

## LUNAR VOLATILES: IMPLICATIONS FOR LUNAR RESOURCE UTILIZATION

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We critically review from a resources perspective the available data on the abundances of the noble gases, hydrogen, carbon, nitrogen, sulfur, fluorine and chlorine in lunar samples. The analytical and mineralogical data relevant to the water content of lunar materials are also reviewed. The factors affecting the abundances of the solar-wind-implanted volatiles are discussed. The extensive analytical data on the Apollo and Luna samples are used to estimate average global inventories of these volatiles in the lunar regolith. The analytical data are also used to discuss some implications of different volatile extraction schemes such as heating and grain size sorting. The chemistry of lunar volcanic gases is discussed with an emphasis on using the chlorofluorocarbon gases as chemical probes of the water abundance in the lunar interior and on the transport of ore-forming metals. Finally, we highlight some of the major unanswered questions about using lunar volatiles as resources and make recommendations for future resource related work on lunar volatiles.

### I. INTRODUCTION

This chapter presents a critical review from a resources perspective of the available data on the abundances of the noble gases and several important chemically reactive volatile elements (H, C, N, S, F, Cl) in lunar materials. These data are an essential input to studies of lunar resource utilization, but in the past have been difficult to use because they are scattered throughout the vast literature on studies of the Apollo and Luna samples.

The chapter is organized as follows. In Sec. II some general information about the factors affecting the abundances of the solar-wind-implanted volatiles is presented. In Sec. III the extensive analytical data on lunar samples are compiled and critically assessed. The mean abundances of the different volatiles in lunar soils at the different Apollo landing sites, in breccias, and in basalts are derived from this assessment. Where data are available, the mean volatile abundances at the different Luna landing sites are also derived. The volatile abundances determined by different laboratories on lunar soils,

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breccias, and basalts are also tabulated. The mean abundance data are then combined with models of volatile abundance as a function of depth in the lunar regolith to give average global inventories for the volatiles. In Sec. IV, some volatile extraction methods are discussed with an emphasis on thermal extraction and grain size sorting applied to lunar soils. In Sec. V, the isotopic and mineralogical evidence for and against indigenous lunar water is reviewed. In Sec. VI, thermodynamic models of lunar volcanic gases are presented and the implications for the transport of ore-forming metals in lunar volcanic gases are discussed. These models also show that the water abundance in the lunar interior (at the time that volcanism occurred) can potentially be probed by analyzing trapped lunar volcanic gases in lunar samples for chlorofluorocarbon (CFC) gases. Finally, in Sec. VII we summarize some of the major remaining unanswered questions about lunar volatiles and recommend studies to answer these questions.

## II. SOLAR-WIND VOLATILES: GENERAL CONSIDERATIONS

The solar wind consists of energetically charged particles that flow radially outward from the solar corona. Essentially, it represents the continuous expansion of the outer layers of the solar atmosphere. The solar wind can be deflected either by a magnetic field or by an atmosphere, but on a body such as the Moon, which has little of either, the solar wind is only stopped by the surface material. The elemental composition of the solar wind is that of the Sun—predominantly H and He, with the abundance of heavier elements generally decreasing with increasing mass. The only elements for which the solar wind makes the largest contribution to lunar inventories are those which are extremely depleted on the Moon: the noble gases and the volatiles H, C and N. Because the abundances of these elements tend to correlate with one another, we will discuss them together. For a more detailed discussion of solar-wind implantation, focusing on the scientific aspects rather than resource evaluation, see Haskin and Warren (1991).

### A. Factors Affecting Abundances

When a solar-wind particle hits a rock (or mineral grain) on the Moon's surface, it penetrates less than  $1\ \mu\text{m}$ , so only material which has been within  $1\ \mu\text{m}$  of the lunar surface will contain implanted solar-wind particles. One result of the shallow implantation depth of the solar wind is that solar-wind-derived volatiles are concentrated in the finest grain sizes, where the surface-to-volume ratio is largest. This is illustrated in Fig. 1 where analytical results for the abundance of  $^3\text{He}$  in different size fractions in lunar soil 10084 are displayed. Analogous studies of the hydrogen, carbon and nitrogen abundance as a function of grain size in lunar soils have been conducted by several groups (see, e.g., Becker and Clayton 1975, 1977; Bustin et al. 1984, 1986; DesMarais et al. 1973; Frick et al. 1988; Gibson et al. 1987; Goel and Kothari 1972; Goel et al. 1974a, b; Kaplan et al. 1970; Müller 1972, 1973, 1974). These results

generally show that H, C and N are also concentrated in the finer grain size fractions of lunar soils. Other investigations in which chemical etching (e.g., by  $F_2$ ) or depth sensitive analytical techniques are employed also show that the solar-wind-derived volatiles are concentrated on the outer surfaces of grains (see, e.g., Becker 1980; Epstein and Taylor 1975; Filleux et al. 1977,1978; Goldberg et al. 1975; Leich et al. 1973,1974). We note that there is some disagreement between the results of the different  $F_2$  stripping experiments (Becker 1980; Epstein and Taylor 1975) with regard to the exact amount of hydrogen on grain surfaces and its speciation. Overall, the large body of analytical data on the noble gases and the chemically reactive solar-wind-derived volatiles show that in principle, beneficiation by grain-size sorting is possible. However, as we discuss below, it is not clear whether this beneficiation method will prove to be practical.

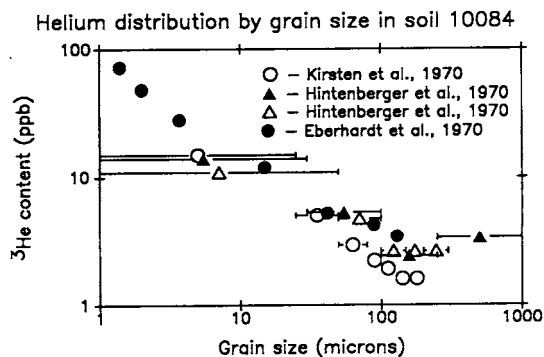


Figure 1. The  $^3\text{He}$  content of Apollo 11 soil 10084 as a function of grain size. The larger contents are found in the smaller particles which have larger surface to volume ratios (figure modified from Swindle et al. 1990).

Another factor affecting the abundance of solar wind volatiles is the "maturity" of a soil—the amount of natural processing that it has undergone. The abundances of the solar-wind-derived volatiles generally correlate with several other parameters that relate to exposure to either the solar wind or impacting micrometeorites. As surface exposure progresses, volatile abundance increases, the mean grain size of the soil decreases, the abundances of agglutinates (composite particles welded together by impacts) increases, and the ratio of fine-grained reduced iron (presumably produced by impact processes) to oxidized iron increases (see, e.g., Morris 1978). Correlations between the abundances of several solar-wind-derived volatiles with the last parameter, usually denoted by  $I_s/\text{FeO}$ , are shown in Fig. 2. Techniques which can remotely sense one of these other parameters (see, e.g., Charette et al. 1976) might be useful in evaluating the resource potential of different areas of the Moon. On the other hand, the maturity of the surface layer, which is all that can be detected by remote sensing, might be unrelated to the maturity

of deeper material, in which case knowledge of surface maturity variations would not be important (Taylor 1991).

The integrated solar-wind fluence varies with location on the Moon as a result of two effects. First, the direction of flow of the solar wind is nearly constant and nearly parallel to the plane of the Moon's rotation, so higher latitudes receive less solar wind per unit area than do equatorial latitudes. Second, the Moon spends about one-quarter of its time (near full Moon) in the tail of the Earth's magnetosphere, which deflects the solar wind (Swindle et al. 1992), so the nearside receives less solar wind exposure than does the farside. The combined result is shown in Fig. 3, which indicates that the central nearside receives <30% of the exposure than that of the central farside. No clear pattern of geography-related variation is apparent in the Apollo samples. The absence of an effect in the Apollo samples might simply reflect the fact that these come from a restricted area on the Moon's nearside. On the other hand, a higher fluence would not translate into a higher volatile abundance of a gas if individual grains are saturated with that gas at the fluence received by the Apollo samples (see Futagami et al. [1990], and Wieler et al. [1980] for different viewpoints and references to other discussions). Currently available data on Luna samples, which come from nearer the eastern limb of the nearside, have higher mean concentrations of all five noble gases and N than do Apollo samples, but, as discussed below, the Luna data only represent a few measurements of mature soils.

The chemistry of the lunar soil seems to be a significant factor only in the case of the light noble gases. Helium (and Ne) contents are much higher (by as much as two orders of magnitude) in ilmenite than in co-existing minerals such as olivine, pyroxene and plagioclase, presumably because diffusion is slower in ilmenite than in the other minerals (see, e.g., Basford 1974; Frick et al. 1988; Hübner et al. 1975; Kirsten et al. 1971; Signer et al. 1977). Because most lunar Ti is in ilmenite, the He and Ne abundances tend to correlate with the Ti abundance (Fig. 4). If maturity effects are taken into account, the correlation is even stronger (Jordan 1989) as illustrated in Fig. 5. As the Ti abundance can be determined remotely by either gamma-ray (Davis 1980) or visible-infrared (Pieters 1978; Johnson et al. 1991) spectroscopy, the He abundance can also be mapped, with some assumptions about maturity variations.

Although there are presumably mineral-dependent abundance effects for the heavier noble gases and H, C and N as well, these are apparently much smaller than the effects for He and Ne (see, e.g., Frick et al. 1988). Therefore, the variations from location to location would be expected to be smaller. We took mean measured volatile abundances and major and minor element abundances for more than 60 Apollo soils, and found that, even if the strong correlations with maturity are taken into account (e.g., by calculating residuals from best-fit lines in plots like Fig. 2), there are few statistically significant (90% level) correlations between the heavy noble gases, H, C or N and other elements. The few correlations present were mostly with Ti, although they

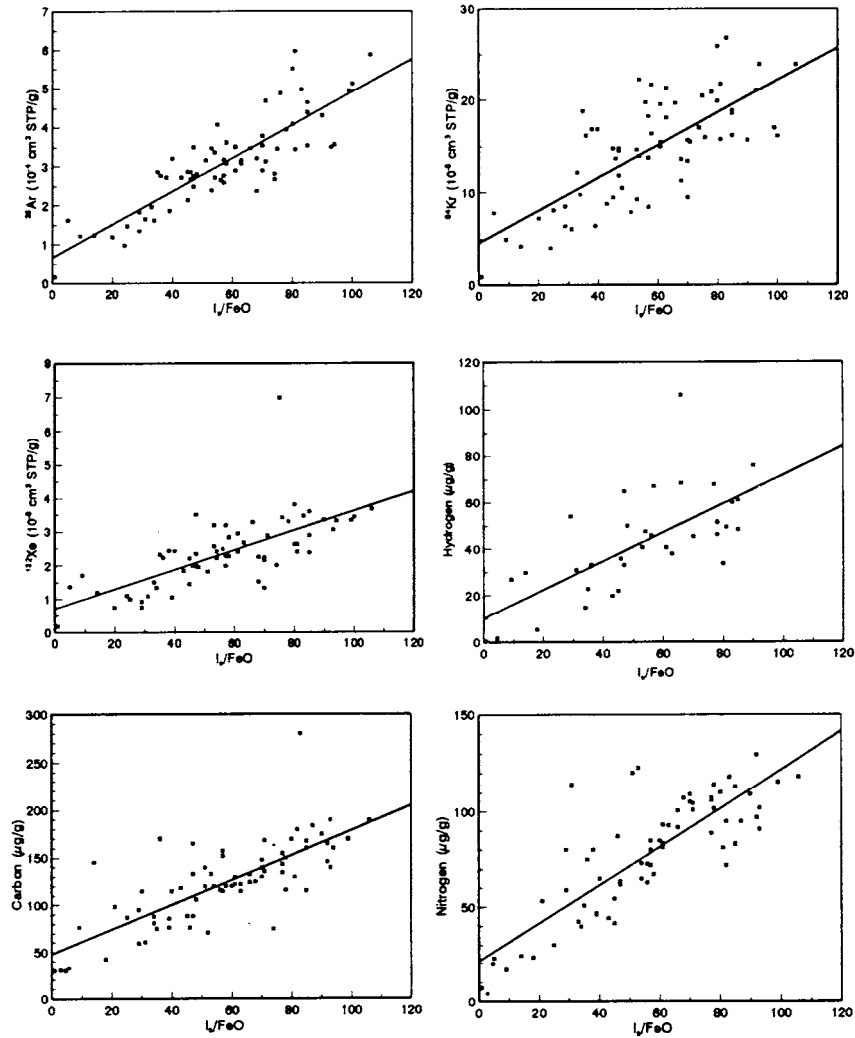


Figure 2. (a) top left: The  $^{36}\text{Ar}$  content of lunar soils as a function of maturity as measured by  $I_s/\text{FeO}$ , the ratio of reduced to oxidized iron. (b) top right: The same for  $^{84}\text{Kr}$ . (c) middle left: The same for  $^{132}\text{Xe}$ . (d) middle right: The same for hydrogen. (e) bottom left: The same for carbon. (f) bottom right: The same for nitrogen. Data for  $^{36}\text{Ar}$ ,  $^{84}\text{Kr}$  and  $^{132}\text{Xe}$  are taken from soil means (references given by Swindle et al. 1990). The data for H, C, and N are taken from literature data compiled by B. Fegley as part of writing this chapter. The  $I_s/\text{FeO}$  data are generally taken from the compilation by Morris (1978). The lines in the figures are unweighted linear least-squares fits to all of the data points in each figure.

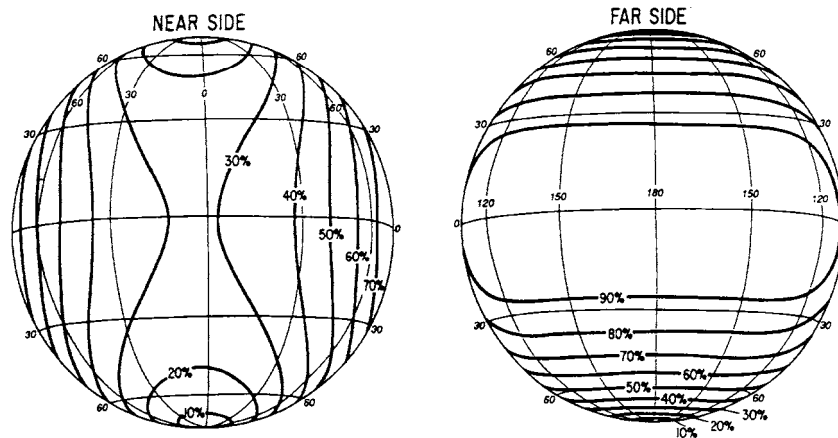


Figure 3. The relative amount of solar-wind exposure for lunar regions as a function of location. The calculations, which are from Swindle et al. (1992), assume that the Moon is completely shielded from the solar wind by the terrestrial magnetotail for 25% of each month, and that the flux is constant otherwise. The contours give percent of maximum (equatorial farside) fluence.

were weaker (and would translate into much smaller abundance variations) than the correlations of these elements with  $I_s/FeO$  or the correlations of He and Ne with Ti.

Finally, some regions with distinctive swirl-like albedo patterns could be the result of deflection of solar-wind ions by local magnetic anomalies (Hood and Schubert 1980; Hood and Williams 1989). If so, there would be corresponding regional variations in the abundance of solar-wind-implanted volatiles, which could be easily mapped.

### III. ESTIMATED INVENTORIES OF LUNAR VOLATILES

In this section, we use the extensive analytical data on the Apollo and Luna samples to make reasonable estimates of the typical abundances of He, Ne, Ar, Kr, Xe, H, C, N, S, F and Cl in soils collected near the surface of the central nearside. These estimates will then be combined with analytical results (where available) and theoretical models on the depth distribution of these elements in the lunar regolith. As a result, we derive average global inventories for each element considered. We also review the abundances of the chemically reactive volatiles H, C, N, S, F and Cl in lunar basalts and breccias. These data, in combination with the data on lunar soils are then used to discuss the water content of the Moon, compositional models for lunar volcanic gases, and theoretical studies of the transport of metals by lunar volcanic gases.

The literature on analyses of the noble gases, H, C, N, S, F and Cl in lunar samples was reviewed by the two authors with Swindle compiling and

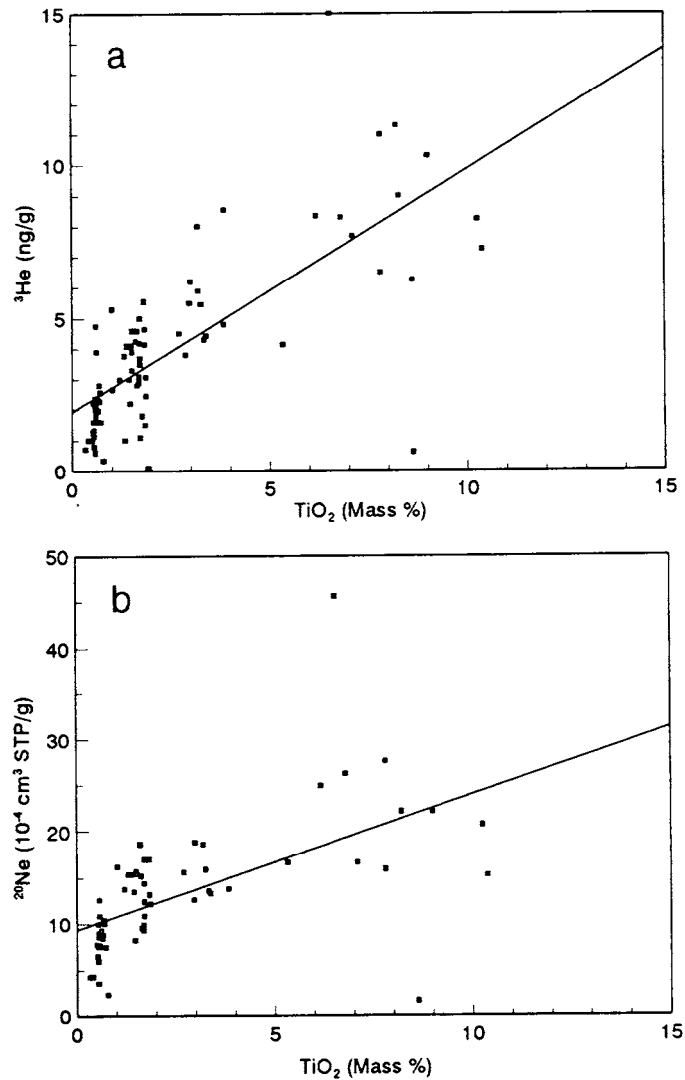


Figure 4. (a) top: A plot of <sup>3</sup>He content (ng g<sup>-1</sup>) vs Ti content (% TiO<sub>2</sub> by mass) in lunar soils. The solid diagonal line is a linear least-squares fit to the values for individual soils. Plot modified from Swindle et al. (1990). (b) bottom: A plot of <sup>20</sup>Ne content (cm<sup>3</sup> STP g<sup>-1</sup>) vs Ti content (% TiO<sub>2</sub> by mass) in lunar soils. The solid diagonal line is a linear least-squares fit to the values for individual soils.

assessing the analytical data for the noble gases and Fegley compiling and assessing the data for the chemically reactive volatiles. The results of this critical assessment are described below starting with the noble gases.

