

Solar wind and cosmogenic rare gases in Luna 16 and 20 soils and their correlation with cosmic ray produced fossil tracks in lunar samples

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Abstract. The elemental and isotopic composition of Ne, Ar, Kr and Xe were measured in the bulk soil samples obtained from Luna 16 and 20 missions. The average cosmic ray exposure ages for the Luna 16 soils is higher than that of Luna 20 soils. The trapped solar wind composition is deduced by correcting for the cosmogenic component and is found to be in good agreement with the values published by others. The correlation between the solar wind gases and cosmic ray tracks in lunar samples is quantitatively studied and some systematic features regarding lunar regolith dynamics are tentatively established.

Keywords. Solar wind gases; lunar soils; cosmic ray exposure ages; solar cosmic ray tracks.

1. Introduction

Noble gas and nuclear particle track studies in the silicate grains of gas rich meteorites have led to the conclusion that the trapped rare gases are of solar composition (Martí 1969, Mazor and Anders 1967, Eberhardt *et al* 1965, Pepin and Signer 1965, Signer and Suess 1963) and the particle tracks are due to solar cosmic rays (Lal and Rajan 1969, Pellas *et al* 1969). It was further suggested that the soil samples from the moon should show similar effects as the surface materials on the moon are also directly exposed to solar wind and solar flare particles. Recent studies on moon samples have confirmed these predictions as they contain large amounts of trapped solar wind noble gases and also reveal particle tracks both due to solar and galactic cosmic ray bombardment. A small fraction of the rare gases in the lunar soil material is contributed by the galactic cosmic-ray spallation on suitable target elements.

In this report, we present our measurements of Ne, Ar, Kr and Xe in a few representative samples from Luna 16 and 20 drill cores. The aims of our investigation are to determine the average abundance and isotopic composition of the solar wind particles trapped in the surface of silicate grains, to determine the average cosmic ray exposure age of the samples, and to look for possible correlation between solar wind and solar cosmic ray effects in lunar soil samples. Comparisons of our data with the literature have been made, wherever possible,

2. Experimental

The mass spectrometer used in our studies is a 4.5", 60° single focusing Reynolds' type glass instrument developed indigenously (Gopalan *et al* 1973). All measurements were made in the static mode, and peaks obtained by magnetic scanning. The instrument was operated at low sensitivity (1.1×10^{-11} and 2.67×10^{-11} cc STP/mv for ^{22}Ne and ^{36}Ar respectively) for Ne and Ar and high sensitivity (1.40×10^{-12} and 1.65×10^{-13} cc STP/mv for ^{86}Kr and ^{132}Xe respectively) for Kr and Xe. An on-line gas extraction and purification was used consisting of high vacuum sample furnace, titanium getters and charcoal traps for gas separation. Samples (typically 1 mg) were heated by RF induction to about 1600° C in a single step. After gas purification, Ne, Ar and Kr + Xe were separated by selective desorption from charcoal at -190°C , -78°C and 100°C , respectively, and introduced into the mass spectrometer individually.

Absolute gas concentrations were calculated from ion beam intensities, calibrated by analysing known volumes (typically 0.01 cc at STP) of air samples before and after each sample run. Isotopic mass discriminations were determined in the same calibrations, based on current values for the isotopic composition of atmospheric noble gases (Nier 1950). Furnace blanks at 1600° C were run periodically and amounted to a couple of per cent of the sample gases.

The Luna samples were received in plastic vials. The samples selected for analysis were weighed and packaged in aluminium foils without further processing. Any atmospheric adsorption during sample handling was sought to be removed by keeping the furnace at 200° C for 48 hr before melting the samples.

