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ДУБНА

E7 - 9636

C.Fiéni, O.Otgonsuren, P.Pellas, V.P.Perelygin,
S.G.Stetsenko, N.N.Gavrilova

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C.Fiéni,* O.Otgonsuren, P.Pellas,* V.P.Perelygin,
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ABUNDANCES OF Z > 52 NUCLEI IN GALACTIC COSMIC RAYS: LONG TERM AVERAGES BASED ON STUDIES OF PALLASITES

* Laboratoire de Mineralogie du Museum and C.N.R.S., Paris, France.

Since the successful studies by Maurette et al.[2,6] of tracks of very heavy cosmic ray nuclei in olivines from pallasites, a large amount of information on energy and charge spectra has been obtained by studying crystals from meteorites and lunar rocks. The existence of cosmic rays with $Z > 30$ was in fact established by means of track length measurements in pyroxenes from the Estherville mesosiderite (Fleischer et al.[13]) and in olivines from the Eagle Station pallasite (Maurette[27]). These data led to a value for the abudance of all elements with $Z > 32$ (VVH group) relative to Fe group (VH group) of about 2.5×10^{-4}, in rather good agreement with the ratios for the "universal" abundances of $\sim 4.5 \times 10^{-4}$ (ref.[41]). By using an alternating etching and polishing technique Maurette et al.[28] were able to measure in pyroxenes of the Johnstown achondrite surface tracks of length exceeding 300 μm. One of the tracks recorded has a length of 1050 μm; this track was attributed to a nucleus with $Z > 80$. A definite progress was made by the development of the technique for revealing TINT and TINCLE type tracks[21] which allowed a direct study of the total recordable length of VVH particle tracks. However, with this method Lal et al. [21] did not obtain tracks longer than ~500 μm. In pigeonite crystals from from lunar rock 12021 Price et al. [24] observed two possible tracks having minimum lengths of ~0.9 and ~1 mm which were assigned to ultraheavy cosmic rays with $Z > 80$. Thus far 3 tracks have been detected whose minimum lengths are in the range of 0.9 - 1 mm in meteoritic or lunar pyroxenes.

We report here on the first measurements of cosmic ray tracks in olivines of pallasites with lengths in the range 0.5 - 0.9 mm.

We believe pallasitic olivines to be the most suitable meteoritic crystals for studying elemental abundances of the VVH primary beam for the following reasons: (a) The large sizes of crystals (up to a few centimeters) and their transparency allow an easy and reliable study of the frequency distribution of lengths of long tracks; (b) The chemical composition of olivines is homogeneous within the same pallasite [7]; (c) The higher threshold ionisation for track registration in olivine as compared to pyroxene lowers the background of iron group tracks by about a factor of 2 [3,23,30] and finally (d) the exposure ages of pallasites are in general higher than those of stony meteorites [29]. For example, the exposure ages of Marjalahti and Eagle Station are about 175 and 50×10^6 y, respectively [19,23].

Furthermore, since in pallasites, olivines are embedded in a nickel-iron matrix, space erosion due to micrometeorites impacts should be less than that in lunar rocks $(\sim 0.5-1) \times 10^{-7}$ cm/y [2,12,17] and stony meteorites [9,11,33].

On the other hand, olivines from pallasites often contain rather high dislocation densities and capillary inclusions which must be distinguished from VVH tracks. Most of the dislocations are, however, easily recognized because they generally occur in groups (nearly parallel) or are curved. The capillary inclusions and hollow channels are definitely oriented along the main crystallographic axes and are visible without any etching.

Due to the low abundance of ultra-heavy nuclei in the cosmic radiation and because of the fact that they are quicky absorbed in meteoritic matter due to nuclear interactions, it is important to choose sampling locations as close as possible to the preatmospheric surfaces. A preliminary survey of ilivines sampled on external regions of our Marjalahti fragment provided the locations with the highest VH track densities $(5-6 \times 10^6\ /cm^2)$. A similar approach was used with the sawn slice of Eagle Station from the Paris Museum. In the latter case the VH track densities measured on external locations were much higher $(1.2 - 1.6 \times 10^7\ /cm^2)$ despite the lower exposure age of Eagle Station. Previous studies by Maurette [27] of the same Eagle Station sample led to values for VH track densities systematically smaller by a factor of 4 compared to our results.