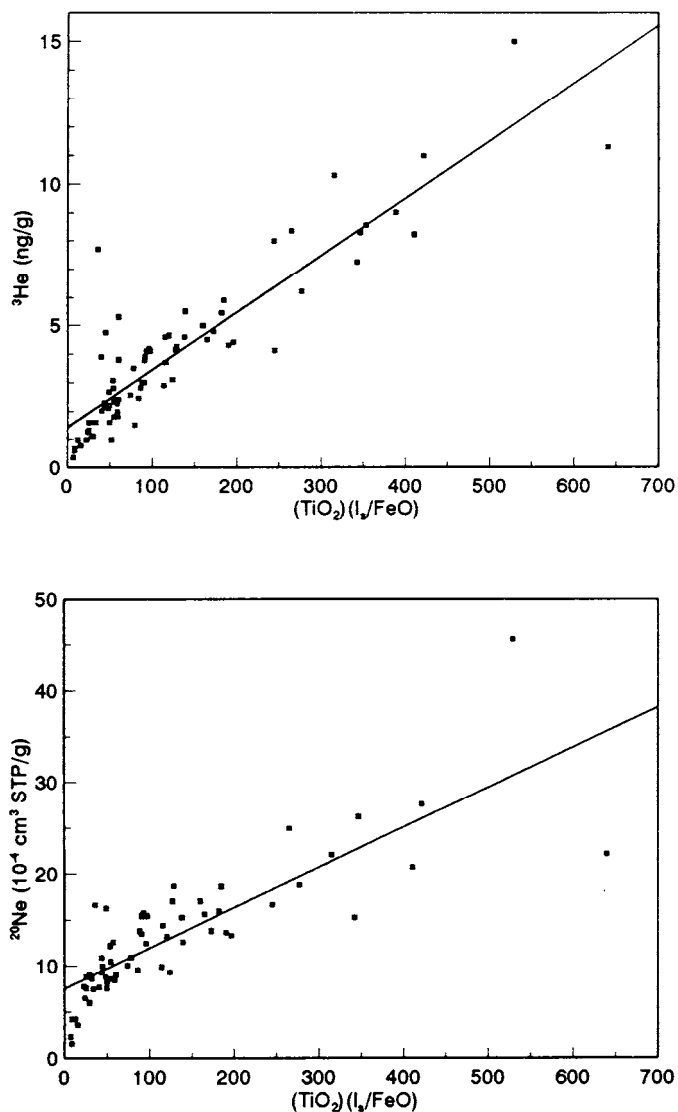


Figure 5. (top) The  $^3\text{He}$  content ( $\text{ng g}^{-1}$ ) plotted against the product of Ti content ( $\% \text{TiO}_2$ ) and  $I_s/\text{FeO}$ . The solid diagonal line is a linear least-squares fit. All soils for which the relevant data are available are included in this plot. Note the smaller amount of scatter relative to that in Fig. 4. Data from compilations by Swindle et al. (1990). (bottom) A similar plot of the  $^{20}\text{Ne}$  content ( $\text{cm}^3 \text{STP g}^{-1}$ ) in lunar soils. Again, note the smaller amount of scatter relative to that in Fig. 4.



### A. Noble Gases

The noble gas data are presented in Tables I (Apollo sites) and II (Luna 16 and 20 sites). The Apollo values in Table I were calculated by first taking the mean of all bulk soil measurements for each soil that has been analyzed (references are given in Swindle et al. 1990), and then combining the values for individual soils to get means and medians for each site. The number of analyses available for a single soil ranges from one (nearly half the soils) to 11 (10084). For the Luna samples, where different identification systems have been used in different reports, all analyses of each site were averaged.

Some values in Table I are skewed by single measurements or single soils. In particular, the Apollo 12 He and Ne give much different means (4.5 ng g<sup>-1</sup> and 13.0 cm<sup>3</sup> STP g<sup>-1</sup>, respectively) if 12030 is deleted. The 12030 numbers come from a single measurement by Heymann et al. (1972). They are about four times as high as the second highest value measured in an Apollo sample (79221). However, the values reported by Heymann's laboratory are not systematically different from other laboratories on the 11 soils where direct comparisons can be made. The Apollo 11 Xe similarly is driven up by one of two measurements of 10010 by Funkhouser et al. (1970) that is about twice as high as most other soils with high Xe contents. Finally, the low end of the Apollo 17 samples is always 74220, the orange soil. Although the 74220 analyses are confirmed by multiple measurements, the soil is certainly unrepresentative. However, enough Apollo 17 soils have been analyzed that it has little effect on the means or medians.

For Ar, Kr and Xe, there is no obvious relationship with landing site for the Apollo sites. Except for Apollo 11, the mean value from each Apollo site is within 1 $\sigma$  of the mean of every other site. The values for Apollo 11 are slightly higher, but this reflects the fact that the few soils analyzed are all mature, and these gases correlate strongly with maturity (Figs. 2a-c). For He and Ne, the Apollo 11 and Apollo 17 means are clearly higher than the other sites. In this case, it reflects the higher mean Ti content of these sites and the correlation of He and Ne with Ti (Fig. 4). The means for the Luna data are higher than the Apollo means for all gases. Although this could indicate a geographic dependence (Sec. II), the Luna 16 and 20 soils are apparently mature (Swindle et al. 1992), and the scatter in reported values is surprisingly high for such a small number of samples.

### B. Hydrogen

The hydrogen contents of lunar samples have generally been determined by four techniques: (1) pyrolysis followed by H<sub>2</sub> analysis by gas chromatography (see, e.g., Bustin et al. 1984,1986; DesMarais et al. 1974; Gibson et al. 1987); (2) pyrolysis followed by manometric measurement of H<sub>2</sub> (see, e.g., Chang et al. 1974b); (3) acid hydrolysis followed by manometric measurement (see, e.g., Chang et al. 1974b) and (4) pyrolysis followed by separation of H<sub>2</sub> and H<sub>2</sub>O which are measured manometrically and isotopically (see, e.g., Epstein and Taylor 1970,1971,1972,1973,1974,1975; Friedman et al.

**TABLE I**  
Noble Gas Contents of Bulk Lunar Soils: Apollo Sites

Mission	No. of Soils	Abundance			
		Min.	Max.	Median	Mean $\pm 1\sigma$
$^3\text{He}$ ( $\text{ng g}^{-1}$ )					
Apollo 11	3	8.0	11.3	9.2	$9.5 \pm 1.7$
Apollo 12	8	2.6	21.0	4.9	$6.6 \pm 5.9$
Apollo 14	7	2.8	4.6	3.5	$3.7 \pm 0.8$
Apollo 15	19	1.0	5.6	3.7	$3.5 \pm 1.1$
Apollo 16	29	0.4	2.8	1.7	$1.7 \pm 0.6$
Apollo 17	24	0.6	15.0	7.2	$6.7 \pm 3.2$
All Soils	90	0.4	21	3.6	$4.2 \pm 3.4$
$^{20}\text{Ne}$ ( $10^{-4} \text{ cm}^3 \text{STP g}^{-1}$ )					
Apollo 11	3	22.1	25.6	23.9	$23.9 \pm 1.8$
Apollo 12	6	7.3	61.4	14.6	$21.1 \pm 20.0$
Apollo 14	7	9.3	15.2	10.9	$11.5 \pm 2.2$
Apollo 15	12	8.2	17.0	14.9	$14.4 \pm 2.4$
Apollo 16	29	2.4	12.6	7.9	$7.6 \pm 2.5$
Apollo 17	18	1.6	45.6	18.7	$19.7 \pm 8.7$
All Soils	75	1.6	61.4	12.6	$13.7 \pm 9.0$
$^{36}\text{Ar}$ ( $10^{-4} \text{ cm}^3 \text{STP g}^{-1}$ )					
Apollo 11	3	3.4	3.9	3.8	$3.7 \pm 0.2$
Apollo 12	6	1.0	2.9	2.6	$2.2 \pm 0.8$
Apollo 14	7	2.4	4.4	3.2	$3.2 \pm 0.6$
Apollo 15	12	1.6	3.6	2.8	$2.7 \pm 0.7$
Apollo 16	29	1.2	5.9	3.5	$3.4 \pm 1.4$
Apollo 17	18	0.2	6.0	3.1	$2.9 \pm 1.2$
All Soils	75	0.2	6.0	3.1	$3.1 \pm 1.2$
$^{84}\text{Kr}$ ( $10^{-8} \text{ cm}^3 \text{STP g}^{-1}$ )					
Apollo 11	3	20.5	22.0	20.9	$21.1 \pm 0.8$
Apollo 12	6	4.0	19.8	13.8	$12.0 \pm 6.4$
Apollo 14	6	9.3	19.7	17.7	$15.8 \pm 4.4$
Apollo 15	10	5.9	23.9	12.0	$13.5 \pm 6.2$
Apollo 16	28	4.9	26.8	14.8	$14.1 \pm 5.7$
Apollo 17	18	0.8	22.3	16.3	$14.9 \pm 6.2$
All Soils	71	0.8	26.8	15.5	$14.5 \pm 5.8$
$^{132}\text{Xe}$ ( $10^{-8} \text{ cm}^3 \text{STP g}^{-1}$ )					
Apollo 11	3	3.0	7.0	3.1	$4.4 \pm 2.3$
Apollo 12	6	1.1	2.5	2.0	$1.7 \pm 0.6$
Apollo 14	6	2.0	3.6	2.9	$2.8 \pm 0.6$
Apollo 15	10	0.8	3.3	1.9	$2.0 \pm 0.8$
Apollo 16	28	0.8	3.8	2.4	$2.4 \pm 0.9$
Apollo 17	18	0.2	3.1	2.3	$2.1 \pm 0.7$
All Soils	71	0.2	7	2.4	$2.4 \pm 1.0$

**TABLE II**  
Noble Gas Contents of Bulk Lunar Soils: Luna Sites

Mission	No. of Soils	Abundance			
		Min.	Max.	Median	Mean $\pm 1\sigma$
$^3\text{He}$ ( $\text{ng g}^{-1}$ )					
Luna 16	5	3.4	10.8	8.4	$7.5 \pm 2.9$
Luna 20	3	1.5	3.0	2.3	$2.3 \pm 0.8$
$^{20}\text{Ne}$ ( $10^{-4} \text{ cm}^3 \text{STP g}^{-1}$ )					
Luna 16	5	11.2	75.1	25.3	$32.6 \pm 25.3$
Luna 20	4	9.2	17.8	11.5/14.3	$13.2 \pm 3.7$
$^{36}\text{Ar}$ ( $10^{-4} \text{ cm}^3 \text{STP g}^{-1}$ )					
Luna 16	5	2.7	8.6	5.4	$5.4 \pm 2.2$
Luna 20	4	4.8	6.5	4.9/5.8	$5.5 \pm 0.8$
$^{84}\text{Kr}$ ( $10^{-8} \text{ cm}^3 \text{STP g}^{-1}$ )					
Luna 16	4	9.0	35.2	22.0/25.3	$22.9 \pm 10.8$
Luna 20	3	13.8	23.6	23.1	$20.2 \pm 5.5$
$^{132}\text{Xe}$ ( $10^{-8} \text{ cm}^3 \text{STP g}^{-1}$ )					
Luna 16	4	1.4	8.5	2.2/5.6	$4.4 \pm 3.3$
Luna 20	3	1.9	3.4	3.0	$2.8 \pm 0.8$

1970,1971,1972; Merlivat et al. 1974,1976; Stievenard et al. 1990). The results of hydrogen determinations on bulk lunar soils are listed by Apollo mission in Table III and by analysis in Table IV.

**TABLE III**  
Hydrogen Contents of Bulk Lunar Soils

Mission	No. of Soils	Hydrogen Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
Apollo 11	1	38	66	46/54.2	$52 \pm 10$
Apollo 12	5	1.9	46	30/38	$31 \pm 14$
Apollo 14	5	35.9	106	61/67	$67 \pm 25$
Apollo 15	14	28	120	52.6	$57 \pm 22$
Apollo 16	15	19	79	36.3/39	$46 \pm 17$
Apollo 17	14	0.2	53	22/25.7	$26 \pm 18$
All Soils <sup>a</sup>	54	0.2	120	45	$46 \pm 16$

<sup>a</sup> Excluding the orange soil 74220, the minimum value becomes  $1.9 \mu\text{g g}^{-1}$  and the median and mean values are unchanged.

The first thing to note is that the reported hydrogen contents range over a factor of 600 from a low of  $\sim 0.2 \mu\text{g g}^{-1}$  reported by Epstein and Taylor (1973) for the orange soil 74220 to a high of  $\sim 120 \mu\text{g g}^{-1}$  reported by Friedman et

**TABLE IV**  
Comparison of Analytical Results for Hydrogen in Bulk Lunar Soils

Reference	No. of Analyses	Hydrogen Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
Bustin et al. 1984	16	17	106	47/50	49 $\pm$ 22
Bustin et al. 1986	5	26	66	39	41 $\pm$ 16
Chang et al. 1974 <i>b</i>	7	0.6	22	7.8	11 $\pm$ 8
DesMarais et al. 1974	18	31	92	62	59 $\pm$ 15
Epstein and Taylor 1970	2	45	46	—	46
Epstein and Taylor 1971	3	1.9	40.7	38	27 $\pm$ 23
Epstein and Taylor 1972	3	35.9	52.6	49.4	46 $\pm$ 10
Epstein and Taylor 1973	4	0.2	61.7	7.9/28.8	25 $\pm$ 30
Epstein and Taylor 1975	13	28.2	58.5	36.3	40 $\pm$ 9
Friedman et al. 1971	2	30	46	—	38
Friedman et al. 1972	5	66	120	85	89 $\pm$ 20
Gibson et al. 1987	5	25.7	54.2	39.2	41 $\pm$ 11
Merlivat et al. 1974	6	28	53	34.5/36.1	38 $\pm$ 9
All Soils <sup>a</sup>	89	0.2	120	45	46 $\pm$ 22

<sup>a</sup> Excluding the two analyses of the orange soil 74220 (0.2 and 0.6  $\mu\text{g g}^{-1}$ ) the minimum value becomes 1.9  $\mu\text{g g}^{-1}$  and the median and mean values are unchanged.

al. (1972) for lunar soil 15271. The latter value appears anomalously high by comparison with another split of the same soil which has a hydrogen content of 38  $\mu\text{g g}^{-1}$  (Bustin et al. 1984). However, the Friedman et al. (1972) value is a combination of hydrogen found as elemental hydrogen and hydrogen found combined in water. If the Friedman et al. (1972) analysis of 15271 were not included in the data set, the highest hydrogen content in a lunar soil would then be 106  $\mu\text{g g}^{-1}$  for 14003 (Bustin et al. 1984).

On the other hand, the low hydrogen content of 74220 is apparently real. Chang et al. (1974*b*) reported a value of 0.6  $\mu\text{g g}^{-1}$  hydrogen for another split of the orange soil. This soil also has an unusually low carbon content of  $\sim 4$  to 11  $\mu\text{g g}^{-1}$  (Epstein and Taylor 1973; Chang et al. 1974*b*; Moore et al. 1974). However, the low hydrogen content of 74220 is probably not representative of the hydrogen contents of most bulk lunar soils. As Table III shows, the median hydrogen content of 54 soils is 45  $\mu\text{g g}^{-1}$  and the mean hydrogen content of the same soils is 46  $\pm$  16  $\mu\text{g g}^{-1}$ . These values do not change if the data for 74220 are excluded from the statistical calculations. Excluding the possibly suspect value of 120  $\mu\text{g g}^{-1}$  for 15271 also has only a small effect on the calculated mean which decreases to 45  $\mu\text{g g}^{-1}$ .

Another important point illustrated by the data in Table III is that the hydrogen contents of bulk lunar soils at the different Apollo sites are about the same. To a first approximation, the mean hydrogen contents of bulk soils at the Apollo 12 and Apollo 17 sites appear slightly lower than the mean

hydrogen contents of bulk soils at the other four Apollo sites. This difference is not significant at the  $1\sigma$  level, but it is important to remember that this simple comparison does not attempt to take any chemical and mineralogical differences into account.