3. Results

The elemental and isotopic abundances of Ne, Ar, Kr and Xe measured in the four samples are listed in tables 1 and 2 along with their zones and depth. For comparison, data on comparative samples by other workers are also given. The xenon fraction of L 1629 was lost due to machine instability. Listed errors in isotopic composition are 1σ statistical errors from peak height measurements and averaging. Total error due to other contributing factors such as variable mass discrimination

Table 1. Elemental and isotopic composition of Ne and Ar in Luna samples

Sample	Zone	^{22}Ne	^{36}Ar	^{20}Ne	^{21}Ne	^{38}Ar	^{40}Ar
		10^{-8} cc	STP/g	^{22}Ne	^{22}Ne	^{38}Ar	^{38}Ar
L-1629 (bulk)	A (0-8 cm)	16.23	51.00	13.056	0.035	0.1881	2.470
L-1630 (bulk)	B (20-28 cm)	8.50	66.17	12.475	0.0368	0.1865	1.478
L-1608 (0-83 μ)	D (28-33 cm)	12.82	83.00	12.838	0.035	0.1870	1.943
L-2004 (0-83 μ)	4 (32-41 cm)	7.07	48.70	12.99	0.037	0.1858	2.01

(Approximately 1 mg of sample is taken for analysis).

Table 2. Elemental and isotopic composition of xenon and krypton in Luna 16 and 20 soil samples

Sample	$\frac{^{136}\text{Xe}}{^{132}\text{Xe}}$	$\frac{^{134}\text{Xe}}{^{132}\text{Xe}}$	$\frac{^{131}\text{Xe}}{^{132}\text{Xe}}$	$\frac{^{130}\text{Xe}}{^{132}\text{Xe}}$	$\frac{^{128}\text{Xe}}{^{132}\text{Xe}}$
	L-1630	0.3022 ±0.0044	0.3673 ±0.0036	0.8256 ±0.0054	0.1683 ±0.0040
L-1608	0.2954 ±0.0040	0.3746 ±0.0042	0.8036 ±0.0060	0.1685 ±0.0030	1.0370 ±0.0060
L-2204	0.2964 ±0.0060	0.3689 ±0.0055	0.8146 ±0.0064	0.1680 ±0.0042	0.9949 ±0.0080
Luna 16 (1-400 μ)*	0.3020	0.3690	0.9360	0.1676	1.031
	$\frac{^{128}\text{Xe}}{^{132}\text{Xe}}$	$\frac{^{126}\text{Xe}}{^{132}\text{Xe}}$	$\frac{^{124}\text{Xe}}{^{132}\text{Xe}}$	$\frac{^{132}\text{Xe}}{^{132}\text{Xe}}$ (10 ⁻⁸ cc STP/g)	
L-1630	0.0942 ±0.0040	0.0095 ±0.0020	0.0109 ±0.0018	3.95	
L-1608	0.0808 ±0.0020	0.0064 ±0.0018	0.0089 ±0.0020	4.13	
L-2204	0.0796 ±0.0041	0.0047 ±0.0015	0.0051 ±0.0015	3.0	
Luna 16 (1-400 μ)*	0.0897	0.0087	0.0071	2.2	
	$\frac{^{84}\text{Kr}}{^{86}\text{Kr}}$	$\frac{^{83}\text{Kr}}{^{86}\text{Kr}}$	$\frac{^{82}\text{Kr}}{^{86}\text{Kr}}$	$\frac{^{80}\text{Kr}}{^{86}\text{Kr}}$	$\frac{^{86}\text{Kr}}{^{86}\text{Kr}}$ (10 ⁻⁸ cc STP/g)
L-1629	3.2640 ±0.0520	0.6767 ±0.0124	0.6929 ±0.0119	0.1348 ±0.0120	12.5
L-1630	3.3490 ±0.0502	0.6994 ±0.0112	0.6969 ±0.0098	0.1579 ±0.0127	13.24
L-1608	3.3347 ±0.0508	0.6786 ±0.0118	0.6828 ±0.0110	0.1388 ±0.0091	9.15
L-2004	3.2541 ±0.0541	0.6677 ±0.0109	0.6664 ±0.0122	0.1455 ±0.0140	7.1
Luna 16 (1-400 μ)*	3.247	0.661	0.653	0.1310	7.8

* Eugster *et al* (1973)

is about 2%. Cumulative error in absolute gas concentration due to errors in weighing, blank correction and volumes of standard air samples is about 20%.

