss of these elements by vaporization. Neither the Apollo 15 green glass spheres nor the Apollo 17 orange glass show any measurable heavy-isotope enrichments in oxygen (Clayton et al., 1973; Taylor and Epstein, 1973), silicon (Taylor and Epstein, 1973), or potassium (Barnes et al., 1973).

We have also examined glassy agglutinates from lunar soils in order to determine whether the glasses are enriched in O^{18} relative to the remainder of the soil. The results are shown in Table 1. No measurable differences were found between the agglutinates and the whole soil. Thus, although the O^{18}-enrichment in the soil is correlated with the agglutinate content, the glass itself does not have an exceptional isotopic enrichment.

By means of a simple model, it is possible to set lower limits on the amounts of material which must be removed from the moon in order to account for the heavy-isotope enrichments found in the regolith. If isotopic fractionation takes

Table 1. Oxygen isotopic compositions of soils and glassy agglutinates.

Sample no.	δO^{18} soil (‰)	δO^{18} agglutinates (‰)
14163	6.14	6.01
15270	5.96	6.00
66081	6.07	6.12

place either by diffusive loss of the elements from a grain or liquid drop, or by diffusive separation in the lunar atmosphere, a fractionation factor equal to the inverse square root of the atomic masses is a good approximation. For a given fractionation factor, the maximum isotope effect in the residual portion will result if a Rayleigh process is assumed, i.e. the residue within the grain, liquid drop or atmosphere remains well mixed. Then the isotopic enrichment in the residue can be calculated as a function of the fraction of the element remaining in the residue. Results are shown in Fig. 2 for the four elements under consideration. The shaded regions along the curves for sulfur and potassium cover the ranges observed for isotopic compositions of whole soil samples: 4–10‰ for sulfur (Kaplan *et al.*, 1970; Kaplan and Petrowski, 1971; Rees and Thode, 1972; Smith *et al.*, 1973), and 5–8‰ for potassium (Barnes *et al.*, 1973). These isotopic compositions imply that, of the original potassium and sulfur which were present in the rock from which the regolith was formed, some 20–30% has been lost from the moon. The "whole-soil" heavy-isotope enrichments for oxygen are only about 0.5‰ (Onuma *et al.*, 1970; Epstein and Taylor, 1970; O'Neil and Adami, 1970) and for silicon are about 0.3‰ (Epstein and Taylor, 1970), implying losses of approximately 1% of these elements from the moon. Since oxygen and silicon are the two most abundant elements in lunar rocks, it can be concluded that the moon has lost at least 1% of the mass of the regolith through processes of micrometeorite bombardment and ion sputtering.

It has been shown by partial fluorination reactions (Epstein and Taylor, 1971) that the small O^{18} and Si^{30} enrichments in the lunar soil are the consequence of very large enrichments localized in a small fraction of the soil, which is especially reactive with fluorine. These large enrichments are indicated schematically in Fig. 2 by hash-marks along the fractionation curves for oxygen and silicon. In the extreme cases, these samples represent residues from which over half of the original oxygen and silicon has been removed. These can hardly be *in situ* residues of thermal volatilization, nor is it feasible to produce them by sputtering of solid particles at low temperatures, since no mechanism exists to maintain the Rayleigh condition of an homogenized residue. It is more likely that the extreme isotope enrichments are in vapor deposits derived from matter volatilized by impact and fractionated in the atmosphere. Large enrichments in S^{34} are found in the finest size-fractions of the soils (Rees and Thode, 1974) which could result from a similar process occuring for sulfur. Tests for surface correlation of the potassium isotope effects have not yet been reported.
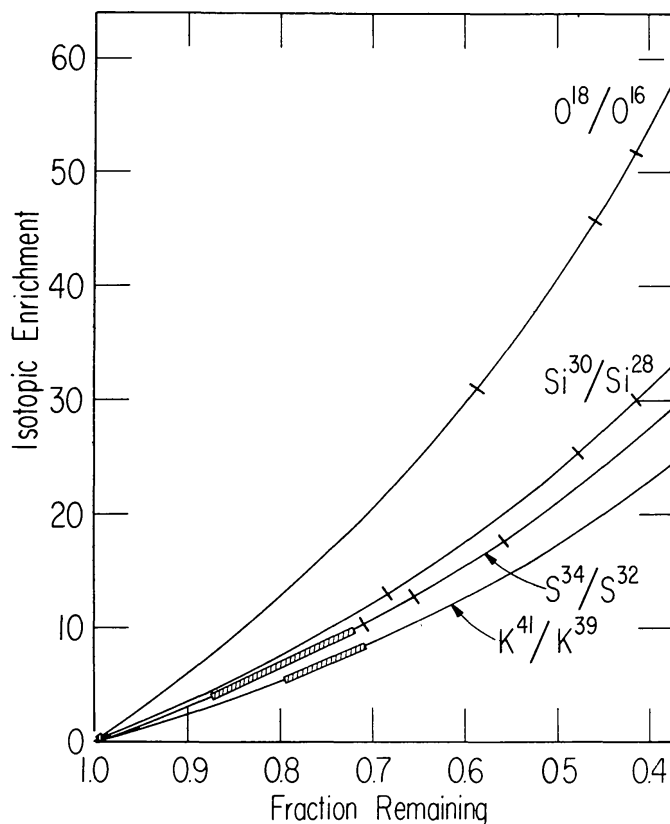
Fig. 2. Rayleigh fractionation curves showing heavy-isotope enrichment (in permil, ‰) as a function of the residual fraction of each element. The cross-hatched regions along the sulfur and potassium curves, and near the origin on the oxygen and silicon curves indicate the ranges of isotopic compositions observed for whole soils. Larger isotopic enrichments, found in surface or reactive sites, are indicated by hash-marks along the curves. Potassium data are from Barnes *et al.* (1973); sulfur data are from Kaplan *et al.* (1970), Kaplan and Petrowski (1971), Rees and Thode (1972, 1974), Smith *et al.* (1973), Kerridge *et al.* (1974); data on silicon and oxygen (whole-soil and stripping reactions) are from Epstein and Taylor (1971, 1972, 1974) and Taylor and Epstein (1973); additional whole-soil oxygen data are from Onuma *et al.* (1970), Clayton *et al.* (1971, 1972, 1973).