When an attempt is made to consider variations in soil properties a slightly different picture emerges. Hydrogen, unlike He, does not correlate with either the Ti content of lunar soils or the product of Ti content and  $I_s/FeO$ . However, as Fig. 2d illustrates, the hydrogen content does correlate with  $I_s/FeO$  itself, with more hydrogen being found in more mature soils with higher  $I_s/FeO$  values. This correlation may be responsible for the generally low hydrogen contents reported by Chang et al. (1974b) in immature and submature soils from the Apollo 17 landing site. But the considerable scatter in Fig. 2d suggests that it may not be economically advantageous to beneficiate soils on the basis of maturity for hydrogen mining.

The hydrogen contents of lunar basalts and breccias (soil and regolith) are given in Table V. The hydrogen contents of breccias are variable and range over 2 orders of magnitude from a low of  $1 \mu\text{g g}^{-1}$  in breccia 76015 (Gibson et al. 1987) to a high of  $101 \mu\text{g g}^{-1}$  in breccia 10061 (Epstein and Taylor 1970). The median and mean values of  $20 \mu\text{g g}^{-1}$  and  $28 \mu\text{g g}^{-1}$ , respectively, are slightly lower than the corresponding values for bulk lunar soils, although the  $1\sigma$  uncertainties on the means for the soils and breccias overlap.

In contrast, the hydrogen contents of lunar basalts are only a few  $\mu\text{g g}^{-1}$  or less with a maximum value of  $12 \mu\text{g g}^{-1}$  for 15058 (DesMarais et al. 1974) and a minimum value of  $<0.6 \mu\text{g g}^{-1}$  for 10072 (Epstein and Taylor 1970). The most recent study of hydrogen in lunar basalts by Stievenard et al. (1990) gives only  $\sim 1 \mu\text{g g}^{-1}$  hydrogen in 70215, a basalt which has been studied extensively by Merlivat et al. (1974,1976) as well. Stievenard et al. (1990) note that the hydrogen in 70215 contains a deuterium-rich component and thus is probably formed by spallogenic processes. Their studies as well as those of Merlivat et al. (1974,1976) on splits of the sample indicates that the indigenous hydrogen content of lunar basalts is below  $1 \mu\text{g g}^{-1}$ . These results have important implications for the formation of lunar ore deposits by volcanic gas transport in the lunar interior (see, e.g., Fegley 1991).

### C. Carbon

The carbon contents of lunar samples have been measured by a variety of techniques: (1) pyrolysis followed by manometry and mass spectrometry (see, e.g., Epstein and Taylor 1970); (2) pyrolysis, or combustion in  $O_2$  followed by gas chromatography (GC) or by gas chromatography mass spectrometry (see, e.g., Burlingame et al. 1971; DesMarais et al. 1973; DesMarais 1978; Gibson and Moore 1973; Moore et al. 1970); (3) solvent extraction followed by GC-MS (see, e.g., Burlingame et al. 1971); and (4) dissolution/hydrolysis with mineral acids and/or deuterated acids followed by gas chromatography, mass spectrometry, or GC-MS (see, e.g., Cadogan et al. 1972; Chang et al. 1970; Sakai et al. 1972; Wszolek et al. 1972). The analytical results for bulk

**TABLE V**  
Hydrogen Contents of Lunar Breccias and Basalts

Reference	No. of Analyses	Hydrogen Abundance $\mu\text{g g}^{-1}$				Mean $\pm 1\sigma$
		Min.	Max.	Median		
<b>Breccias</b>						
Epstein and Taylor 1970	1	101	101	101	101	101
Friedman et al. 1970	3	50	66	60	59 $\pm$ 10	
Friedman et al. 1972	4	8	38	20	22 $\pm$ 15	
Gibson et al. 1987	13	1	60.4	3.5	16 $\pm$ 23	
Merlivat et al. 1974	2	34.5	39.1	—	37	
All Breccias	23	1	101	20	28 $\pm$ 28	
<b>Basalts</b>						
DesMarais et al. 1974	2	4	12	—	8	
Epstein and Taylor 1971	1	<0.6	<0.6	<0.6	<0.6	
Friedman et al. 1971	3	3	9	6.6	6.2 $\pm$ 3.5	
Friedman et al. 1972	2	2	2.8	—	2.4	
Gibson et al. 1987	16	1	3.8	1.8	2.0 $\pm$ 0.7	
Merlivat et al. 1974	3	0.65	2.42	1.51	1.5 $\pm$ 0.9	
Merlivat et al. 1976	8	1.25	2.96	1.55/1.98	2.0 $\pm$ 0.6	
Stievenard et al. 1990	1	0.97	0.97	0.97	0.97	
All Basalts	36	<0.6	12	2	2.7 $\pm$ 2.4	

**TABLE VI**  
Carbon Contents of Bulk Lunar Soils

Mission	No. of Soils	Carbon Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
Apollo 11	2	116	226	155/157	162 $\pm$ 27
Apollo 12	13	23	250	120	113 $\pm$ 46
Apollo 14	12	42	186	120	117 $\pm$ 35
Apollo 15	35	2.5	175	98	94 $\pm$ 44
Apollo 16	33	31	280	140	125 $\pm$ 52
Apollo 17	39	3.5	200	130/135	124 $\pm$ 45
All Soils	134	2.5	280	120	124 $\pm$ 45

lunar soils are listed in Tables VI and VII.

The reported carbon contents range over 2 orders of magnitude, but the median and mean values are within a factor of 3 of the corresponding values for hydrogen. The data in Table VI apparently indicate that the carbon contents of bulk soils at the Apollo 11 site are slightly higher than those at other sites. This apparent difference is not statistically significant and because of the small number of samples does not bias the calculated median and mean. The highest carbon content reported for any bulk lunar soil is  $280 \mu\text{g g}^{-1}$  for 64421 (Moore et al. 1973). The lowest reported carbon content is  $2.5 \mu\text{g g}^{-1}$  for 15426 (Modzeleski et al. 1972). There are no replicates for either analysis. Four analyses of different splits of the orange soil 74220 give carbon contents of  $3.5 \mu\text{g g}^{-1}$  (Epstein and Taylor 1973),  $5 \mu\text{g g}^{-1}$  (Moore et al. 1974),  $11 \mu\text{g g}^{-1}$  (Chang et al. 1974b), and  $100 \mu\text{g g}^{-1}$  (Moore et al. 1974). A comparison of the results obtained by different analytical groups in Table VII suggests some possible discrepancies between the different laboratories. The generally lower values reported by Wszolek et al. (1972) are for the Apollo 15 deep core (15001–15006) and for sample 15426. These values were determined by DF hydrolysis and are lower than carbon contents determined by other groups (Epstein and Taylor 1975; DesMarais et al. 1973) using pyrolysis on different splits of the same samples. The carbon contents determined by Modzeleski et al. (1972) for Apollo 15 samples are also generally lower than analyses by other groups on other splits of the same samples. However, within the calculated  $1\sigma$  uncertainties, the results of Wszolek et al. (1972) and Modzeleski et al. (1972) overlap those of other investigators. The calculated mean carbon content of bulk soils will not be significantly changed by removing these data from the computations.

Table VIII displays the carbon contents of lunar basalts and breccias. As with hydrogen, the calculated mean value is slightly lower than that for bulk soils, but overlaps the soil value within the  $1\sigma$  uncertainties. The reported carbon contents for breccias range over almost a factor of 80, and span almost the entire range covered by bulk lunar soils. The maximum of  $385 \mu\text{g g}^{-1}$  is

**TABLE VII**  
Comparison of Analytical Results for Carbon in Bulk Lunar Soils

Reference	No. of Analyses	Carbon Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
Burlingame et al. 1970	2	168	185	—	176
Chang et al. 1970	1	157	157	157	157
Chang et al. 1974a	7	11	88	50	56 $\pm$ 28
Chang et al. 1974b	6	129	160	135/141	140 $\pm$ 11
DesMarais et al. 1973	22	52	125	81	83 $\pm$ 19
DesMarais et al. 1975	12	36	152	120/123	111 $\pm$ 35
Epstein and Taylor 1970	2	143	155	—	149
Epstein and Taylor 1971	3	30	145	125	100 $\pm$ 68
Epstein and Taylor 1972	3	43	89	60	64 $\pm$ 27
Epstein and Taylor 1973	4	3.5	141	95/97	84 $\pm$ 67
Epstein and Taylor 1975	8	51	216	101/144	123 $\pm$ 50
Friedman et al. 1970	1	188	188	188	188
Friedman et al. 1971	3	116	139	133	129 $\pm$ 14
Friedman et al. 1972	5	51	110	99	88 $\pm$ 25
Gibson and Moore 1973	4	131	184	150/154	155 $\pm$ 26
Kaplan and Petrowski 1971	9	37	140	112	105 $\pm$ 28
Kaplan et al. 1970	4	116	170	143/147	144 $\pm$ 26
Kaplan et al. 1976	10	81	154	125/127	122 $\pm$ 24
Kerridge et al. 1975b	14	31	180	92/126	116 $\pm$ 43
Kerridge et al. 1978a	18	23	163	111/119	106 $\pm$ 43
Modzeleski et al. 1972	26	2.5	100.5	35.2/35.4	41 $\pm$ 18
Moore and Lewis 1975	37	59	200	140	142 $\pm$ 31
Moore et al. 1970	2	142	226	—	184
Moore et al. 1971	22	23	250	130/135	122 $\pm$ 53
Moore et al. 1972	15	42	186	135	128 $\pm$ 36
Moore et al. 1973	56	29	280	130	128 $\pm$ 43
Moore et al. 1974	40	5	200	140	131 $\pm$ 42
Sakai et al. 1972	2	100	138	—	119
Wszolek et al. 1972	6	29	92	36/43	49 $\pm$ 25
All Soils	344	2.5	280	120	124 $\pm$ 45

higher than the maximum carbon content of  $280 \mu\text{g g}^{-1}$  in bulk soils. The carbon contents of basalts vary over a smaller range of  $\sim 5$  to  $70 \mu\text{g g}^{-1}$  with a mean of  $26 \mu\text{g g}^{-1}$ .

The carbon analyses of breccias are generally in good agreement. In the case of 14321, the different results are 21, 22, 28, 32, 43  $\mu\text{g g}^{-1}$  (Moore et al. 1972), 24  $\mu\text{g g}^{-1}$  (Epstein and Taylor 1972), and 27  $\mu\text{g g}^{-1}$  (Sakai et al. 1972). For 79135, the results are  $150 \mu\text{g g}^{-1}$  (Moore et al. 1974), and 136 and  $157 \mu\text{g g}^{-1}$  (Becker and Epstein 1981). However, there is also



**TABLE VIII**  
Carbon Contents of Lunar Breccias and Basalts

Reference	No. of Analyses	Carbon Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
<b>Breccias</b>					
Becker and Epstein 1981	6	99	385	138/152	188 $\pm$ 96
Epstein and Taylor 1970	1	262	262	262	262
Epstein and Taylor 1972	1	24	24	24	24
Friedman et al. 1970	3	113	218	181	171 $\pm$ 62
Friedman et al. 1972	4	22	50	45/50	42 $\pm$ 14
Kaplan et al. 1970	4	132	198	137/181	162 $\pm$ 32
Kaplan et al. 1976	2	62	76	—	69
Moore et al. 1970	2	102	230	—	166
Moore et al. 1971	2	65	120	—	93
Moore et al. 1972	19	21	225	80	94 $\pm$ 66
Moore et al. 1973	24	5	160	39/45	54 $\pm$ 42
Moore et al. 1974	6	21	150	54/85	73 $\pm$ 46
Sakai et al. 1972	1	27	27	27	27
All Breccias	75	5	385	65/70	93 $\pm$ 73
<b>Basalts</b>					
DesMarais 1978	9	5.3	41.1	8.6	16 $\pm$ 12
Friedman et al. 1971	3	27	61	42	43 $\pm$ 20
Friedman et al. 1972	2	7.3	7.7	—	7.5
Kaplan and Petrowski 1971	2	14	19	—	16
Moore et al. 1970	2	64	70	—	67
Moore et al. 1972	1	35	35	35	35
Moore et al. 1973	6	10	27	13/16	16 $\pm$ 6
Moore et al. 1974	3	31	65	54	50 $\pm$ 20
All Basalts	28	5.3	70	18/19	26 $\pm$ 20

considerable disagreement between some replicate analyses, apparently due to heterogeneous carbon contents. For example, Becker and Epstein (1981) report 220 and 385  $\mu\text{g g}^{-1}$  for 10018, and Moore et al. (1972) report 140 and 210  $\mu\text{g g}^{-1}$  for 14047. Presumably this heterogeneity accounts for at least some of the scatter in the analytical results.

A comparison of the carbon analyses of basalts indicates that some real disagreements exist between different analytical groups. For example, the three basalts 15058, 70215 and 75035 have been analyzed for carbon by DesMarais (1978) and Moore et al. (1973,1974). In all cases higher carbon contents are reported by Moore and colleagues. The mean carbon content for the lunar basalts analyzed by Moore et al. (1970,1972,1973,1974), repre-

senting nearly half the total data, is  $34 \pm 23 \mu\text{g g}^{-1}$ , while the mean carbon content for lunar basalts analyzed by all other investigators is  $20 \pm 16 \mu\text{g g}^{-1}$ . However, both values are well within the  $1\sigma$  uncertainty of the total mean, and they disagree with each other by less than the  $1\sigma$  uncertainty in either.

#### D. Nitrogen

Table IX gives the nitrogen contents of bulk lunar soils at the different Apollo landing sites and at two of the Luna landing sites. These data were obtained by the following analytical techniques: (1) pyrolysis and/or combustion in  $\text{O}_2$  followed by mass spectrometry (see, e.g., Becker and Clayton 1975; Kerridge et al. 1975*a, b*); (2) dissolution/hydrolysis with mineral acids followed by mass spectrometry (see, e.g., Chang et al. 1974*a, b*; Kerridge et al. 1975*a, b*); (3) neutron activation analysis (see, e.g., Goel and Kothari 1972); (4) pyrolysis followed by gas chromatography (see, e.g., Moore et al. 1970); (5) Kjeldahl analysis for chemically bound N (see, e.g., Müller 1972); and (6) spark source mass spectrometry (see, e.g., Morrison et al. 1970).

**TABLE IX**  
Nitrogen Contents of Bulk Lunar Soils

Mission	No. of Soils	Nitrogen Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
Apollo 11	2	93	153	110	$121 \pm 27$
Apollo 12	12	5.9	140	71.1	$67 \pm 34$
Apollo 14	2	80	92	80/92	86
Apollo 15	19	18	135	96	$81 \pm 31$
Apollo 16	41	4	141	90	$85 \pm 34$
Apollo 17	35	7.3	130	73	$72 \pm 29$
LUNA 16	1	242	242	242	242
LUNA 20	1	102	102	102	102
All Soils	111	7.3	242	84	$81 \pm 37$

The reported nitrogen contents vary over a factor of 30 from a low of  $7.3 \mu\text{g g}^{-1}$  in 74220 (Chang et al. 1974*b*) to a high of  $242 \mu\text{g g}^{-1}$  in the Luna 16 soil (Goel et al. 1974*b*). There are no replicates by other groups for either analysis. Nitrogen, like carbon and hydrogen, does not show clear variations between the different Apollo sites. However, the one set of analyses for the Luna 16 soil show markedly higher nitrogen contents. As shown in Fig. 2f, nitrogen contents show a strong correlation with  $I_s/\text{FeO}$ .

A comparison of the nitrogen analyses by different groups is given in Table X. The lowest values were reported by Chang et al. (1974*b*) for submature and immature Apollo 17 soils including the orange soil 74220. The nitrogen contents reported by Chang et al. (1974*b*) follow a trend also exhibited by hydrogen and carbon in these soils and confirmed for these two elements by analyses done by other groups. With the exception of the Luna 16 analyses

reported by Goel et al. (1974*b*), no other group clearly stands out from the rest on the high side. However, a soil by soil comparison of analyses done by different groups generally shows that the results of Moore and colleagues tend to be higher than other analyses on the same soil. Given the agreement between the other analytical groups, especially the UCLA and Chicago laboratories, some of the results of Moore and colleagues may be anomalously high. However, their median and mean values do not appear out of line with the other values summarized in Table X and do not affect the overall mean.

**TABLE X**  
Comparison of Nitrogen Analyses of Bulk Lunar Soils

Reference	No. of Analyses	Nitrogen Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
Becker and Clayton 1975	15	43	114	83	80 $\pm$ 27
Becker and Clayton 1977	9	22.7	70	44	45 $\pm$ 13
Becker and Clayton 1978	2	37	75	—	56
Becker et al. 1976	3	17	62	51	43 $\pm$ 27
Chang et al. 1974 <i>a</i>	7	7.3	54.6	23.4	29 $\pm$ 17
Chang et al. 1974 <i>b</i>	8	88	135	96/98	101 $\pm$ 15
Goel and Kothari 1972	1	80	80	80	80
Goel et al. 1974 <i>a</i>	8	65	242	102/120	119 $\pm$ 54
Goel et al. 1974 <i>b</i>	3	102	242	102/242	195 $\pm$ 83
Kaplan et al. 1976	10	33	110	90/91	79 $\pm$ 27
Kerridge et al. 1975 <i>a</i>	29	15	125	68	72 $\pm$ 33
Kerridge et al. 1975 <i>b</i>	14	4	125	68/83	74 $\pm$ 35
Kerridge et al. 1978 <i>b</i>	18	5.9	74.8	53.6/54.6	47 $\pm$ 26
Kothari and Goel 1973	10	58	141	95/98	101 $\pm$ 26
Moore and Lewis 1975	40	61	137	96/97	99 $\pm$ 19
Moore et al. 1970	2	102	153	—	128
Moore et al. 1971	15	46	140	96	94 $\pm$ 28
Morrison et al. 1970	1	110	110	110	110
Morrison et al. 1971	1	40	40	40	40
Müller 1972	5	80	109	92	91 $\pm$ 12
Müller 1973	9	70	124	96	97 $\pm$ 20
Müller 1974	10	23	110	70/73	69 $\pm$ 27
Müller et al. 1976	3	93.4	110	94	99 $\pm$ 10
Wszolek et al. 1972	6	18	89	21/36	45 $\pm$ 33
All Soils	229	4	242	87	81 $\pm$ 37

The nitrogen analyses of basalts and breccias, which are summarized in Table XI, follow the trends already noted for hydrogen and carbon. The mean nitrogen content of breccias is slightly lower than that of bulk soils, but the two overlap within their mutual uncertainties. Likewise, the nitrogen contents of basalts are again markedly lower, although the nitrogen analyses display

more variability than the carbon analyses. The results for basalts apparently fall into two groups with one group being  $\leq 2 \mu\text{g g}^{-1}$  and the other group being higher than  $\sim 20 \mu\text{g g}^{-1}$ . It is tempting to ascribe the higher analyses to contamination and/or other analytical problems. If this explanation is valid, then the true nitrogen content of lunar basalts is  $\sim 1 \mu\text{g g}^{-1}$  or less (see, e.g., DesMarais 1978).