4. Discussion

(a) Trapped gases

The two main components of noble gases present in lunar soil are trapped solar wind ions and cosmic ray spallation products. Such a strictly two-component mixture in the case of Ne can be represented by the following equation (Eberhardt *et al* 1970).

$$\frac{^{22}\text{Ne}}{^{21}\text{Ne}_m} = \frac{^{21}\text{Ne}_e}{^{21}\text{Ne}_m} \left[\left(\frac{^{22}\text{Ne}}{^{21}\text{Ne}} \right)_e - \left(\frac{^{22}\text{Ne}}{^{21}\text{Ne}} \right)_t \right] + \left(\frac{^{22}\text{Ne}}{^{21}\text{Ne}} \right)_t \quad (1)$$

where

$$\left(\frac{^{22}\text{Ne}}{^{21}\text{Ne}} \right)_m, \left(\frac{^{22}\text{Ne}}{^{21}\text{Ne}} \right)_t \quad \text{and} \quad \left(\frac{^{22}\text{Ne}}{^{21}\text{Ne}} \right)_e$$

are the isotopic ratios measured in a sample, in the trapped solar wind and spallation components respectively. The concentration of solar wind ions in lunar soil particles is surface correlated while that of cosmic ray spallation products is volume correlated (Eberhardt *et al* 1970, Hintenberger *et al* 1972). A set of grain size fractions of a bulk soil sample will therefore differ in the relative concentrations of the trapped and spallogenic components and plot on a straight line in a $(^{22}\text{Ne}/^{21}\text{Ne})_m$ vs $(1/^{21}\text{Ne})_m$ diagram according to the above equation. Our bulk soil data conform within experimental errors with such correlation lines obtained

Table 3. Cosmic ray exposure ages of Luna 16 and 20 samples

Sample	$^{21}\text{Ne}_e$	$^{38}\text{Ar}_e$ (10^{-8} cc STP/g)	$^{136}\text{Xe}_e$	Exposure Ages (m.y) ^a		
				^{21}Ne	^{38}Ar	^{136}Xe
L-1629	40.20	156.0	..	304	912	..
L-1630	41.83	112.5	2.0	317	660	930
L-1608	36.60	189.0	2.0	278	1105	440
L-2004	34.80	60.0	(0.2)	210	351	(200)
*Luna 16 (Average) (20-22 cm)	58	..	1.02	440	..	330
†Luna 16 (A zone)	865
‡Luna 16 (29-31 cm)	2.10	1090
**Luna 20 (44)	286	614

^a Production rates (cc STP/g.m.y.): $^{21}\text{Ne} = 0.132 \times 10^{-6}$; $^{38}\text{Ar} = 0.170 \times 10^{-6}$
 $^{136}\text{Xe} = 1.3 \times 10^{-21}$ cc STP/year/PPMBa.

* Eugster (1973). † Pepin *et al* (1972); ‡ Kaiser (1972). ** Heymann *et al* (1973).

by Eberhardt *et al* (1970) on size fractions of Apollo 11 soil and so are consistent with the trapped gas composition of $(^{22}\text{Ne}/^{21}\text{Ne})_t = 31.0 \pm 1.0$ given by the above workers. Assuming the departure of our data from the correlation line is due to minor variations in the concentration of spallogenic ^{21}Ne , the same for individual samples can be calculated from eq. (1) and using $(^{22}\text{Ne}/^{21}\text{Ne})_e = 1.17$ (Eberhardt *et al* 1970). The production rates used and $(^{21}\text{Ne})_e$ exposure ages calculated are shown in table 3.

A similar comparison of our argon data with the grain size correlation obtained by Pepin *et al* (1972) on Luna 16 indicates that the trapped $(^{38}\text{Ar}/^{36}\text{Ar})$ ratio is close to 0.185. Using $(^{38}\text{Ar}/^{36}\text{Ar})_e = 1.53$ (Huneke *et al* 1972) spallation ^{38}Ar concentrations and ^{38}Ar exposure ages are derived and shown in table 3. Since the $(^{38}\text{Ar})_e$ concentrations are about a few per cent of the total ^{38}Ar , the ^{38}Ar ages have relatively large errors.