In order to increase the statistics and to favour the registration of long tracks, we choose to study crystals whose orientation was mostly at right angles to the preatmospheric surface [13]. The surface area in the case of Marjalahti olivines was usually larger than 4 mm^2 and a number of crystals has observational surfaces between 10 and 20 mm^2.The preliminary study of Eagle Station was performed on smaller crystal sizes.

After polishing the surface of olivine crystals, they were etched with WH solution of Krishnaswami et al.[20] slightly modified by adding 4g. of oxalic acid instead of 1 g. per liter of solution, the final pH being adjusted to 8.00 ± 0.05. The increase in the oxalic acid concentration led to a decrease in the half cone angle of VVH tracks from $(0.5 - 0.6)^{\circ}$ to $(0.3 - 0.4)^{\circ}$ both for the Marjalahti and Eagle Station olivines. The etching temperature was 110°C; to prevent evaporation, the etching was performed in teflon closed containers.

The total etchable length for tracks due to VVH nuclei was obtained by using either the "TINCLE" method of Lal et al.[21], which reveals deep-seated tracks inside the crystal along dislocation planes, pre-existing craks or fissures, or using a modified "TINT" method which consists of irradiating the crystals with a perpendicular artificially accelerated Xe beam (7.3 MeV/a.m.u.) producing tracks of about 55 μm length[34]. With our etching conditions, the most favourable "host-Xe" track density to reveal TINTS of VVH nuclei tracks inside the crystals was found to be about $5 \times 10^6 /cm^2$.

It has recently been demonstrated that artificially accelerated Fe nuclei of 3.16 MeV/a.m.u., produce average etchable track lengths of 13.5 μm in terrestrial olivine[34].Similar experiments with 6.5 MeV/a.m.u. Fe nuclei were carried out in previously annealed olivines from Marjalahti and Eagle Station. The accelerated Fe ions were implanted at an angle of 30° to the polished crystal surfaces and we used the "host-Xe -TINT" technique to measure the total recordable track lengths. A track-length histogram for the artificially implanted Fe ions in olivine of Marjalahti is shown in Fig. 1a; it can be compared with fossil Fe group track lengths in both Marjalahti and Eagle Station olivines (Figs. 1b, 1c and 1d).

The broadening of Fe track length distribution is primarily due to the difference in rate of etching of Fe tracks which cross totally - or only partially - the "host-Xe" tracks. The same average value for track lengths of accelerated Fe nuclei was obtained in the case of Eagle Station Station olivines; therefore we may conclude that the significant difference in fayalite content between the olivines of the two pallasites (Buseck and Goldstein, 1969) does not affect the etchable track lengths of Fe nuclei.

By comparing Fig. 1a with 1b and 1c-1d, it appears that the predominant fossil iron peak in Marjalahti histogram is shifted and broadened to short track lengths (peaking at about 7 μm) more significantly than that of Eagle Station (peaking at about 10.5 μm). The latter value is in good agreement with the fossil Fe track lengths measured in olivines from the Patwar mesosiderite[34]. Annealing experiments performed on accelerated Fe nuclei tracks in olivines have demonstrated that the length reduction