**TABLE XI**  
Nitrogen Contents of Lunar Breccias and Basalts

Reference	No. of Analyses	Nitrogen Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
Breccias					
Becker and Clayton 1975	6	4.3	62	48	$37 \pm 23$
Becker and Epstein 1981	6	72.3	114.1	74.3/83	$87 \pm 17$
Becker et al. 1976	3	1.8	7.5	2.5	$4 \pm 2$
Frick et al. 1988	1	114	114	114	114
Goel and Kothari 1972	8	22	136	41/55	$58 \pm 34$
Kaplan et al. 1976	2	41	55	—	48
Kothari and Goel 1973	3	16	74	22	$37 \pm 26$
Moore et al. 1970	2	98	125	—	112
Moore et al. 1972	2	57	130	—	94
Müller 1972	1	131	131	131	131
All Breccias	34	1.8	136	62	$61 \pm 40$
Basalts					
Becker and Clayton 1975	3	0.6	3.5	1	$1.7 \pm 1.2$
Becker et al. 1976	1	0.5	0.5	0.5	0.5
DesMarais 1978	9	0.1	1.0	0.7	$0.6 \pm 0.4$
Goel and Kothari 1972	1	21	21	21	21
Kothari and Goel 1973	1	18	18	18	18
Moore et al. 1970	2	30	116	—	73
Müller 1972	3	<10	64	—	—
Müller 1974	1	<8	<8	<8	<8
Müller et al. 1976	7	3	127	5	$39 \pm 55$
All Basalts	28	0.1	127	3.1	$19 \pm 38$

### E. Sulfur

Sulfur contents of lunar samples have been determined by the following methods: (1) spark source mass spectrometry (Bouchet et al. 1971; Morrison et al. 1970); (2) dissolution/hydrolysis with mineral acids followed by mass spectrometry (see, e.g., Chang et al. 1974a, b; Kaplan et al. 1970; Kerridge et al. 1975a, b; Rees and Thode 1972); (3) pyrolysis or combustion in  $\text{O}_2$  followed

by mass spectrometry (see, e.g., DesMarais 1978); (4) X-ray fluorescence (see, e.g., Palme et al. 1978; Wänke et al. 1975); (5) fusion and SO<sub>2</sub> titration (see, e.g., Moore et al. 1972); and (6) combustion in O<sub>2</sub> followed by quantitative infrared spectroscopy on SO<sub>2</sub> (see, e.g., Gibson and Moore 1973). The analytical results for bulk lunar soils are listed in Tables XII and XIII, and the sulfur contents of lunar basalts and breccias are displayed in Table XIV. These data illustrate several important points.

**TABLE XII**  
Sulfur Contents of Bulk Lunar Soils

Mission	No. of Soils	Sulfur Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
Apollo 11	2	640	1750	770	922 $\pm$ 466
Apollo 12	14	530	1100	756	745 $\pm$ 108
Apollo 14	9	682	1149	900	865 $\pm$ 133
Apollo 15	18	337	950	654/669	638 $\pm$ 123
Apollo 16	35	320	829	592	591 $\pm$ 141
Apollo 17	37	212	1330	790/795	786 $\pm$ 261
All Soils	115	212	1750	704/706	715 $\pm$ 216

First, sulfur is much more abundant (on a mass basis) in lunar soils than H, C and N, and is the second most abundant chemically reactive volatile on an atomic basis. The S/C mass ratio is  $\sim 6$ , and the H/S atomic ratio is  $\sim 2$ . The high sulfur abundance means that extraction of other volatiles (e.g., by heating) may also be accompanied by the release of large amounts of sulfur gases if the major S-bearing mineral, troilite (FeS) decomposes during the extraction process.

Second, the sulfur content of lunar soils is much less variable than the H, C and N contents and varies by less than a factor of 8. The lowest value of  $212 \mu\text{g g}^{-1}$  was reported by Chang et al. (1974a) for the orange soil 74220 and the highest value of  $1750 \mu\text{g g}^{-1}$  was reported by Bouchet et al. (1971) for 10084. However, both of these extreme values may be incorrect. Two replicate analyses on 10084 by Kaplan et al. (1970) give  $770 \mu\text{g g}^{-1}$  sulfur, and other analyses of 74220 give higher sulfur contents in the range of  $442 \mu\text{g g}^{-1}$  (Chang et al. 1974b) up to  $830 \mu\text{g g}^{-1}$  (Gibson and Moore 1973; Gibson and Andrawes 1978b). Other sulfur determinations by Bouchet et al. (1971), who used spark source mass spectrometry, are generally higher than analyses by other groups on the same samples. Chang et al. (1974b) used acid hydrolysis to determine sulfur in 74220 and other Apollo 17 soils. They commented on a relationship between decreasing sulfur content and increasing <sup>34</sup>S isotopic enrichment in different aliquots of 74220 but stated that it is not due to incomplete H<sub>2</sub>S release by acid hydrolysis of troilite. Furthermore, we note that a comparative study of acid hydrolysis and fusion techniques by Rees and Thode (1974) shows good agreement between results

**TABLE XIII**  
Comparison of Sulfur Analyses of Bulk Lunar Soils

Reference	No. of Analyses	Sulfur Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
Bouchet et al. 1971	2	1100	1750	—	1425
Chang et al. 1974a	9	212	1199	578	623 $\pm$ 297
Chang et al. 1974b	5	556	950	687	709 $\pm$ 131
Cripe and Moore 1975	3	610	640	610/640	630 $\pm$ 17
Gibson and Andrawes 1978b	16	410	830	555/560	753 $\pm$ 147
Gibson and Moore 1973	20	474	794	645/652	642 $\pm$ 110
Gibson and Moore 1974	12	560	1300	822/850	915 $\pm$ 244
Kaplan and Petrowski 1971	2	620	714	—	667
Kaplan et al. 1970	4	640	770	680/770	715 $\pm$ 57
Kaplan et al. 1976	10	360	710	670/680	643 $\pm$ 99
Kerridge et al. 1975b	13	365	829	673	617 $\pm$ 150
Kerridge et al. 1975c	19	320	810	534	552 $\pm$ 155
Kerridge et al. 1978	18	594	811	763/765	752 $\pm$ 53
Moore et al. 1972	5	900	1000	930	940 $\pm$ 36
Moore et al. 1974	21	710	1330	950	950 $\pm$ 174
Morrison et al. 1970	1	530	530	530	530
Palme et al. 1978	1	740	740	740	740
Rees and Thode 1972	13	549	1149	745	765 $\pm$ 145
Rees and Thode 1974	9	500	1014	620	709 $\pm$ 188
Sakai et al. 1972	2	337	920	—	629
Wänke et al. 1975	3	500	585	510	532 $\pm$ 38
All Soils	188	212	1750	704/706	715 $\pm$ 216

obtained by three different methods. Thus, there is no reason to doubt the low value obtained for 74220, except that it is unconfirmed.

Finally, a comparison of the sulfur contents of soils, basalts and breccias also shows much less variability of only a factor of 2 between these materials. Unlike the solar-wind-derived volatiles, sulfur is more abundant in basalts than in either breccias or soils.

#### F. Fluorine and Chlorine

Fluorine and chlorine analyses of lunar samples have been done by the following methods: (1) neutron activation analysis for F and Cl (see, e.g., Jovanovic and Reed 1973, 1974); (2) halogen extraction by pyrohydrolysis followed by F analysis with a fluoride-sensitive electrode (see, e.g., Palme et al. 1978; Wänke et al. 1972); (3) neutron activation analysis for Cl (see, e.g., Wänke et al. 1972); and (4) spark source mass spectrometry for F and Cl (see, e.g., Bouchet et al. 1971; Morrison et al. 1971). Most of the analyses in the literature have been done by methods (1) to (3). The results for fluorine are given in Tables XV–XVII and the results for chlorine are given in Tables XVIII–XX.

**TABLE XIV**  
Sulfur Contents of Lunar Breccias and Basalts

Reference	No. of Analyses	Sulfur Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
Breccias					
Cripe and Moore 1975	8	230	2100	910	926 $\pm$ 496
Gibson and Moore 1973	1	844	844	844	844
Gibson and Moore 1974	9	720	1020	890	866 $\pm$ 98
Kaplan et al. 1970	4	1070	1120	1070/1120	1095 $\pm$ 25
Kaplan et al. 1976	2	660	870	—	765
Moore et al. 1972	8	580	1020	910/950	846 $\pm$ 163
Moore et al. 1974	4	380	1110	500/860	713 $\pm$ 290
Palme et al. 1978	4	420	2150	470/1000	1010 $\pm$ 696
Rees and Thode 1974	3	400	828	500	576 $\pm$ 224
Wänke et al. 1975	4	560	1100	660/750	768 $\pm$ 203
Wänke et al. 1976	1	236	236	236	236
Wänke et al. 1977	9	420	1040	610	668 $\pm$ 197
All Breccias	57	230	2150	810	818 $\pm$ 339
Basalts					
Cripe and Moore 1975	1	1970	1970	1970	1970
DesMarais 1978	7	642	1850	1400	1213 $\pm$ 479
Gibson and Moore 1974	6	1580	2770	2210	2105 $\pm$ 400
Gibson et al. 1975	15	600	2770	965	1274 $\pm$ 632
Gibson et al. 1976	32	1580	2770	1850/1865	1871 $\pm$ 238
Gibson et al. 1977	18	560	1520	1105/1110	1099 $\pm$ 262
Kaplan and Petrowski 1971	1	754	754	754	754
Moore et al. 1974	3	1860	3140	2040	2347 $\pm$ 566
Rees and Thode 1974	2	1223	1581	—	1402
Wänke et al. 1975	8	485	1620	1350/1370	1132 $\pm$ 442
Wänke et al. 1976	1	715	715	715	715
All Basalts	94	485	3140	1580	1513 $\pm$ 562

Fluorine contents of bulk lunar soils range from a low of  $14 \mu\text{g g}^{-1}$  in 67455 (Wänke et al. 1973) to a high of  $230 \mu\text{g g}^{-1}$  in 74241 (Jovanovic and Reed 1973). The mean F content given in Table XV has a large uncertainty which is 66% of the mean. However, the large scatter is probably not due to analytical errors. Because of the small number of analyses there are few multiple analyses of the same sample, but there are no obvious systematic differences involving the two most extensive data sets, those of the Mainz and Argonne groups. Considering only the data of Wänke and colleagues, the

**TABLE XV**  
Fluorine Contents of Bulk Lunar Soils

Mission	No. of Soils	Fluorine Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
Apollo 11	1	66	96	86	$83 \pm 18$
Apollo 12	2	60	63	60/61	$61 \pm 1$
Apollo 14	2	106	145	—	126
Apollo 15	2	45	59	—	52
Apollo 16	8	14	105	39/47	$48 \pm 28$
Apollo 17	10	17	230	58	$77 \pm 61$
All Soils	25	14	230	59/60	$68 \pm 45$

**TABLE XVI**  
Comparison of Fluorine Analyses of Bulk Lunar Soils

Reference	No. of Analyses	Fluorine Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
Bouchet et al. 1971	2	63	96	—	80
Jovanovic and Reed 1973	5	47	230	102	$109 \pm 65$
Jovanovic and Reed 1974	8	17	61	38/52	$35 \pm 24$
Morrison et al. 1970	1	66	66	66	66
Morrison et al. 1971	1	60	60	60	60
Wänke et al. 1972	7	45	145	61	$80 \pm 32$
Wänke et al. 1973	5	14	210	41	$64 \pm 62$
Wänke et al. 1974	4	31	71	58/59	$55 \pm 15$
Wänke et al. 1975	1	39	39	39	39
All Soils	34	14	230	59/60	$68 \pm 45$

mean F content is  $70 \pm 40 \mu\text{g g}^{-1}$  if the KREEP sample 14163 ( $145 \mu\text{g g}^{-1}$  F) is included and  $66 \pm 38 \mu\text{g g}^{-1}$  is 14163 is excluded. Considering only the data of Reed and colleagues, the mean F content is  $63 \pm 59 \mu\text{g g}^{-1}$ . Within the uncertainties the two calculated mean F contents are identical, although the results of Wänke and colleagues have a smaller uncertainty.

The chlorine contents of lunar soils range from a low of  $8 \mu\text{g g}^{-1}$  in 71501 (Jovanovic and Reed 1974) to a high of  $350 \mu\text{g g}^{-1}$  in 10084 (Morrison et al. 1970). Again, only a small number of Cl analyses have been done and apparently no Cl data are available for Apollo 15 soils. The formal  $1\sigma$  uncertainty on the mean Cl content of lunar soils is larger than the mean value itself. If the two largest Cl contents of  $350 \mu\text{g g}^{-1}$  (reported for 10084 by Morrison et al. [1970]) and  $280 \mu\text{g g}^{-1}$  (reported for 14163 by Wänke et al. [1972]) are excluded, then the mean Cl content becomes  $30 \pm 20 \mu\text{g g}^{-1}$ .

The calculated F/Cl mass ratio in lunar soils in which both elements have been analyzed is  $2.4 \pm 1.4$ . This can be contrasted with the F/Cl mass ratio of



**TABLE XVII**  
Fluorine Contents of Lunar Breccias and Basalts

Reference	No. of Analyses	Fluorine Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
Breccias					
Palme et al. 1978	1	19	19	19	19
Wänke et al. 1972	2	101	119	—	110
Wänke et al. 1974	6	14	90	31/40	44 $\pm$ 25
Wänke et al. 1975	2	15	36	—	26
Wänke et al. 1976	1	15	15	15	15
Wänke et al. 1977	5	31	94	59	64 $\pm$ 23
All Breccias	17	14	119	40	54 $\pm$ 32
Basalts					
Wänke et al. 1975	5	27	78	49	54 $\pm$ 19
Wänke et al. 1976	1	22	22	22	22
All Basalts	6	22	78	43/49	49 $\pm$ 21

**TABLE XVIII**  
Chlorine Contents of Bulk Lunar Soils

Mission	No. of Soils	Chlorine Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
Apollo 11	1	27	350	84	154 $\pm$ 191
Apollo 12	1	24	48	30	34 $\pm$ 14
Apollo 14	2	40	280	—	160
Apollo 15	0	—	—	—	—
Apollo 16	10	12	41	19/22	24 $\pm$ 11
Apollo 17	16	8	103	18	30 $\pm$ 24
LUNA 16	1	24	24	24	24
LUNA 20	1	26	26	26	26
All Soils	32	8	350	25	44 $\pm$ 67

$\sim 0.09$  in CI chondrites. The lunar ratio is larger because Cl is depleted in lunar soils relative to CI chondrites where its abundance is about  $700 \mu\text{g g}^{-1}$ . The F/Cl mass ratio in lunar breccias and basalts in which both elements have been analyzed is  $4.0 \pm 4.8$ . Again, the high value is due to a Cl depletion.

### G. Depth Profiles of Volatiles in the Lunar Regolith

To find the total inventories, we need only to combine the estimated volatile contents with estimates of the variations in volatile abundance with location and with depth within the regolith.