The low abundant isotopes of Kr and Xe are quite strongly affected by noble gases resulting from cosmic ray spallation reactions on Ba, Sr and rare earth elements. We have corrected the measured Xe and Kr of each sample for spallation contribution by assuming an average spallation spectrum for Kr and Xe (Funkhouser *et al* 1970, Hohenberg *et al* 1970, Kaiser 1972) and one isotope ratio in the trapped xenon, namely, $(^{126}\text{Xe}/^{136}\text{Xe})_t = 0.0141$ (Eberhardt *et al* 1972) and $(^{136}\text{Xe})_e = 0$. The spallation ^{126}Xe concentrations and the corresponding exposure ages are given in table 3.

We assign an error of about 30% for our ^{21}Ne and ^{126}Xe exposure ages. ^{38}Ar exposure ages are subject to much larger errors as the cosmogenic argon is almost masked by trapped argon. Table 3 also lists the exposure ages measured by other workers for comparable Luna 16 and 20 samples. The results of different workers are concordant within stated experimental errors for comparable samples and by the same method. For example, our ^{21}Ne ages for Luna 16 samples agree within 30% with that for a Luna 16 grain size fraction measured by Eugster *et al* (1973). However, the ages for the same sample by different methods do not sometimes agree. The Luna 16 and 20 exposure ages appear to be similar to those of Apollo soils, though there is a possibility of Luna 16 soil being older.

The isotopic abundances of the trapped Kr and Xe derived as mentioned earlier from the measured spectra agree well with the isotopic composition of surface correlated Kr and Xe in the Apollo 12 soil designated as BEOC-12001 (Eberhardt *et al* 1972) and with the spectrum for the solar wind component of Apollo 11 soil (designated as SUCOR) reported by Podosek *et al* (1971). It may be pointed out that the BEOC and SUCOR composition were calculated by a direct method using a comparison of different lunar soil samples or their size fractions, without the necessity of any assumptions about possible solar wind and spallation spectra, or relative spallation production rates of different elements. The agreement of our trapped gas spectrum with the BEOC-12001 composition is shown in figure 1 where the two spectra are compared using the atmospheric Xe composition as reference in the usual δ notation:

$$\delta_{132}^M = \left[\frac{(^m\text{Xe}/^{132}\text{Xe})_m}{(^m\text{Xe}/^{132}\text{Xe})_{\text{atm}}} - 1 \right] \times 100$$

where m refers to either BEOC or our data. Light isotopes have been omitted due to large spallation contribution in them and relatively large measurement

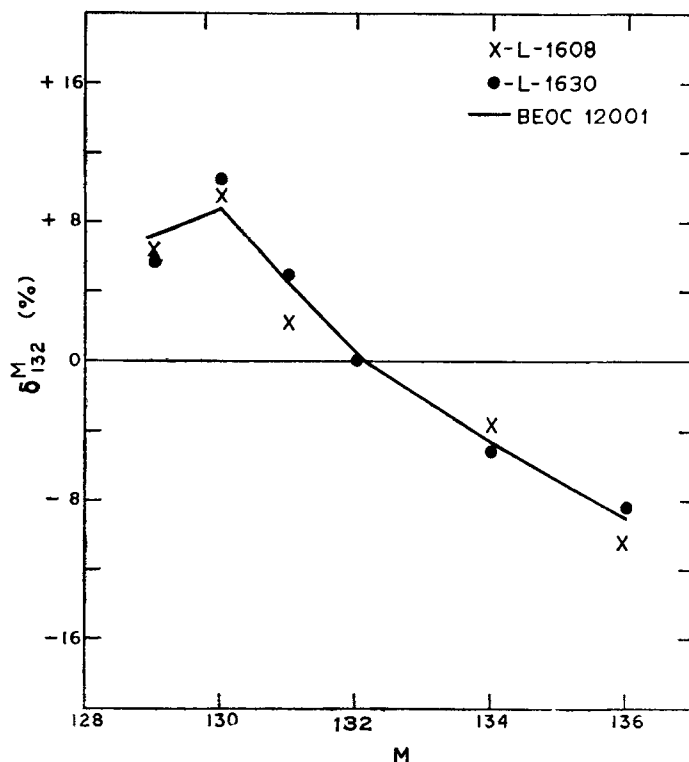


Figure 1. Comparison of the deduced isotopic Xe composition from Luna 16 soil fractions after correcting for spallogenic contribution, with the surface correlated solar Xe from BEOC-12001 data of Eberhardt *et al* (1972).

errors. The overall agreement of our calculation with BEOC spectrum justifies the validity of our assumption for spallation correction to the heavier isotopes of Xe.