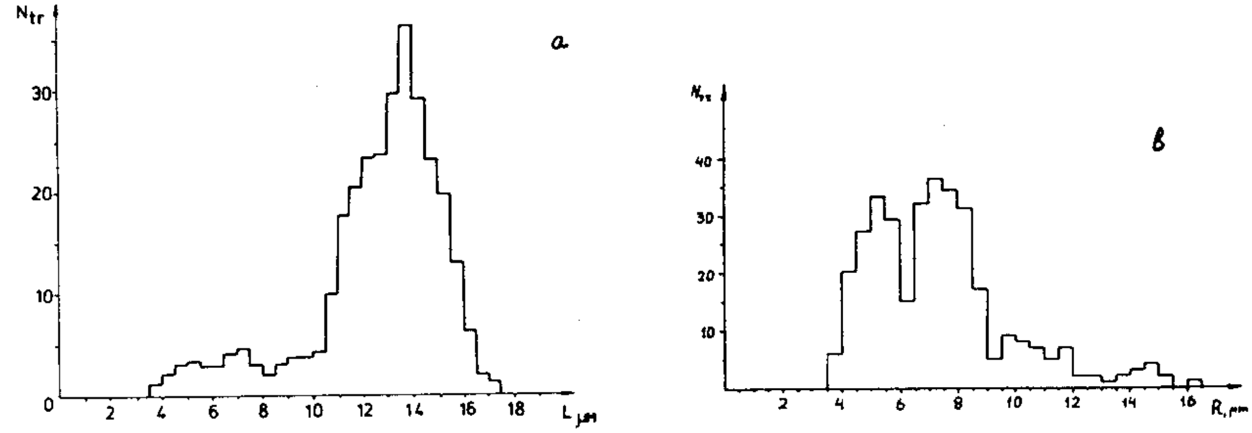
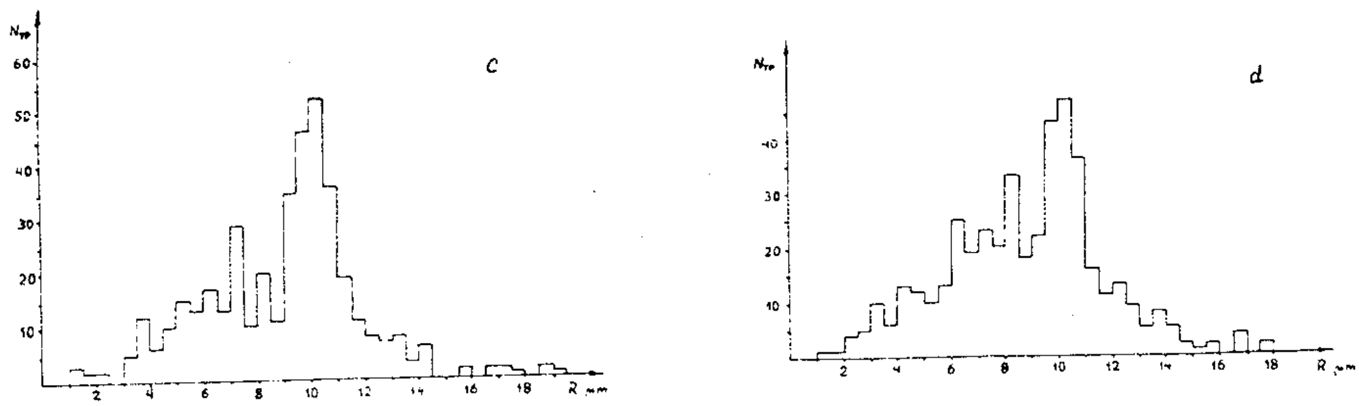


Fig. 1. a) Histogram showing the frequency distribution in track lengths due to accelerated **Fe** nuclei (E= 6.5 MeV/amu) in pre-annealed Marjalahti olivines. The tail extending to short lengths is due to **Cr** ions present as contamination in the **Fe** beam. Track length distribution of fossil cosmic ray tracks (Z <30) in b) Marjalahti olivines. As explained in the text, the shortening of **Fe** groups tracks is probably due to annealing effects during space exposure. c) Eagle Station olivines (location close to surface). d) Eagle Station olivines (internal location).



could be attributed to the radiation damage recovery of the highest energy portion of the particle path /15,34/. Since our annealing experiments for artificial Fe tracks performed on olivines from both Marjalahti and Eagle Station have shown an identical radiation damage recovery behaviour, despite the different chemical composition of the olivines, and since the mean track length of the predominant peak in Eagle Station is larger than in Marjalahti, we tentatively attribute the stronger fading of latent tracks in Marjalahti olivines as being due to differences in orbital elements; for instance a closer approach to the sun of the Marjalahti meteorite as compared with the orbit of Eagle Station object. However, the more pronounced space annealing conditions in the case of Marjalahti olivines did not alter markedly the $^{3}He/^{21}Ne$ ratio /29/ when corrected for differences in the composition of olivines and using the elemental production rates given by Bogard and Cressy /6/. This of course implies that etchable lengths of Fe tracks in olivine is a much sensitive monitor of any heating in space than ^{3}He diffusion losses in the same mineral /24/.

To obtain the value of track densities in the case of Marjalahti olivines for the same space annealing conditions as for Eagle Station, we made a linear correction for the reduction in the mean track length for the predominant peak ascribed to Fe nuclei, namely reduction from 10.5 μm to 7 μm. Furthermore because the threshold ionisation for VH particle track registration is higher in olivine than in pyroxene, a second correction

factor must be applied to precise the sampling depth. For ascertaining the preatmospheric depth, we made use indeed of the St Severin pyroxene data for which accurate track densities versus depth have been measured /9,22,24,42/. According to different authors the average-track density ratio for pyroxenes and olivines is equal to about 2 /3,24,30/.