**TABLE XIX**  
Comparison of Chlorine Analyses of Bulk Lunar Soils

Reference	No. of Analyses	Chlorine Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
Bouchet et al. 1971	2	48	84	—	66
Jovanovic and Reed 1973	15	13	72	26	30 $\pm$ 16
Jovanovic and Reed 1974	9	8	103	16	31 $\pm$ 28
Morrison et al. 1970	1	350	350	350	350
Morrison et al. 1971	1	30	30	30	30
Wänke et al. 1970	1	27	27	27	27
Wänke et al. 1971	1	24	24	24	24
Wänke et al. 1972	2	40	280	—	160
Wänke et al. 1973	2	12	20	—	16
Wänke et al. 1974	4	12	25	15/16	17 $\pm$ 5
Wänke et al. 1975	1	15	15	15	15
All Soils	39	8	350	25	44 $\pm$ 67

**TABLE XX**  
Chlorine Contents of Lunar Breccias and Basalts

Reference	No. of Analyses	Chlorine Abundance $\mu\text{g g}^{-1}$			
		Min.	Max.	Median	Mean $\pm 1\sigma$
Breccias					
Jovanovic and Reed 1981	1	195	195	195	195
Palme et al. 1978	2	18	51	—	35
Wänke et al. 1974	6	12	270	26/104	115 $\pm$ 110
Wänke et al. 1975	2	10	33	—	21
Wänke et al. 1976	1	7	7	7	7
Wänke et al. 1977	5	4	30	20	20 $\pm$ 9
All Breccias	17	4	270	26	65 $\pm$ 86
Basalts					
Jovanovic and Reed 1980	2	1.4	5.9	—	3.7
Wänke et al. 1975	5	3.5	6	4.9	5 $\pm$ 0.9
Wänke et al. 1976	1	4	4	4	4
All Basalts	8	1.4	6	4.5/4.9	4.5 $\pm$ 1.5

The largest uncertainty in these calculations stems from our incomplete knowledge of the distribution of solar-wind-implanted volatiles with depth beneath the surface of the regolith. As the regolith is stirred ("gardened") by impacts, grains that had been on the surface are mixed to some depth. The solar-wind-rich layer could be as deep as the regolith itself, which, on average,

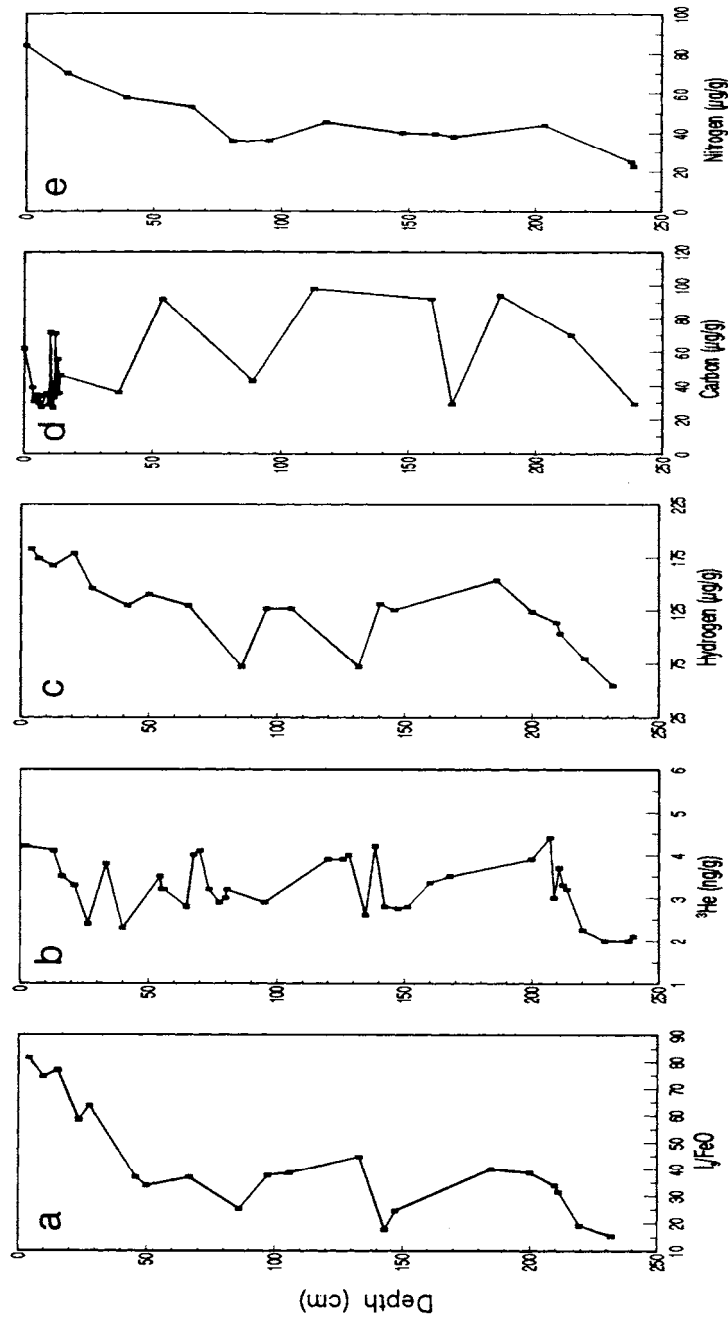


Figure 6. Profiles with depth for left:  $^{15}\text{O}/\text{FeO}$ ; (center left)  $^3\text{He}$ ; (center) hydrogen; (center right) carbon and (right) nitrogen in the Apollo 15 deep drill core (samples 15001 to 15006). The  $^{15}\text{O}/\text{FeO}$  and H data are from Gibson and Bustin (1987); the  $^3\text{He}$  data are from the compilation in Appendix A of Swindle et al. (1990); the C data are from DesMarais et al. (1973), Mozeleski et al. (1972), and Wszolek et al. (1972), and the N data are from Becker and Clayton (1977), Smith et al. (1973), and Wszolek et al. (1972). Note that the H data from Gibson and Bustin (1987) are for the sub- $10\ \mu\text{m}$  grain fraction.

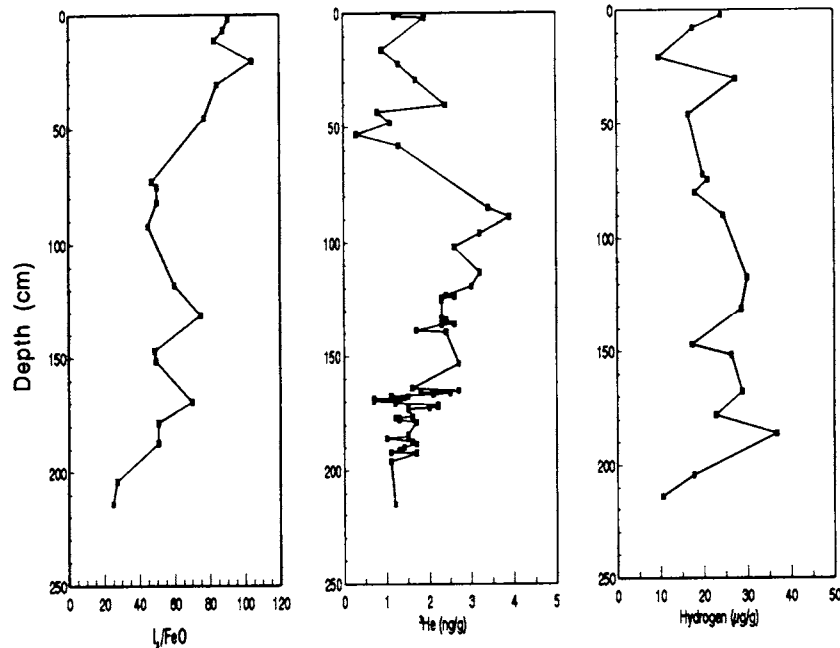


Figure 7. Profiles with depth for (left)  $I_s/FeO$ ; (center)  $^3He$ , and (right) hydrogen in the Apollo 16 deep drill core (samples 60001 to 60007). The  $I_s/FeO$  and H data are from Gibson et al. (1988) and the  $^3He$  data are from Appendix A of Swindle et al. (1990).

is about 8 m deep in the maria and 15 m deep in the highlands (McKay et al. 1991, and references therein). On the other hand, quantitative models of regolith evolution typically predict something resembling an exponential decrease in the abundance of surface-exposed grains with depth, falling to  $1/e$  of its surface value in a depth of about 3 m (see, e.g., Arnold 1975). Only three Apollo drill cores extend to greater than 1 m depth, and the deepest of these (Apollo 17) extends to only 3 m depth. The data from these drill cores (Figs. 6–8) are ambiguous, because the random variations (resulting from variations in maturity of layers of soil found at different depth) are large enough to mask the difference between a uniform abundance with depth and a substantial fall-off with depth. Most estimates of the total lunar inventory of volatiles assume a uniform concentration to a depth of 2 or 3 m and nothing deeper (see, e.g., Wittenberg et al. 1986; Haskin 1989; Taylor 1991), which tends to be closer to the exponential model considered above (i.e., a lower limit). For our estimates, we will assume a uniform concentration to 3 m depth and nothing deeper, but note that present evidence does not rule out the possibility that this accounts for only 20 to 30% of the total inventory.

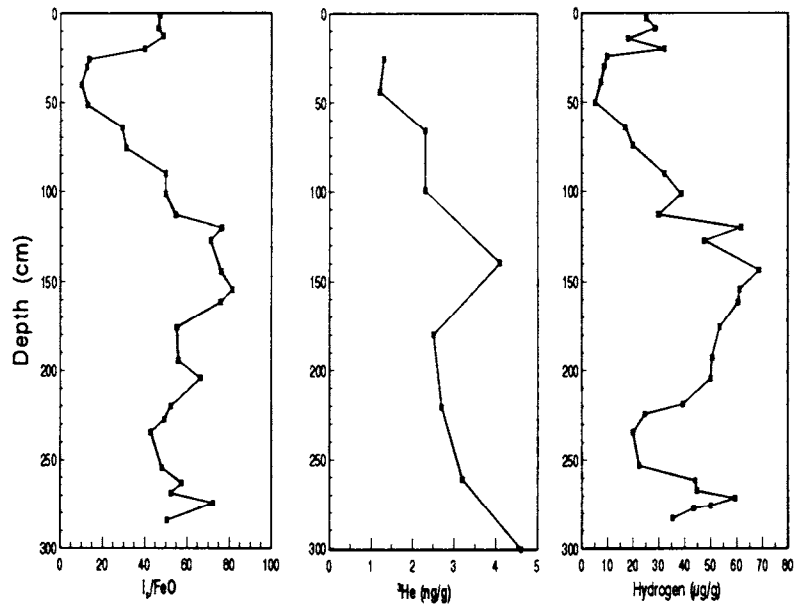


Figure 8. Profiles with depth for (a) left:  $I_2/FeO$ , (b) center:  $^3He$ , and (c) right: hydrogen in the Apollo 17 deep drill core (samples 70001 to 70009). The  $I_2/FeO$  and H data are from Gibson et al. (1988) and the  $^3He$  data are from Appendix A of Swindle et al. (1990).

## H. Geographic Effects

In our estimates, we also ignore the possibility of latitude- and longitude-dependent effects. However, if saturation does not occur until doses reach three or more times those received at the central nearside, the total inventories could be a factor of 2 larger than our estimates. (Our estimates could also be about 10% too high, if saturation occurs at a fluence very close to that seen by the Apollo samples.)

For He and Ne, we also must somehow account for the variation in Ti content. We will use the method of Swindle et al. (1990), who averaged the Ti contents in mare and highland areas overflowed by the Apollo orbiter gamma-ray spectrometers and assumed that those averages were typical of the Moon as a whole. They then converted these into  $^3He$  abundances by using a plot analogous to Fig. 5a. This approach emphasizes the equatorial regions, but there is no evidence of systematic compositional variations with latitude. We will then use the average weight ratios of  $^4He/^3He$  and  $Ne/^3He$  to calculate the abundances of those species.

## I. Global Inventory Estimates

Our estimates are given in Table XXI. Several points should be emphasized from a resource perspective. First, none of these elements should be con-

sidered abundant. Typical concentrations are  $125 \mu\text{g g}^{-1}$  or less, with the exception of sulfur. Second, the solar-wind volatiles come as a package; mining for one of them is mining for all of them. If mining of  $^3\text{He}$  occurs, for every ton of  $^3\text{He}$  extracted, more than  $10^3$  tons each of H, C, N and  $^4\text{He}$  will also be extracted. Because these are valuable resources in their own right, any  $^3\text{He}$  mining scheme should include processing of these gases as well. Third, these estimates are conservative; for reasons we have stated above, the actual inventories could be as much as an order of magnitude larger. Fourth, if we have underestimated the inventories, a substantial portion of those inventories must be at depths of  $>3$  m and/or on the farside; this has obvious implications for mining scenarios.

**TABLE XXI<sup>a</sup>**  
Globally Averaged Inventory of Volatiles on the Lunar Surface

Volatile	Concentration in Soil	Estimated Inventory ( $10^9$ kg)	
		This Work	Other Values
$^3\text{He}$	$4.2 \pm 3.4 \text{ ng g}^{-1}$	$0.84 \pm 0.69$	0.45–4.6 (Swindle et al. 1990); 1.1 (Wittenberg et al. 1989); 0.4–0.5 (Taylor 1991)
$^4\text{He}$	$14.0 \pm 11.3 \mu\text{g g}^{-1}$	$2890 \pm 2300$	
$^{20}\text{Ne}$	$1.2 \pm 0.8 \mu\text{g g}^{-1}$	$250 \pm 160$	
$^{36}\text{Ar}$	$0.50 \pm 0.19 \mu\text{g g}^{-1}$	$100 \pm 40$	
$^{84}\text{Kr}$	$0.54 \pm 0.22 \text{ ng g}^{-1}$	$0.10 \pm 0.04$	
$^{132}\text{Xe}$	$0.14 \pm 0.06 \text{ ng g}^{-1}$	$0.03 \pm 0.01$	
H	$46 \pm 16 \mu\text{g g}^{-1}$	$9,300 \pm 3,200$	8,000 (Haskin 1989)
C	$124 \pm 45 \mu\text{g g}^{-1}$	$25,200 \pm 9,100$	15,000 (Haskin 1989)
N	$81 \pm 37 \mu\text{g g}^{-1}$	$16,500 \pm 7,500$	8,000 (Haskin 1989)
S	$715 \pm 216 \mu\text{g g}^{-1}$	$145,000 \pm 44,000$	
F	$70 \pm 47 \mu\text{g g}^{-1}$	$14,200 \pm 9,500$	
Cl <sup>a</sup>	$44 \pm 67 \mu\text{g g}^{-1}$	$8,900 \pm 13,600$	

<sup>a</sup> As shown in Table XVIII, the formal  $1\sigma$  uncertainty on the Cl concentration is greater than the concentration itself. If the two highest values of  $350 \mu\text{g g}^{-1}$  (Morrison et al. 1970) and  $280 \mu\text{g g}^{-1}$  (Wänke et al. 1972) are excluded the mean  $\pm 1\sigma$  becomes  $30 \pm 20 \mu\text{g g}^{-1}$ . The corresponding inventory is then  $6100 \pm 4100$  in units of  $10^9$  kg.

There are two other possible uncertainties, both most serious for hydrogen, that should also be mentioned. First, some unknown (but perhaps large) fraction of the hydrogen measured in lunar soils may be terrestrial contamination, so the true regolith inventory could be less than that estimated by us and others (see, e.g., Haskin 1989). This possibility is suggested by the isotopic studies of  $\text{H}_2$  released from lunar samples (see, e.g., Epstein and Taylor 1970, and subsequent papers).

Second, as originally suggested by Watson et al. (1961), and later ex-

panded upon by Arnold (1976,1979,1987), it is possible that there are permanently shadowed regions near the lunar poles which remain cold enough to trap significant amounts of water ice. The estimated water inventory in the permanently shadowed regions is in the range of  $10^{16}$  to  $10^{17}$  g (Arnold 1979), or about 1 to 10 times as much hydrogen as contained as  $H_2$  in lunar soils. The potential sources of lunar water are the solar-wind reduction of  $Fe^{2+}$  to Fe metal in the lunar regolith, the impact of water-bearing meteorites, cometary impact, and water degassed from the lunar interior. If the water from these sources could have been efficiently transported to the permanently shadowed regions at the lunar poles (covering  $\sim 0.02\%$  of the lunar surface according to Hodges [1980]), then deposits with column densities of  $10^3$  g  $cm^{-2}$ , or greater, may have formed (Hodges 1991). However, a recent re-examination of the efficiency of the transport process by Hodges (1991) implies that transport is not efficient and that the amount of water ice in lunar polar deposits is probably too small to be of practical interest. Instead, Hodges (1991) proposes that the water is assimilated into the lunar regolith, which on average would give  $30 \mu moles g^{-1}$  of  $H_2O$  in lunar soils. This alternative requires efficient exchange of lunar water for terrestrial water in all lunar samples, none of which show evidence for any water other than terrestrial contamination (see Sec. V below). However, presumably this alternative can be tested with more careful analyses of existing lunar samples or by the collection of new samples under very carefully controlled conditions. Likewise, remote sensing observations from lunar polar orbit, or observations by robotic probes deployed on the lunar surface are apparently required to test unambiguously the concept of water ice deposits in the lunar polar regions.

#### IV. EXTRACTION OF VOLATILES FROM LUNAR SOILS

The extensive laboratory studies of volatiles in lunar samples suggest two obvious ways to extract solar-wind-derived volatiles from lunar soils: (1) heating bulk soils, and (2) grain-size sorting of bulk soils followed by volatile extraction from the finest size fractions. In the case of He and Ne, beneficiation by concentration of ilmenite could also be imagined. In this section, we examine these issues in detail to explore whether or not they may turn out to be practical. We begin by considering volatile release by heating bulk soils.

##### A. Thermal Extraction Schemes

Most studies of the resource potential of solar-wind gases assume that gas will be extracted by simple heating, although the exact mechanism, such as microwaves or concentrated solar energy, varies from study to study (see, e.g., Houdashelt et al. 1989). This technique clearly will work, because virtually all the solar-wind-derived gas is released by a temperature of about  $700^\circ C$  (see Figs. 9 and 10). However, whether or not the outcome is desirable for volatile extraction will depend to some extent on the chemical speciation of the volatiles released from the heated soil and how they can be separated from

one another. The physical properties of the evolved volatiles such as their condensation temperatures, thermal conductivities, viscosities, and so on may also have an influence on engineering designs for an extraction facility.

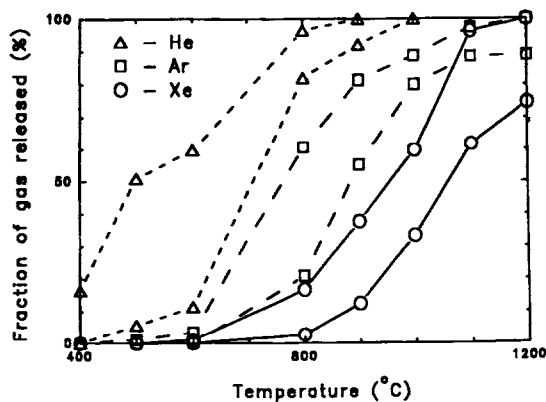


Figure 9. Release patterns for He, Ar and Xe in lunar soils heated to progressively higher temperatures (stepwise heating). Data are from Basford et al. (1973), Hohenberg et al. (1970), Pepin et al. (1970) and Srinivasan et al. (1972). Note the progressively higher release temperatures in going from He to Ar to Xe.