It is of interest to compare the relative abundances of the trapped noble gases measured in Luna samples with the results of other workers as well as those found in other known reservoirs such as meteorites and atmosphere. The $^{86}\text{Kr}/^{36}\text{Ar}$ and $(^{132}\text{Xe}/^{36}\text{Ar})$ ratios in our samples range from $(3.7 - 6.8) \times 10^{-4}$ and $(0.5 - 0.6) \times 10^{-4}$ respectively. These are in good agreement with the results of Eberhardt *et al* (1972) and Bogard *et al* (1972). However, the $^{20}\text{Ne}/^{36}\text{Ar}$ ratios in the L-1630, 1608 and 2004 are lower by about a factor of 2 as compared with Apollo 12 bulk fines (Eberhardt *et al* 1972) and Apollo 14 fines (Bogard *et al* 1972). This difference may be either due to the increased content of plagioclase in Luna soil (Heymann *et al* 1973) with its apparently poor retentivity for Ne or due to differential trapping and implantation of solar wind gases (Bogard and Nyquist 1972).

The abundances of trapped gases relative to ^{36}Ar in our measurements are compared in figure 3 with the cosmic abundances (Aller 1961), atmosphere (Nier 1950), the gas rich meteorite Pesyanoe (Marti 1969) and the Luna 16 soil data of Kaiser (1972). The overall relative abundance pattern of the trapped gases is generally similar to the one predicted for the solar wind gases observed in gas-rich meteorites (Marti 1969) and Eberhardt *et al* (1970).

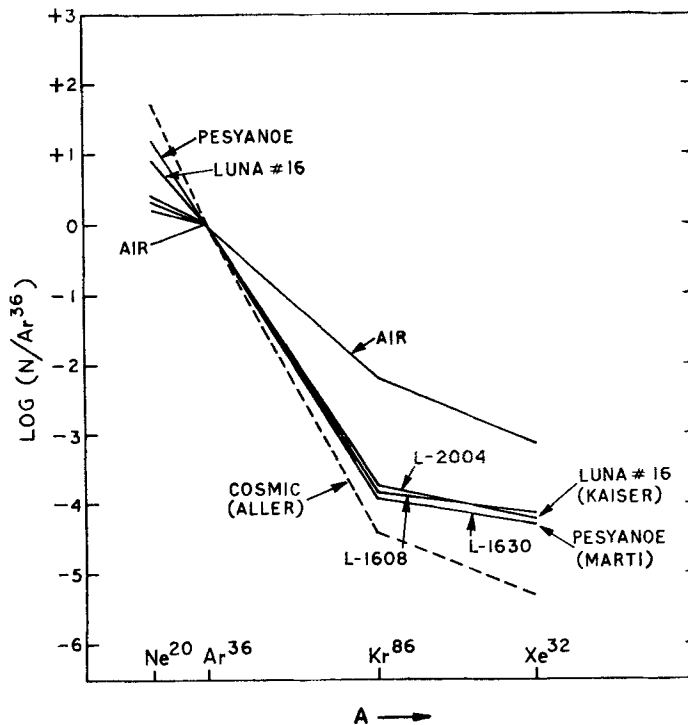


Figure 2. Abundance of trapped noble gases relative to ^{36}Ar observed in Luna 16 and 20 bulk soil samples. These data are compared with those of Pesyanoë (Marti 1969), and Luna 16 (Kaiser 1972) as well as cosmic abundances (Aller 1961) and air (Nier 1950). The lines representing Kaiser (1972) and Marti (1969) data could not be shown separately as they agree closely to those of L-1608, 1630 and 2004 for Ar, Kr and Xe. The deviations could be indicated only for Ne.