In Table 1 are listed Fe group track densities measured in olivines and also the corrected values for differences in pyroxene/olivine registration characteristics. The calculated distances to preatmospheric surface and kinetic energy range are also given in the same table. We assumed an upper limit for space erosion of 5×10^{-9} cm/y in the case of Marjalahti; for Eagle Station the magnitude of erosion is negligible because of its shorter exposure age.

Figs. 1c and 1d show VH track length distributions for two depths in Eagle Station. The shielding differences between the two locations (about 9 g/cm^2 versus 29 g/cm^2) could explain the apparent increase of abundance of elements belonging to Cr group (V + Cr + Mn) as due to nuclear fragmentation. Note that the derived abundances of Cr group would be higher than the observed relative abundances of shorter length tracks considering the lower probability of recording shorter tracks due to V, Cr and Mn. With this correction we obtain for the abundance ration (V + Cr+Mn/Fe) a value of 1.0-1.5, in sharp disagreement with the recent results (0.29) of Lund et al. /25/ in the same energy interval. Similar very large abundance ratios of Cr group relative to Fe were also found in

Table 1

VH track densities and deduced preatmospheric radial depths of samples in Marjalahti and Eagle Station pallasites

Locations	Measured track densities in olivine* ($10^6/cm^2$)	Normalized track densities (pyrox.equivalent) ($10^6/cm^2$)	Distance to present surface (cm)	Deduced distance of the preatmospheric surface (cm) Chondritic Composition	Pallasitic Composition	Kinetic energy of VH nuclei (GeV/amu) $\varepsilon = 0$	$\varepsilon < 5 \times 10^{-9}$ cm/y
Marjalahti+							
n° 11	6.1	18.3	0.5	6.7	4.8	0.68	0.78
9	5.0	15.0	0.5	7.3	5.2	0.72	0.82
5	3.1	9.3	0.5	9.0	6.4	0.84	0.94
6	0.9	2.7	4–6	14.5	10.4	1.17	1.30
7	0.7	2.1	4–6	16.0	11.4	1.30	1.45
Eagle Station+							
n° 11	15	30	0.5–1	2.5	1.8	0.35	
10	10.5	21	2–3	3.5	2.5	0.45	
25	4.1	8.2	6.8–7.5	6.0	4.3	0.63	
23	2.5	5.0	10–10.5	8.0	5.7	0.77	

* Cosmic ray exposure ages: Marjalahti: 174×10^6 y /19,29/; Eagle Station: 51×10^6 y /29/.

+ Mean track density (β angle = 45°).

meteoritic or lunar crystals by other groups /1,31,32/. This apparent overabundance might be ascribed as due to the poor charge resolution of VH particle tracks in silicates, especially if space annealing processes shift the track lengths to lower values.

A total surface of 860 mm^2 (corresponding to a volume of 62 mm^3) was scanned in the case of Marjalahti olivines and the total recordable track lengths of VVH nuclei (i.e., tracks whose both ends are contained in the crystal volume) were measured.

Crystals with the highest VH track densities (see table 1) were selected and only tracks longer than 40 μm were measured. Our preliminary study of Eagle Station olivines was carried out on a surface of 60 mm^2 (~3.5 mm^3) in the track density region of 1.2 - $1.5 \times 10^6 /cm^2$. Figs. 2a and 2b show track length distributions of VVH nuclei in olivines of the two pallasites. In the case of Eagle Station the measured track lengths, when lengths exceed 300 μm, should be considered as lower limits because of the high VH particles densities and the cone angle condition which set limits to average maximum etchable track lengths, in agreement with the model calculations of Bhandari et al. /4/; these cases where measured track lengths should be considered as minimum values are shown as shaded in Fig. 2a.