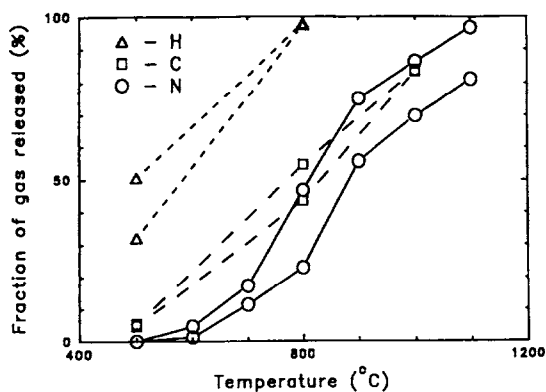


Figure 10. The release patterns for the chemically reactive volatiles H, C and N for lunar soils heated to progressively higher temperatures under vacuum. Data are from Becker (1980), Becker and Clayton (1977,1978), Norris et al. (1983) and Thiemens and Clayton (1980). Note the significantly higher temperatures required for quantitative release of C and N relative to hydrogen.

Fortunately, the analytical literature is a good guide to what to expect in this regard. Epstein and Taylor (1970) found that ~20 to 33% of the



total hydrogen in lunar soils was evolved as H<sub>2</sub>O. However, the observed D/H ratio and oxygen isotopic composition suggests that most, if not all, of the water released from lunar soils is probably terrestrial contamination (see, e.g., Epstein and Taylor 1970,1971,1972,1973,1974,1975; Merlivat et al. 1972,1974,1976; Stievenard et al. 1990). Thus, hydrogen will probably be evolved dominantly as H<sub>2</sub>.

The situation is a little more complex for carbon. Acid hydrolysis of lunar samples decomposes cohenite [ideally (Fe,Ni)<sub>3</sub>C], which is the major carbide present in lunar samples (see, e.g., Goldstein et al. 1976), and yields CH<sub>4</sub> along with lesser amounts of higher hydrocarbons (see, e.g., Cadogan et al. 1972; Chang et al. 1970). Pyrolysis and inorganic gas release studies yield a mixture of CO + CO<sub>2</sub> + CH<sub>4</sub> (see, e.g., Chang et al. 1974a; DesMarais et al. 1973; Gibson and Moore 1972). Studies of the gases released from lunar rocks and soils upon crushing show that CO > CO<sub>2</sub> ~ CH<sub>4</sub> in lunar basalts (Gibson and Andrawes 1978a).

Nitrogen is released as N<sub>2</sub>. Ernsberger (1972) reports that laser heating of individual soil particles yields approximately equal amounts of N<sub>2</sub> and CH<sub>4</sub>. Pyrolysis studies of lunar soils commonly yield N<sub>2</sub>, although NH<sub>3</sub> has also been reported (see, e.g., Simoneit et al. 1972). The NH<sub>3</sub>/N<sub>2</sub> ratio was much less than one, and the NH<sub>3</sub> may be a contaminant. Crushing experiments on lunar rocks and soils release N<sub>2</sub> only (Gibson and Andrawes 1978a). Inorganic gas release studies also show only N<sub>2</sub> (Gibson and Moore 1972).

Sulfur is released primarily as H<sub>2</sub>S and SO<sub>2</sub>. Hydrolysis of lunar soils commonly gives the former, while pyrolysis commonly gives the latter (see, e.g., Chang et al. 1974a, b; DesMarais 1978). The inorganic gas release study of Gibson and Moore (1972) reported both H<sub>2</sub>S and SO<sub>2</sub>.

Finally, apparently no studies have been done on the speciation of F and Cl released from lunar soils upon heating or upon the release profiles. These elements will readily combine with hydrogen to form HF and HCl upon heating. The mean H, S, F and Cl contents in lunar soils correspond to an atomic ratio H/(S + F + Cl) ~ 1.7, which implies that all F and Cl will be released as HF and HCl, respectively.

### **B. Grain-Size Sorting**

The hydrogen content of lunar soils as a function of grain size has been studied by DesMarais et al. (1974) and Gibson and collaborators (Bustin et al. 1984,1986; Gibson et al. 1987). Related studies of hydrogen distribution with depth in lunar soil grains have been done using F<sub>2</sub> etching (Becker 1980; Epstein and Taylor 1975) and nuclear techniques (Leich et al. 1973,1974). These studies generally indicate that hydrogen is indeed concentrated near grain surfaces and in finer grain-size fractions of lunar soils, but the interpretation of the results is not totally unambiguous (cf. Becker 1980).

DesMarais et al. (1974) studied the hydrogen content of grain-size fractions of five Apollo 15 and 16 soils. Not surprisingly they found more hydrogen in the finer grain-size fractions. DesMarais et al. (1974) resolved

both surface- and volume-correlated hydrogen components in their samples and claimed that the soils were not saturated with respect to hydrogen. Instead, in their view, the hydrogen content of lunar soils is controlled by a kinetic steady state in which the hydrogen implanted by the solar wind is balanced by hydrogen lost by diffusive escape and sputtering. This lack of saturation for hydrogen is similar to the lack of saturation which is observed for helium.

DesMarais et al. (1974) did not present data for the weights of the different size fractions and a complete mass balance relating the hydrogen contents of the different size fractions to the hydrogen content of the bulk sample is not possible for their samples. However, this was done by Gibson and colleagues (Bustin et al. 1984,1986; Gibson et al. 1987). Their data for the hydrogen content of different grain size fractions of five lunar soils are presented in Table XXII. The replicate analyses reported in their abstracts have been averaged together and the  $1\sigma$  uncertainties were calculated using small number statistics (Wilson 1952). The bulk soil hydrogen contents are taken from our compilation of literature data.

**TABLE XXII**  
Hydrogen Content of Lunar Soils as a Function of Grain Size<sup>a</sup>

Grain-Size Fraction ( $\mu\text{m}$ )	Hydrogen Abundance $\mu\text{g g}^{-1}$ ( $\pm 1\sigma$ )				
	10084	12070	15021	60501	71501
<20	133 $\pm$ 25	108 $\pm$ 0.5	148 $\pm$ 22	134 $\pm$ 18	154 $\pm$ 50
20-45	32 $\pm$ 4	25 $\pm$ 10	54 $\pm$ 6	46 $\pm$ 6	59 $\pm$ 21
45-75	29 $\pm$ 8	10 $\pm$ 11	25 $\pm$ 4	27 $\pm$ 20	39 $\pm$ 37
75-90	25 $\pm$ 8	7 $\pm$ 4	21 $\pm$ 2	14 $\pm$ 2	25 $\pm$ 31
90-150	18 $\pm$ 4	5 $\pm$ 6	18 $\pm$ 5	9 $\pm$ 0.5	6 $\pm$ 3
150-250	22 $\pm$ 18	5 $\pm$ 5	8 $\pm$ 5	4 $\pm$ 2	2 $\pm$ 0
250-500	17 $\pm$ 2	6 $\pm$ 6	13 $\pm$ 7	4 $\pm$ 0.4	11 $\pm$ 15
500-1000	5 $\pm$ 5	6 $\pm$ 5	9 $\pm$ 3	4 $\pm$ 3	3 $\pm$ 2
Weighted Sum	52 $\pm$ 7	32 $\pm$ 3	56 $\pm$ 5	47 $\pm$ 5	47 $\pm$ 11
Bulk Soil <sup>b</sup>	53 $\pm$ 11	33 $\pm$ 6	54 $\pm$ 12	34 $\pm$ 6	23 $\pm$ 5

<sup>a</sup> Mean  $\pm 1\sigma$  of values from Bustin et al. (1984,1986) and Gibson et al. (1987).

<sup>b</sup> Mean  $\pm 1\sigma$  of literature values for the hydrogen content of the bulk soil.

A comparison of the weighted sum of the hydrogen contents of the different grain-size fractions with the literature data on bulk soil hydrogen contents shows generally good agreement for soils 10084, 12070 and 15021. In these three cases the calculated and observed hydrogen contents are almost identical and always agree within the combined  $1\sigma$  uncertainties. However, this is not the case for soils 60501 and 71501 where the weighted sums are considerably larger than the observed bulk values. This could be explained by contamination from terrestrial water vapor on the finest grain size fractions. This would then lead to higher apparent hydrogen contents in these fractions and to a weighted sum larger than the bulk soil value. However, the hydrogen

contents of the  $<20 \mu\text{m}$  fractions of all five soils are similar and in any case it is not clear why only two of the five samples would be affected by terrestrial water vapor.

Figure 11 presents the same data in graphical form. In this case, the percentage of the total soil mass in each grain-size fraction is compared to the percentage of total hydrogen in each grain-size fraction. If we consider only the three samples which display mass balance, 61 to 74% of the total hydrogen is in the sub  $20 \mu\text{m}$  grains, and 78 to 87% of the total hydrogen is in the sub  $45 \mu\text{m}$  grains. However, 22 to 26% of the total mass resides in the sub  $20 \mu\text{m}$  grains and 40 to 46% of the total mass is in the sub  $45 \mu\text{m}$  grains. The corresponding hydrogen enrichment factors for the sub  $45 \mu\text{m}$  fractions of the three soils are 1.8 (10084), 2.2 (12070), and 1.8 (15021). In other words, the hydrogen enrichment in the fine-grained fractions is only about a factor of 2. Given this small enrichment, the economics of grain-size beneficiation may not be viable.

Likewise, Fig. 12 illustrates similar data for  $^4\text{He}$ . These data, which are from R. O. Pepin (personal communication, 1991; Pepin et al. 1975) show similar trends for the He enrichment factor. Specifically, for the sub  $37 \mu\text{m}$  fractions, the He enrichments are 2.1 (10084), 2.0 (12033), 2.9 (15531), 2.9 (67701) and 2.3 (70181). More results for He are shown in Table XXIII, where we summarize He results from three laboratories that analyzed both bulk samples and samples with grain size less than about  $20 \mu\text{m}$  from the same soil for five or more soils. Again, the results are consistent with average enhancement factors of about 2. As the  $<20 \mu\text{m}$  fraction typically contains about 20% of the mass of the soil (Butler et al. 1973,1974), this means that such a grain-size separation would lose more than half the He. Whether this would be useful would depend on the detailed economics of the mining, sorting and extraction systems.

**TABLE XXIII**  
He Enrichment in Fine-Grained Soil Particles

Laboratory	$\text{He}_{<20\mu\text{m}}/\text{He}_{\text{bulk}}$	No. of Samples	Notes <sup>a</sup>
Bogard	$1.84 \pm 0.51$	10	1
Hintenberger	$1.56 \pm 0.45$	11	2
Kirsten	$2.39 \pm 0.52$	5	3

<sup>a</sup> Notes: (1) Data from Bogard et al. (1973,1974) and Bogard and Hirsch (1975). (2) Data from Hintenberger et al. (1970,1971,1974,1975) and Hintenberger and Weber (1973). One sample  $<30 \mu\text{m}$  and one sample  $<25 \mu\text{m}$  are included. (3) Data from Kirsten et al. (1972,1973).

Finally we note that because the solar-wind-derived volatiles are located near the surfaces of grains, a technique that released the gas in the outer  $1 \mu\text{m}$  of every grain would extract virtually all of the gas (a small amount of gas does migrate to the interior of grains). In the laboratory, this is often done

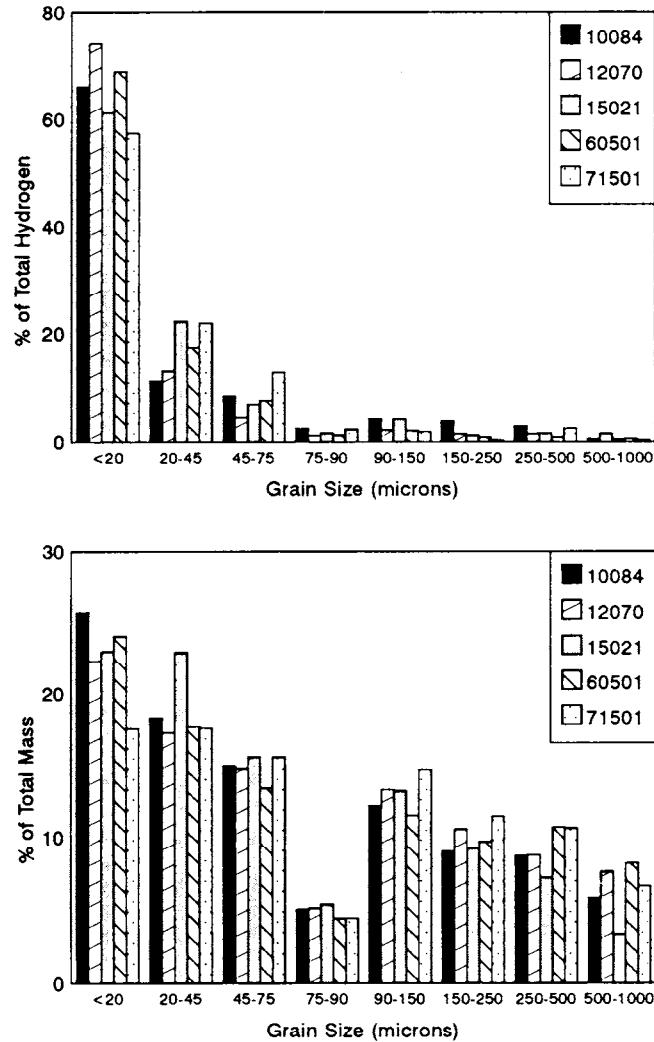


Figure 11. (top) The percentage (by mass) of total hydrogen found in different grain-size fractions of lunar soils from the five Apollo landing sites. (bottom) The percentage of the total mass in each grain-size fraction of the same soils. Note that the smallest grain-size fraction in this figure approximately corresponds to the sum of the two smallest size fractions in Fig. 12. The hydrogen content and mass as a function of grain size are from Bustin et al. (1984,1986) and Gibson et al. (1987).

by etching with mineral acids or  $F_2$ . While that might not be practical on the Moon, it might be possible to achieve similar results through the mutual abrasion of agitated particles. Such a scheme has not been investigated in detail.

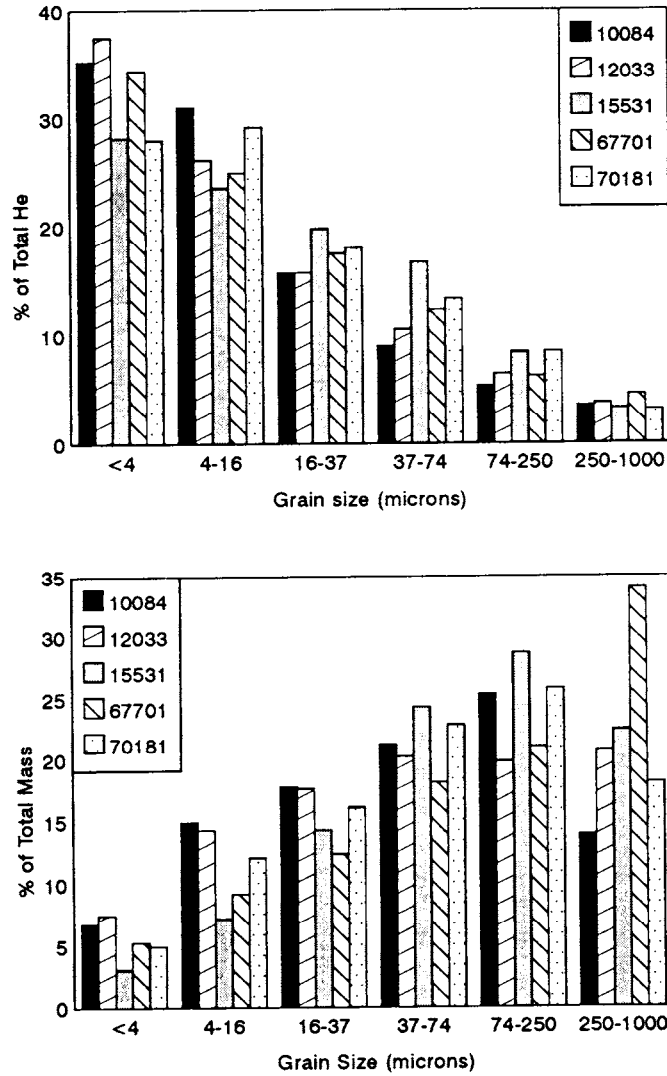


Figure 12. (top) The percentage (by mass) of total <sup>4</sup>He in different grain-size fractions of lunar soils from the five Apollo landing sites. (bottom) The percentage of the total mass in each grain-size fraction of the same soils. Note that the two smallest size fractions in this figure approximately correspond to the smallest size fraction in Fig. 11. The <sup>4</sup>He content and mass as a function of grain size are from R. O. Pepin (personal communication, 1991) and Pepin et al. (1975).

### C. Ilmenite Separation

Beneficiation by ilmenite separation seems plausible for He extraction, because He is much more abundant (sometimes by more than a factor of 100) in ilmenite than in other lunar materials. However, separation of ilmenite from soil is apparently extremely difficult (see, e.g., Heiken and Vaniman 1990), and even if it could be achieved, such a separation would miss the common He- and Ti-rich compound particles, such as agglutinates and breccias (whose high He and Ti contents presumably reflect ilmenite grains that became a part of the compound particles).