It is not clear how closely the trapped gas spectrum of the lunar soil obtained in this work and elsewhere represents the primary solar wind composition. Some of the processes responsible for this alteration are mass fractionation, lunar atmospheric phenomena, rare gas diffusion (Eberhardt *et al* 1970, 1972, Podosek *et al* 1971) and possibly the solar flare-induced nuclear reactions in lunar regolith materials (Rao *et al* 1971).

(b) *The relation between solar cosmic ray tracks and solar wind rare gases in lunar soil samples*

The silicate grains located at the uppermost layers of lunar regolith are not only subjected to the solar wind implantation but also to the solar flare bombardment. The result of the solar flare bombardment of the above grains is a very high track density $\geq 10^8/\text{cm}^2$ and/or track density gradient due to very heavy nuclei in solar flares (Arrhenius *et al* 1971), whereas the solar wind implantation produces very high concentrations of solar wind rare gases in the surface layers of those grains (Eberhardt *et al* 1972). It follows that grains in the lunar soil with high track densities should show high solar wind concentrations.

Using the cosmic ray track data of the lunar soil grains, Arrhenius *et al* (1971) proposed that two parameters N_{r}/N and ρ_{q} are sufficient to characterise the dura-

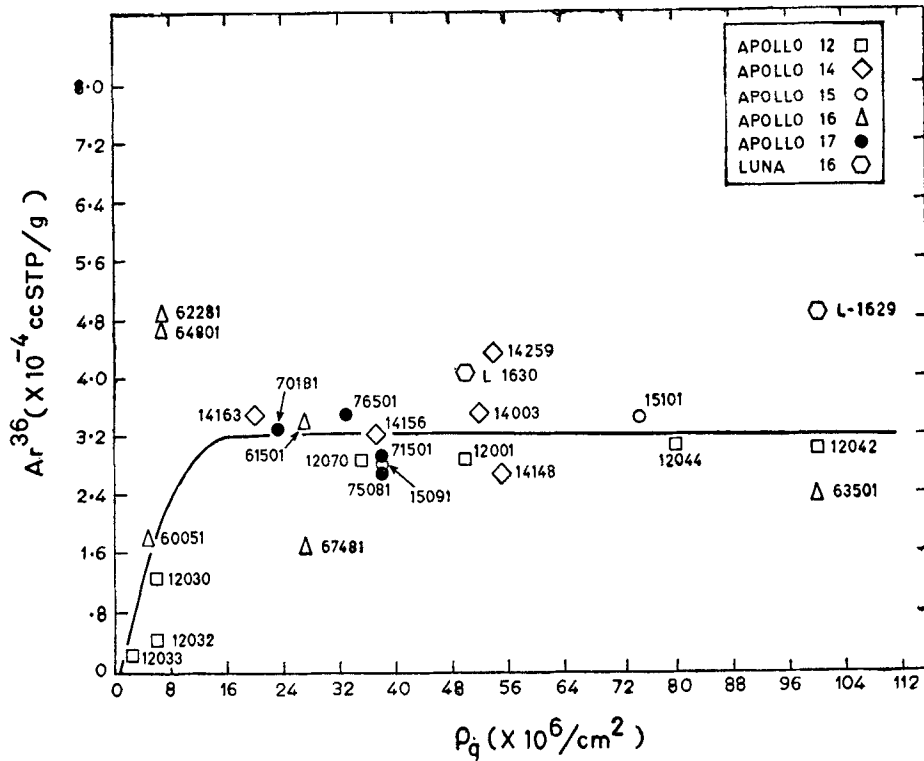


Figure 3. Correlation between the measured ^{36}Ar contents with the ρ_q (the quartile, track density) values given for several Apollo and Luna samples (Arrhenius *et al* 1971 and Bhandari *et al* 1972 and 1973). ^{36}Ar contents are taken from Hintenberger *et al* (1971), Heymann *et al* (1972), Pepin *et al* (1972), Kirsten *et al* (1972), Bogard *et al* (1972), Berdot *et al* (1972).