Could losses of sulfur and potassium of 20–30% have gone undetected by ordinary elemental analyses? The range of sulfur contents of lunar surface rocks is broader than that of lunar soils. At the Apollo 12, 14, 15, and 16 sites, the vast majority of rocks *and* soils analyzed contain 500–800 ppm sulfur (Kaplan and Petrowski, 1971; Moore *et al.*, 1972; Rees and Thode, 1972, 1974; Gibson and Moore, 1973; Smith *et al.*, 1973; Cripe and Moore, 1974). The basalts at the Apollo 11 and 17 sites are much richer in sulfur (1500–2800 ppm), whereas soils in these areas are only moderately richer (600–1300 ppm) (Kaplan *et al.*, 1970; Gibson and Moore, 1974; Chang *et al.*, 1974; Kerridge *et al.*, 1974; Rees and Thode, 1974; Moore *et al.*, 1974). Thus, although the coverage is too sparse to permit good averaging over the lunar surface, the sulfur analyses are entirely compatible with the hypotheses of considerable volatile transport and of loss of a large fraction from the moon. At any particular site on the moon, local variations of sulfur

content of soils is dominated by mixing in various proportions of sulfur-rich and sulfur-poor components (Cripe and Moore, 1974; Gibson and Moore, 1974).

Comparison between igneous rock analyses and soil analyses is even more difficult in the case of potassium, due to the ubiquitous admixture of a KREEP component in the soils, bringing in large amounts of potassium from a source not represented among the major igneous rock types sampled. Thus the substantial net loss of potassium from the regolith could not have been observed on the basis of elemental analyses alone. The increase of potassium content with decreasing grain size (and hence increasing specific surface area) in several soils (Evenson et al., 1973), has been cited as evidence for vapor transport and redeposition, but has been considered primarily in terms of addition rather than removal of potassium from the regolith.

It has long been recognized that potassium–argon dating of lunar soils is seriously complicated by the presence of large amounts of reimplanted Ar^{40} (Heymann and Yaniv, 1970). It is evident from the discussion above that the regolith system is open with respect to potassium, as well as argon, so that whole-soil "ages" do not measure single lunar events (Pepin et al., 1972, 1974).

We may also examine the implication of the observation of major losses of potassium to abundance of the related element, rubidium. Gibson et al. (1973) measured the loss of potassium and rubidium from lunar rocks and soils by heating in vacuum, and found rubidium to be lost preferentially. Their experiments were designed to simulate conditions in a hot ejecta blanket, and thus involved heating at temperatures near 1000°C for many hours. Under these conditions, the relative volatilities of potassium and rubidium are probably determined by their relative equilibrium vapor pressures. However, in the rapid heating in a lunar micrometeorite impact, the relative volatile loss is more likely to be governed by transport processes, favoring potassium over rubidium. Furthermore, after vaporization, removal from the moon is easier for potassium than for rubidium due to its lower mass. Hence, large losses of potassium do not necessarily imply large losses of rubidium. Major rubidium losses would be inconsistent with the common observations of Rb–Sr model ages near 4.6 b.y. However, Nyquist et al. (1973) have shown that rubidium losses from soils are not entirely negligible, leading to a correlation between increased model ages (Rb loss) and soil maturity, as measured by the content of glassy agglutinates.

CONCLUSIONS

Several recent papers have noted that stable isotope variations of some light elements imply significant degrees of volatilization and vapor phase transport (Epstein and Taylor, 1971; Barnes et al., 1973; Gibson et al., 1973; Housley et al., 1974). In this paper we have summarized the data concerning oxygen, silicon, sulfur, and potassium, and have emphasized the fact that material balance requires removal of substantial amounts of these elements from the moon. The data for oxygen and silicon imply a mass loss from the moon of at least 1% of the mass of the regolith. The data for potassium and sulfur imply removal of at least

20–30% of the original complement of these elements from the regolith and thence from the moon. These four elements were selected for discussion because of the wealth of isotopic evidence available. It is clear that similar conclusions about loss from the moon must also hold for many other major and minor elements for which similar isotopic evidence is not available.

Thermal volatilization by micrometeorite impact is the important first step leading to loss of volatiles. Some material may reach escape velocity in the impact process, the remainder is redeposited on solid surfaces where it is susceptible to removal by ion sputtering. It is not evident that chemical interactions with implanted solar wind hydrogen or processes specifically involving radiation-damaged surfaces play any significant role in removal of these elements from the regolith and from the moon.

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