## V. LUNAR WATER

One of the first, and apparently one of the most firm, conclusions drawn from examination of lunar samples is their anhydrous nature. The virtually unanimous agreement upon this point is illustrated by the following list taken from Charles et al. (1971): (1) searches for hydrous material in lunar rocks have been singularly unsuccessful, with the few exceptions to the contrary being discussed in Sec. V(B) below; (2) spectral studies by Adams and McCord (1970), which are cited in Charles et al. (1971), reveal that lunar materials completely lack bands due to  $\text{OH}^-$  and  $\text{H}_2\text{O}$ ; (3) electron microprobe analyses of lunar apatites [ideally  $\text{Ca}_5(\text{PO}_4)_3(\text{F}, \text{Cl}, \text{OH})$ ] show F and Cl in amounts high enough to indicate that OH is either absent or minor (also see Frondel 1975, and references therein); (4) major element analyses of the Apollo 11 samples revealed no detectable  $\text{H}_2\text{O}$  (see, e.g., Engel and Engel 1970); (5) the high  $\text{Fe}^{2+}/\text{Fe}^{3+}$  ratios indicate the lack of a suitable oxidizing agent such as  $\text{H}_2\text{O}$ ; (6) no aqueous fluid inclusions were found in the Apollo 11 samples (Roedder and Weiblen 1970); and (7) the isotopic composition of the trace amounts of water found in lunar samples are consistent with contamination by terrestrial water vapor (Epstein and Taylor 1970).

In Sec. V.A we briefly review some of this evidence in the context of lunar resource utilization. We begin by discussing the isotopic evidence showing that water in lunar samples may simply be due to terrestrial water vapor contamination and then describe the small amount of mineralogical evidence which implies the presence of water in the lunar interior at some point in the past.

### A. Isotopic Evidence

The  $\text{H}_2$  and  $\text{H}_2\text{O}$  contents and isotopic compositions of lunar samples have been studied by three groups: (1) Epstein and Taylor (1970, 1971, 1972, 1973, 1974, 1975); (2) Friedman and colleagues (Friedman et al. 1970, 1971, 1972); and (3) by French workers (Merlivat et al. 1972, 1974, 1976; Stievenard et al. 1990). Their data for  $\text{H}_2$  contents are presented in Tables III–V and their data for water contents and isotopic composition are summarized in Table XXIV.

The highest water contents, about  $14 \mu\text{moles g}^{-1}$  are found in lunar breccias and soils, while lower contents of about  $2 \mu\text{moles g}^{-1}$  are found in

lunar basalts. The mean contents of all three groups overlap due to the large uncertainties on the means for breccias and soils, but it is clear that basalts contain the least water. However, the isotopic results suggest that even this small amount of water is dominated by terrestrial contamination.

The observed D/H ratios in the samples analyzed span a wide range from 21 ppm in breccia 10060 ( $\delta D = -865$  per mil) to 150 ppm ( $\delta D = -37$  per mil) in basalt 15555. However, the mean D/H values observed in the basalts, breccias, and soils only range from 85 to 136 ppm and all three means overlap within the  $1\sigma$  uncertainties (see Table XXIV).

As stressed by Epstein and Taylor in their papers, the isotopic composition of water extracted from lunar samples is generally indistinguishable from that of terrestrial water. This point is vividly illustrated in Fig. 13. This figure shows that the D/H ratio, and where determined the  $^{16}\text{O}/^{18}\text{O}$  ratio, in lunar samples is the same as that of atmospheric water vapor in Pasadena, Calif. Given this similarity, both the Caltech group and the French workers have concluded that there is no unambiguous evidence that the water extracted from lunar samples originated on the Moon instead of being merely terrestrial contamination. (But see the papers by Friedman and colleagues for an opposing viewpoint.)

However, as noted recently by Hodges (1991), assuming that the water produced by the solar-wind reduction of  $\text{Fe}^{2+}$  to Fe metal is retained by the Moon, then the mean water content of lunar soils should be about  $15 \mu\text{moles g}^{-1}$ , or about the same as that observed for lunar soils and breccias. If the probable water input due to cometary and meteoritic impacts is also considered, then this figure becomes about  $30 \mu\text{moles g}^{-1}$ . Thus, the observed water contents are not inconsistent with independent theoretical predictions.

## B. Mineralogical Evidence for Water

The mineralogical evidence for water falls into two categories: (1) amphiboles and micas found in lunar samples; and (2) the presence of  $\beta$ -FeOOH, the mineral akaganéite, in the rusty rock 66095 and various other lunar samples. We consider the occurrences of amphiboles and micas first and rely on Frondel (1975) who summarizes the relevant observations.

*Amphiboles and Micas.* Gay et al. (1970) found an amphibole in a vug in a sample of 10058, a medium-grained basalt. The electron microprobe analysis corresponds to a low-alumina Na-amphibole known as magnesioarfvedsonite with the general formula  $(\text{Na},\text{K},\text{Ca})_3 (\text{Mg},\text{Mn},\text{Fe})_3 ([\text{OH}]?,\text{F})_2$ . The amphibole contains  $1.2 \pm 0.3$  mass % F and gave a low total of 97.1%. The X-ray diffraction data for the crystal are also consistent with the identification as magnesioarfvedsonite. Charles et al. (1971) found loose grains of an amphibole in a package with a fragment of the porphyritic basalt 12021. The electron microprobe analysis shows low amounts of F (0.4%) and Cl (0.2%) and a low total of 97.5%. The microprobe analysis is consistent with an aluminoschermakite which has the general formula  $(\text{Ca},\text{Na},\text{K})_{2.5} (\text{Mg},\text{Fe},\text{Mn})_4$

**TABLE XXIV**  
Water Content and Isotopic Composition in Lunar Samples

Sample Number	Sample Type	Water $\mu\text{moles g}^{-1}$	D/H ppm	$\delta D^a$ (per mil)	Reference
12051	Basalt	0.45	142	-88	Friedman et al. 1971
15555	Basalt	0.49	150	-37	Friedman et al. 1972
70215	Basalt	2.7	130	-165	Merlivat et al. 1974
70215 <sup>b</sup>	Basalt	3.9 $\pm$ 2.7	135 $\pm$ 9	-133 $\pm$ 58	Merlivat et al. 1976
70215 <sup>c</sup>	Basalt	3.3	140	$\sim$ -100	Stievenard et al. 1990
75035	Basalt	2.48	141.4	-92	Merlivat et al. 1974
75035	Basalt	2.8	116.5	-252	Merlivat et al. 1974
Mean		2.3 $\pm$ 1.3	136 $\pm$ 11	-127 $\pm$ 71	
10046	Breccia	25.3	38	-756	Friedman et al. 1970
10046	Breccia	20.6	66	-576	Friedman et al. 1970
10060	Breccia	8.4	21	-865	Friedman et al. 1970
10061	Breccia	8.9	92	-409	Epstein and Taylor 1970
14305	Breccia	8.8	144	-76	Friedman et al. 1972
15299	Breccia	12.7	147.7	-52	Merlivat et al. 1974
Mean		14.1 $\pm$ 7.2	85 $\pm$ 53	-454 $\pm$ 340	
10084	Soil	11.07	116	-255	Epstein and Taylor 1970
10084	Soil	11.58	114	-268	Epstein and Taylor 1970
12030	Soil	12.4	123	-210	Friedman et al. 1971
12033	Soil	7.5	142	-88	Epstein and Taylor 1971
12042	Soil	13.5	134	-140	Epstein and Taylor 1971
12070	Soil	11.8	120	-230	Epstein and Taylor 1971



14240	Soil	17.4	121	-223	Epstein and Taylor 1972
14422	Soil	10.4	104	-332	Epstein and Taylor 1972
15001	Soil	25.1	132	-152	Epstein and Taylor 1975
15004	Soil	25.1	126	-191	Epstein and Taylor 1975
15006	Soil	30.9	125	-198	Epstein and Taylor 1975
15021 <sup>d</sup>	Soil	15.1	127	-185	Epstein and Taylor 1973
15100	Soil	23.7	44.6	-714	Friedman et al. 1972
15301	Soil	7.8	121	-223	Epstein and Taylor 1972
15600	Soil	2.6	146.5	-59	Merlivat et al. 1974
15600	Soil	3.8	140.9	-95	Merlivat et al. 1974
62221 <sup>d</sup>	Soil	18.5	130	-165	Epstein and Taylor 1973
64421	Soil	15.6	134	-140	Epstein and Taylor 1973
65513 <sup>e</sup>	Soil	~20	—	—	Epstein and Taylor 1974
66095	Soil	40.0	142	-88	Merlivat et al. 1974
68501	Soil	5.2	137.9	-115	Merlivat et al. 1974
72501	Soil	5.0	142	-88	Merlivat et al. 1974
72501	Soil	4.6	143	-82	Merlivat et al. 1974
74220	Soil	4.6	139	-108	Epstein and Taylor 1973
74240	Soil	11.0	117	-248	Epstein and Taylor 1975
78501	Soil	5.8	144.5	-72	Merlivat et al. 1974
Mean		13.8±9.2	127±20	-185±129	

<sup>a</sup> The  $\delta D$  values were all recalculated using  $D/H = 155.76$  ppm for Standard Mean Ocean Water (SMOW) as done by Merlivat et al. (1974, 1976). <sup>b</sup> Mean of eight values determined for 70215 (Merlivat et al. 1976). <sup>c</sup> Calculated from data of Stievenard et al. (1990) by mass balance. <sup>d</sup> Approximate values for  $D/H$  and  $\delta D$  because Epstein and Taylor (1973) combined the high temperature fractions of  $H_2$  and  $H_2O$  for 15021 and 62221. <sup>e</sup> The high temperature  $H_2O$  fraction for 65513 was lost by Epstein and Taylor (1973) and no  $\delta D$  value was given.

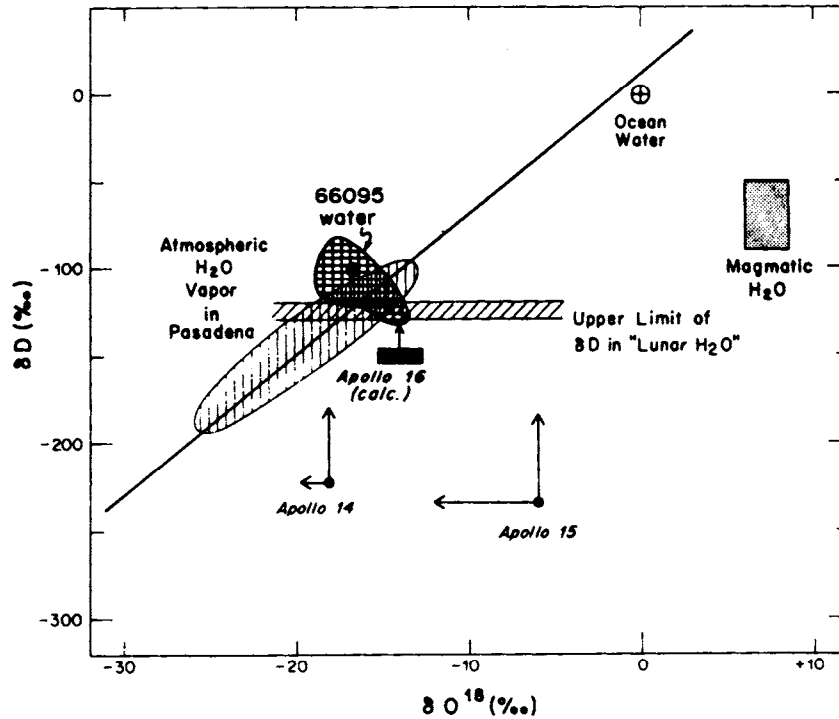


Figure 13. A comparison of the hydrogen and oxygen isotopic composition of water released from lunar soils with the H and O isotopic composition of terrestrial water. The Apollo samples plotted are 14321, 15301, 64221 and 66095. The O and H isotopic composition of these samples was determined by Epstein and Taylor (1973,1974). The cross-hatched horizontal band is their estimate for the upper limit of D/H in "lunar" water. The diagonal line is the locus of terrestrial meteoric waters. The arrows show the directions in which the isotopic data should be shifted to account for O and H isotopic exchange during sample processing. The isotopic similarity of "lunar" water to terrestrial water suggests that all water recovered from lunar samples is a terrestrial contaminant (figure taken from Epstein and Taylor 1974).



Mason et al. (1972) reported a few grains of hornblende in a thin section of breccia 14319,13 and in fines from 14163. The electron microprobe analysis corresponds to a titanian pargasite or a kaersutite with the general formula  $(\text{Ca}_{1.88} \text{Na}_{0.79} \text{K}_{0.35}) (\text{Mg}_{1.84} \text{Mn}_{0.05} \text{Fe}_{2.47} \text{Al}_{0.18} \text{Ti}_{0.45}) \text{Al}_2 \text{Si}_6 \text{O}_{23}$  and the total is 100.5%. In this case, the microprobe total does not show any evidence for a mass deficit which could be attributed to OH or water, but hornblendes are water-bearing minerals (see, e.g., Deer et al. 1963).

Gay et al. (1970) also reported a few small crystals of mica in fines from 10084. The electron microprobe analysis corresponds to that of a biotite

[general formula  $K_2(OH,F)_4(Mg,Fe,Al)_8(Si,Al)_8O_{20}$ ] and has a low total of 94.9%. The X-ray diffraction pattern is also consistent with a mica. However, as Frondel (1975) notes, the possibility that this material is a contaminant cannot be ruled out.

Drever et al. (1971) also reported flakes of an unknown mica in fines from 10084. In this case, only a qualitative probe analysis was possible because of the small size of the flakes. The X-ray diffraction pattern is consistent with various dioctahedral phyllosilicates such as muscovite or kaolinite. Drever et al. (1971) also reported rare  $\mu\text{m}$ -sized flakes of a mica on the surface of 10017, a fine-grained basalt. The  $b$  parameter in an X-ray pattern was consistent with biotite, chlorite, or a serpentine.

Although the identification of amphiboles in trace amounts seems firm, the inference that these minerals contain OH rests solely on mass deficits in microprobe analyses. Furthermore, the presence of trace amounts of micas is less firm and may be due to contamination. Thus, the available evidence by itself is not a compelling argument for water in the lunar interior.

*"Rust" on Lunar Rocks.* Mineralogical examination of Apollo 14 samples showed the presence of rust-like alteration on samples from every station and on breccia 66095 in particular (see, e.g., El Goresy et al. 1973; Taylor et al. 1973). This alteration product was originally believed to be goethite  $\alpha\text{-FeOOH}$  (see, e.g., El Goresy et al. 1973), but was later shown to be akaganéite  $\beta\text{-FeOOH}$  by Taylor et al. (1974). This phase was interpreted by some as being lunar in origin (see, e.g., El Goresy et al. 1973), but by others as being merely a reaction product between lawrencite  $\text{FeCl}_2$  and terrestrial water vapor, either in the Apollo module or on the Earth (see, e.g., Taylor et al. 1974).

The evidence in favor of an origin by reaction with terrestrial water vapor, either in the spacecraft, or upon return to Earth, is as follows. Taylor et al. (1974) showed that lawrencite, the probable precursor, is rapidly oxidized and hydrated to form akaganéite at  $25^\circ\text{C}$  and 40% relative humidity. Epstein and Taylor (1974) showed that the O and H isotopic composition of the akaganéite in breccia 66095 is identical to that of atmospheric water vapor (e.g., in Pasadena, Calif.) and that the amounts of water released and the isotopic composition of the water released from a terrestrial goethite and a synthetic akaganéite are (with the exception of the  $\delta^{18}\text{O}$  values in the synthetic sample) also virtually identical to that of the lunar sample. Taken together, these two studies considerably weaken the case for a lunar origin of the akaganéite in 66095 and strongly suggest a terrestrial origin.

## VI. LUNAR VOLCANIC GASES

Evidence for lunar volcanic gases comes from the presence of vesicles and vugs in lunar rock samples (e.g., LSPET 1969; Taylor et al. 1991) and from the volatile-rich surface coatings on the Apollo 15 green and Apollo 17 orange glasses (see, e.g., Meyer et al. 1975). Detailed examination of these surface coatings (see, e.g., Butler and Meyer 1976; Chou et al. 1975; Goldberg et

al. 1976; Wasson et al. 1976) shows that the coatings are probably condensates from a volcanic gas. The chemistry of this gas phase is of interest for several reasons: (1) to place constraints on the water abundance in the lunar interior at the time that volcanism occurred; (2) to identify the major gaseous species of volatile and ore-forming metals; and (3) to model the transport mechanisms which may have fractionated and concentrated metals into lunar "ore deposits." Fegley (1991,1992) used mass balance arguments and thermodynamic calculations to constrain the chemistry of lunar volcanic gases. He modeled lunar volcanic gases as modifications of terrestrial volcanic gases with two important differences. First, lunar volcanic gases were probably more reducing than terrestrial volcanic gases. Oxygen fugacity measurements on lunar samples (see, e.g., Sato 1976,1979; Sato et al. 1973) show that lunar rocks are generally about 4 orders of magnitude more reduced than terrestrial volcanic rocks and lie at or below the iron-wüstite (IW) buffer (e.g., see Fig. 14). Previous theoretical models of lunar volcanic gases have also assumed that they are more reducing than their terrestrial counterparts (see, e.g., Naughton et al. 1972; Wellman 1970). Second, the absence of indigenous water (and thus hydrogen) on the Moon means that lunar volcanic gases will be much drier than terrestrial volcanic gases which are generally rich in steam. As reviewed above in Secs. III.B and V, there is no firm evidence for indigenous water in lunar samples and the amounts of H<sub>2</sub> in lunar basalts is  $\leq 0.5 \mu\text{moles g}^{-1}$ . In the case of the orange soil 74220, the H<sub>2</sub> contents reported by Epstein and Taylor (1973) and Chang et al. (1974b) are 0.1 to 0.3  $\mu\text{moles g}^{-1}$ . Epstein and Taylor (1973) also found 4.6  $\mu\text{moles g}^{-1}$  H<sub>2</sub>O with a  $\delta D = -117$  per mil in 74220, but the isotopic composition is consistent with that of atmospheric water vapor in Pasadena, Calif. It is therefore most plausibly explained by contamination.