tion and the pattern of lunar surface irradiation. N_{H}/N is the fraction of grains exposed in the uppermost layer within about a few hundred microns from the surface (where N_{H} is the number of grains having high track density $\geq 10^8/\text{cm}^2$ or track density gradient in a total N number of grains. ρ_q is the quartile track density (a value such that a quarter of the grains in a sample have track densities lower than ρ_q) and it is an index of time for which a layer is exposed at the lunar surface (Arrhenius *et al* 1971). We have plotted in figure 3, the ρ_q values given by Arrhenius *et al* (1971) and Bhandari *et al* (1972, 1973) for a large number of bulk samples from Apollo and Luna missions against the ^{36}Ar concentrations measured by several groups, including our Luna data. In spite of the scatter of points, due to experimental errors in figure 3, a trend indicating that the solar wind-trapped ^{36}Ar saturates at a ρ_q (no. of tracks/ cm^2) value of about 16×10^6 could be easily seen. This corresponds to about 15 m.y. for producing a saturation ^{36}Ar concentration in the grains present in an average thickness of ~ 3 cm for the lunar soil layer. Since the ^{36}Ar saturation is indicative of a layer derived by Bhandari *et al* (1973) from of ~ 20 m.y. for an average lunar soil well-mixed soil layer, the mixing time scales a plot of N_{H}/N versus ρ_q is consistent with the present value. The average residence time of a grain at the top of the lunar regolith such that it attains a track density $\geq 10^8/\text{cm}^2$ is deduced to be $\sim 10^4$

years by Arrhenius *et al* (1971) from these track measurements. We calculate such a parameter from the rare gas measurements in the following way. From figure 3 we get a saturation concentration of ^{36}Ar to be $\sim 3 \times 10^{-4}$ cc STP/g. The present day solar wind flux of ^4He is 6.3×10^6 atoms/cm²/sec and a $^4\text{He}/^{36}\text{Ar}$ ratio of 1.8×10^4 (Eberhardt *et al* 1972, Marti 1971). Assuming that the average grain size is about 60μ in diameter which roughly corresponds to the median grain size of most of the soils and a solar wind penetration depth of $\sim 0.3 \mu$ (Eberhardt *et al* 1971), we find about 500 years are sufficient for the grains to remain exposed to the solar wind at the top of the lunar regolith to acquire the required ^{36}Ar concentrations. Knowing the average value of ~ 500 years for the surface residence time, one calculates the effective time-scales for mixing an average lunar soil layer of ~ 3 cm thickness using the micrometeorite influx rate deduced on the basis of microcrater counts on lunar rock surface (Gault *et al* 1972). It is found that the micrometeorite flux in the size range $0.1 - 0.5$ cm is sufficient to produce the mixing of the average soil layer in about 15 m.y. which is in agreement with value reported by Bhandari *et al* (1973) from track measurements.

5. Conclusion

In Luna 16 and 20 bulk soil samples taken at different depths of the drill cores, Ne, Ar, Kr and Xe were measured in total melting runs. Using standard procedures, the gross composition is decomposed into cosmogenic and solar wind components. The average cosmic ray exposure ages for these soils are determined by ^{21}Ne , ^{38}Ar and ^{136}Xe methods and our values agree with those determined by Eugster *et al* (1973) and Pepin *et al* (1972). The measured Xe and Kr isotopic data are corrected for spallogenic contribution, using their average cosmogenic spectra and the trapped solar wind composition thus deduced is in agreement, within experimental errors, with that reported for BEOC by Eberhardt *et al* (1972). The total ^{36}Ar contents of several Apollo and Luna samples, including the data reported here, are compared with the nuclear track data reported by Bhandari *et al* (1972, 1973) in these soil samples and the resultant correlation between them yields a value of ~ 500 years for the average residence time for these soil grains to attain solar wind saturation ^{36}Ar concentration of $\sim 3 \times 10^{-4}$ cc STP/g. From these data, in a qualitative way, we calculate a mixing time scale of ~ 15 m.y. for the average lunar surface layer of about 3 cm thickness.

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