Fegley (1991,1992) noted that these two factors will lead to first-order differences between the chemistry of lunar and terrestrial volcanic gases. He also pointed out that changes in the ratios of sulfur and the halogens, although also important, will be less significant for the overall chemistry of the gas.

A representative model composition studied by Fegley (1991,1992) is displayed in Table XXV. This was derived by starting with a measured composition for a terrestrial volcanic gas from Hawaii and removing water (i.e., removing H and the corresponding stoichiometric amount of O needed for water). The oxidation state was also adjusted by setting the O/C atomic ratio to 1.2. The Br and I abundances were specified by assuming CI chondritic Cl/Br and Cl/I ratios. Ideal gas chemical equilibrium calculations were then done from 500 to 2000 K at pressures from 0.01 to 100 bar. This pressure range was chosen because it is appropriate for volcanic gases originating at different depths within the Moon (see, e.g., Wellman 1970). The thermodynamic data used in the calculations were taken from standard compilations such as the JANAF Tables (Chase et al. 1985) and Gurvich et al. (1989).

In addition to considering the chemistry of the elements listed in Table XXV, the major rock-forming elements (Ca, Mg, Si, Al, Ti, Fe), volatile

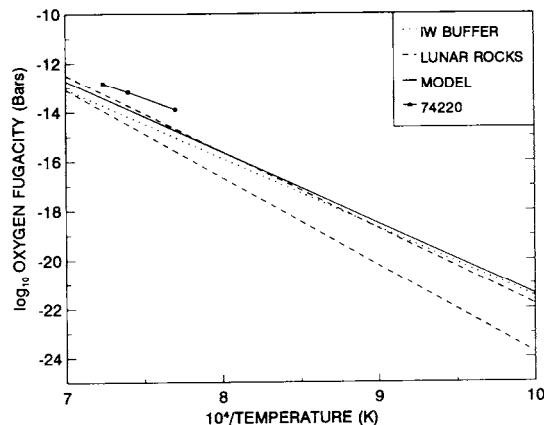


Figure 14. The measured oxygen fugacities as a function of temperature for lunar samples are compared with the iron-wüstite (IW) buffer and with the oxygen fugacities calculated for the lunar volcanic gas model of Fegley (1992). The oxygen fugacity of the orange soil 74220 is plotted separately because it is higher (e.g., more oxidizing) than most lunar samples (figure modified from Fegley 1992).

elements (P, Na, K, Zn, Cu), and ore-forming metals (Ni, Cr, Mn, Co, V) were also included in the calculations. Although these elements are of interest, these abundances in lunar volcanic gases are not at all well constrained. However, studies of terrestrial volcanic gases and sublimates from these gases (see, e.g., Symonds et al. 1987; White and Waring 1963) show that elements other than those listed in Table XXV generally have negligible abundances relative to those of the major constituents of terrestrial volcanic gases. Furthermore, as noted by Fegley (1991), theoretical models of the vaporization of the rock-forming elements from silicate magmas show negligible vapor pressures for compounds formed by these elements (Fegley and Cameron 1987). The more volatile elements (P, Na, K, Zn, Cu) will have higher vapor pressures, but even so the abundances of their gases over silicate magmas are expected to be less than those of gases formed by S, F, Cl and C. Therefore, the chemistry of the additional elements considered was calculated by giving them low abundances, for example, mole fractions of  $10^{-9}$  or less. As a result, the chemistry of these elements can be calculated without affecting the overall mass balance of the system.

Some of the results of the calculations are displayed in Figs. 14–18. Figure 14 shows that the calculated oxygen fugacity ( $f_{O_2}$ ) of the model lunar volcanic gas is basically the same as that as IW and is a good match to the measured  $f_{O_2}$  values for lunar samples. Figures 15–17 show the major Cl, F and S gases as a function of temperature at a total pressure of 1 bar. In contrast to terrestrial volcanic gases where steam dominates, the calculated  $H_2O$  abundance is totally insignificant and is not graphed because its abundance is below 0.1 ppb by volume. The low abundance of water vapor is a result of

**TABLE XXV**  
Lunar Volcanic Gas Model

Element	Abundance (%)
H	0.1
O	46.0
C	38.4
Cl	8.5
S	5.1
F	1.7
N	0.2
Br	190 ppm
I	14 ppm

removing H from the system. However, an important consequence of the low H elemental abundance is that the most abundant H-bearing gases in this case, and in other plausible models of lunar volcanic gases, are HCl and HF. As shown in Fig. 17, H<sub>2</sub>S is also totally insignificant and is not graphed because its abundance is also below 0.1 ppb by volume.

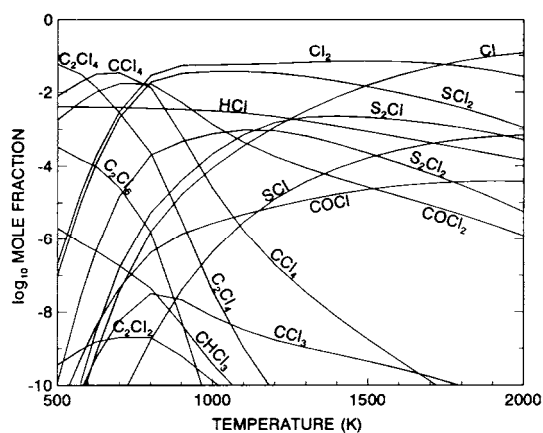


Figure 15. The calculated abundances of the major Cl-bearing compounds in the lunar volcanic gas model of Fegley (1992) are plotted as a function of temperature for a total pressure of 1 bar. Note that the chlorofluorocarbon (CFC) gases constitute a significant fraction of total Cl compounds. Also note that several other gases such as Cl<sub>2</sub>, CCl<sub>4</sub> and C<sub>2</sub>Cl<sub>4</sub> are generally more abundant than HCl over the entire temperature range considered (figure modified from Fegley [1992] with the inclusion of more Cl-bearing gases in the thermodynamic database).

Although HCl and HF are the two major H-bearing gases they are not the major Cl- and F-bearing gases. This is because the H/Cl and H/F atomic ratios of ~0.01 and 0.06, respectively, are too low to bind chemically all Cl and F in the form of hydrogen halides. Instead, Cl and F form other gases

such as  $\text{Cl}_2$ ,  $\text{CCl}_4$ ,  $\text{C}_2\text{Cl}_4$ ,  $\text{COF}_2$ ,  $\text{COCIF}$ ,  $\text{CF}_4$  and the chlorofluorocarbons  $\text{CClF}_3$ ,  $\text{CCl}_2\text{F}_2$ , and  $\text{CCl}_3\text{F}$ . The results in Fig. 15 show that the major Cl gases are Cl and  $\text{Cl}_2$  (at  $T \geq 800$  K) and  $\text{CCl}_4$  and  $\text{C}_2\text{Cl}_4$  (at  $T < 800$  K). The results in Fig. 16 show that the major F gases are  $\text{COF}_2$  (above 900 K), and  $\text{COCIF}$  and  $\text{CF}_4$  at lower temperatures. However, the chlorofluorocarbon gases together make up about 10% of the total Cl abundance and several tens of % of the total fluorine abundance. This is a dramatic change from terrestrial volcanic gases where calculations show that chlorofluorocarbons typically have volume mixing ratios in the range of  $10^{-28}$  to  $10^{-42}$  over the same temperature range (500 to 2000 K).

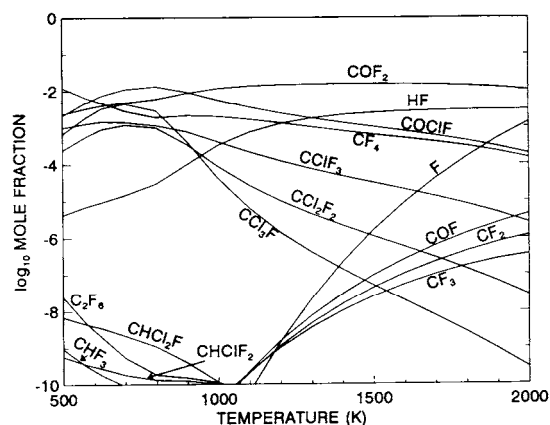


Figure 16. The calculated abundances of the major F-bearing compounds in the lunar volcanic gas model of Fegley (1992) are plotted as a function of temperature for a total pressure of 1 bar. Note that the chlorofluorocarbon (CFC) gases constitute a significant fraction of total F compounds. Also note that several other gases such as  $\text{COF}_2$ ,  $\text{COCIF}$ ,  $\text{CF}_4$  and  $\text{CCl}_3\text{F}$  are generally more abundant than HF over the entire temperature range considered (figure modified from Fegley [1992] with the inclusion of more F-bearing gases in the thermodynamic database).

The qualitatively different halogen chemistry predicted for lunar volcanic gases can be understood by considering how halogen chemistry depends on the abundances of H, Cl and F. Volcanic gases with the bulk abundance ratio  $\text{H}/(\text{F} + \text{Cl}) > 1$  will have HF and HCl as the dominant halogen gases, as in terrestrial volcanic gases. However, volcanic gases with  $\text{H}/(\text{F} + \text{Cl}) < 1$  will have HF and HCl as the dominant H gases, but not as the dominant halogen gases. In the intermediate case, volcanic gases with  $\text{H}/(\text{F} + \text{Cl}) = 1$  will display more complex chemistry involving HF, HCl and other halogen gases.

These considerations are of interest for lunar resources because the predicted large abundances of the chlorofluorocarbons are a direct consequence of the anhydrous and reducing conditions assumed to prevail in the lunar interior. The chlorofluorocarbons can thus be viewed as probes of conditions

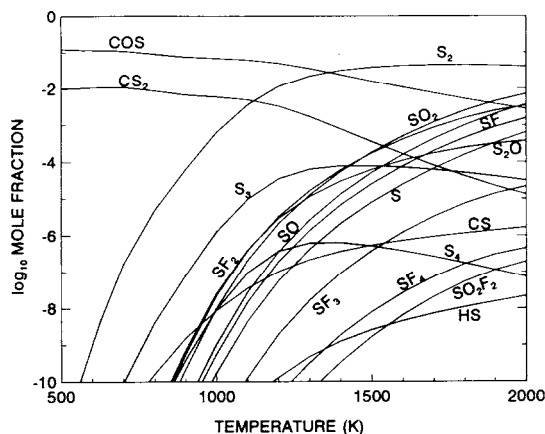


Figure 17. The calculated abundances of the major S-bearing gases in the same model are plotted as a function of temperature for a total pressure of 1 bar. Note the absence of  $\text{H}_2\text{S}$ , which has an insignificant abundance at all temperatures considered (figure from Fegley 1992).

in the lunar interior, and if any remained trapped in lunar glasses or rocks, their abundances in principle provide important constraints on compositional conditions at the time the gases were generated.

Finally, Fig. 18 illustrates the chemistry of two of the metals, Cu and Fe, which were also included in the thermodynamic calculations. Fegley (1991) showed that with very few exceptions the chlorides and fluorides were the major species for the rock-forming elements, volatile elements, and ore-forming metals included in his calculations. Similar results were reported later by Colson (1992). The results in this figure illustrate the same point. Copper chemistry in lunar volcanic gases is predicted to be dominated by  $\text{CuCl}$  over a wide temperature range, down to about 800 K where the trimer  $(\text{CuCl})_3$  takes over. Unpublished calculations by Fegley (1992) also predict similar behavior for Cu in terrestrial volcanic gases. These predictions are in accord with the observations by Murata (1960) of  $\text{CuCl}$  in volcanic gases.

In the case of Fe, two gases  $\text{FeF}_3$  and  $\text{FeCl}_3$  are the dominant Fe gases over the 500 to 2000 K temperature range, with small amounts of  $\text{FeCl}_2$  also being formed at temperatures below 1000 K. As noted in Fegley (1991) other transition metals display qualitatively similar chemistry.

Finally we note that the results presented in Fig. 18 and the more extensive results discussed in Fegley (1991) and Colson (1992) are complemented by the proposal of Gillett (1990) that chloride magmas may have fractionated and concentrated metals in the lunar interior.



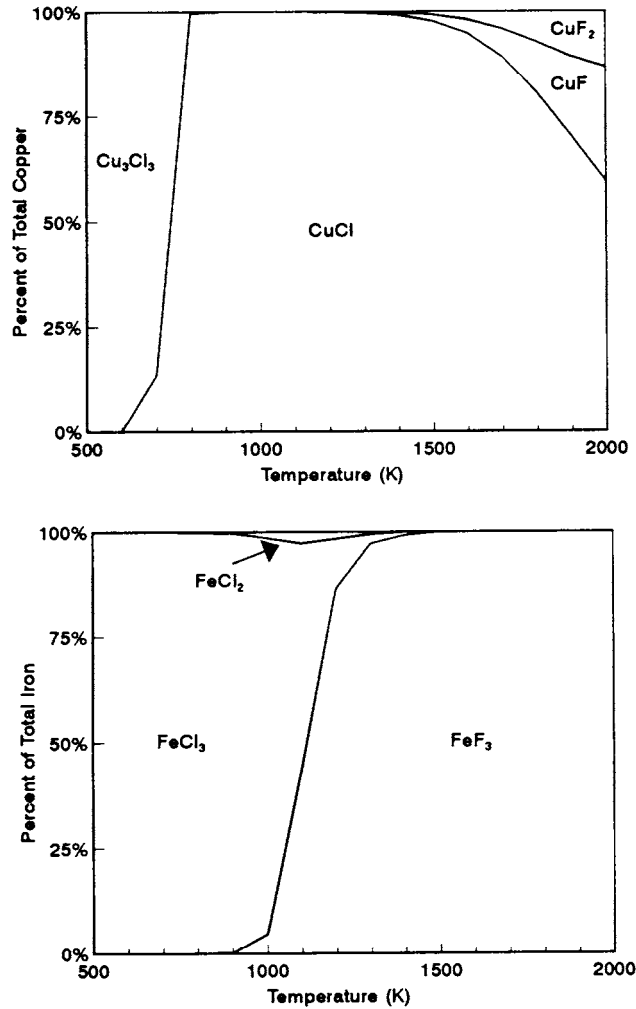


Figure 18. (top) The gas phase chemistry of copper as a function of temperature for the model lunar volcanic gas composition illustrated in Figs. 14–17. Note that in this graph the abundance scale expresses the percent of total Cu which is in each gas. Copper is a relatively volatile ore-forming metal and CuCl(g) has been observed in terrestrial volcanic gases (Murata 1960). (bottom) The same for iron, except that the abundance scale expresses the percent of total Fe which is in each gas. Fe-bearing minerals are found in sublimates collected from volcanic gases and Fe is enriched in the aerosol + vapor of volcanic gases relative to the magma (see, e.g., Symonds et al. 1987).

## VII. SUMMARY

In this section we summarize what we regard as the major remaining unanswered questions with regard to the utilization of lunar volatiles as resources. We also present our recommendations for future work specifically directed towards the utilization of lunar volatiles as resources.

### A. Remaining Unanswered Questions

The major remaining unanswered questions of interest from a resources perspective include the following:

1. What is the average distribution of solar-wind-implanted volatiles with depth? Different reasonable models lead to order-of-magnitude differences in calculated total inventories, by far the largest uncertainty for most volatiles.
2. Are exposed surfaces of Apollo soils saturated with any or all of the solar-wind-implanted volatiles? If not, limb and farside locations become much more promising for resources.
3. Is there water trapped near the lunar poles? If the amount is significant, this would be by far the most valuable source of H on the Moon.
4. What is the actual abundance of hydrogen and of water in lunar materials? The existing laboratory data for hydrogen indicate a mean abundance of about  $50 \mu\text{g g}^{-1}$  in bulk lunar soils, but this may be high due to contamination by terrestrial water. In many cases, no isotopic data were taken when hydrogen abundances were determined, and thus potentially important information about the degree (if any) of terrestrial contamination is unavailable. In the case of water, the actual water content of lunar materials is still unknown, because all the water detected could be explained by terrestrial contamination.

### B. Recommendations for Future Work

The following studies are of interest from a resources perspective:

1. Determination of the abundance of solar-wind-implanted volatiles at depths of  $>3$  m for several sites, either by using drill cores, trenches, or penetrators. As this recommendation implies, either returned samples or *in-situ* observations could be used to get the required data.
2. Determination of the feasibility of various extraction techniques for solar-wind-implanted volatiles. In particular, extraction by acid etching (preferably in a closed system involving recycling of the aqueous phase) or abrasion should be studied from an engineering perspective, and compared with thermal extraction processes. If lunar fluorine extraction schemes are feasible, then etching by  $\text{F}_2$  may also be a possibility.
3. Determination of the water abundance (if any) in the lunar polar regions. A polar orbiting mission with a gamma-ray spectrometer would be able

to detect economically important deposits (see, e.g., Feldman et al. 1991; Metzger and Drake 1990).

4. Determination of the actual hydrogen and water contents of different types of lunar materials with an emphasis on lunar soils and breccias. Microanalytical techniques such as the ion microprobe and improved stepwise heating techniques, e.g., as employed by Stievenard et al. (1990), may be very useful for these studies. The combined determination of abundances and isotopic compositions is highly recommended.

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