Because of the higher statistics in the case of Marjalahti, we shall focus our attention to these results (see Fig. 2b which shows the track length distribution of 1424 tracks). The observed low abundance of

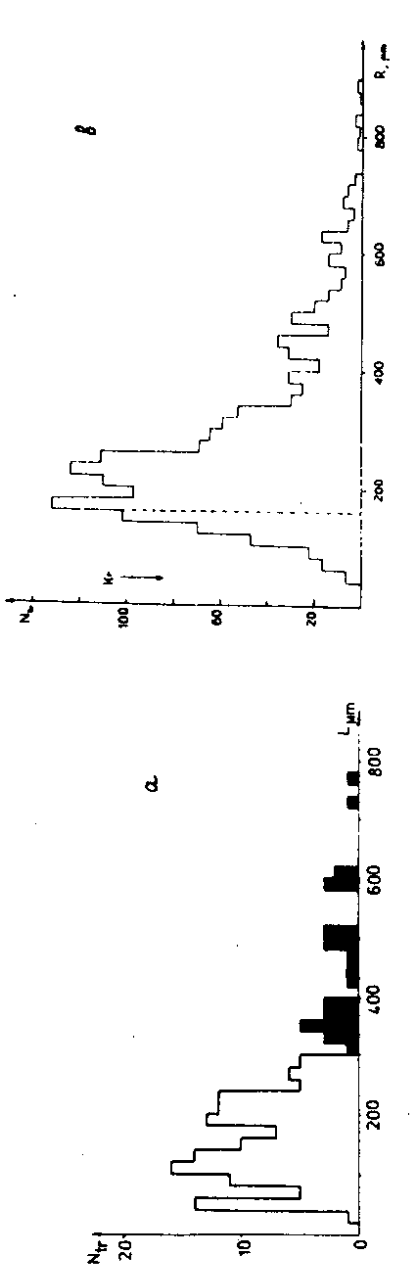


Fig. 2. Track lengths distribution of ultraheavy nuclei ($Z > 36$). a) Eagle Station olivines: for tracks longer than 300 μm minimum lengths only were measured (shaded portions) (see text). b) Marjalahti olivines: complete recordable lengths. For the sake of comparison average recordable track length for Kr ions (E=10.35 MeV/amu) is also shown (Price et al.[35]).

of tracks of lengths in the range 40-160 μm due to an experiment artefact due to geometric conditions: shorter tracks have indeed a lower probability to intersect cracks or "host-Xe " tracks by which the etchant penetrates inside the crystal. Therefore we shall only consider track lengths longer than 160 μm.

The main difficulty in deciphering the Fig. 2b histogram is to assign with confidence given track lengths to charge numbers or even track length intervals to charge groups. The reason is the lack of adequate calibrations with accelerated ions of very high Z in the range 40-92. It thus becomes necessary to theoretically estimate the expected lengths of ultraheavy cosmic rays in olivine which was done by using the following two procedures.

Firstly, one may attempt to extrapolate track length -Z relationship for the charge region 26-36, for which there are already good calibrations for olivine [15, 35] to larger Z, on the basis of simple theoretical considerations. For this, we use the Katz and Kobetich model [18]. Our experimental data for muscovite mica irradiated with accelerated Ne, S, Ar, Ti, Cr ions and Fe cosmic rays displayed a very good agreement with the predicted threshold for registration of these ions in mica together with the predicted track lengths. Taking into account the higher threshold for registration in the case of olivine, we deduced the expected recordable track lengths for Cr, Fe, Zn, Ge and Kr ions; the agreement between predictions and observed values was good. In view of this we felt it was appropriate to use the

Katz and Kobetich model[18] for calculating track lengths of nuclei of higher Z. The extrapolation, together with the experimental data for accelerated Fe, Zn, Ge and Kr ions, is shown in Fig. 3. We are fully aware of the drastic extrapolation conditions to very high Z and very high threshold energies for the predicted recordable lengths as they are shown in Fig. 3. At this point in time, however, there seems to be no way of verifying the predictions. Hopefully future calibration experiments, which are badly needed, will settle the question.

The second approach consisted of normalizing the track lengths distribution to the most prominent peaks of the solar abundances (charge groups 48-58 and 76-92) provided that low abundances and gap are found in the distribution which would correspond to the charge regions 62-75 and 83-89. We observe in fact a rough agreement between the smoothly descreasing track lengths spectrum from 160 up to 900 μm (Fig. 2b) and the solar abundances [8]. However, it is not clear that the expected peaks and gaps are significantly resolved in the histogram. Of course, source(s) abundances have to be duly modified to take into account effects of propagation through the interstellar medium which would flatten down the peaks in the neutron magic numbers regions. Moreover the expected effects of 25-30 g/cm^2 shielding of pallasitic material and the differential partial annealing of latent tracks in space throughout the exposure time should produce additional shifting, broadening and overlapping resul-

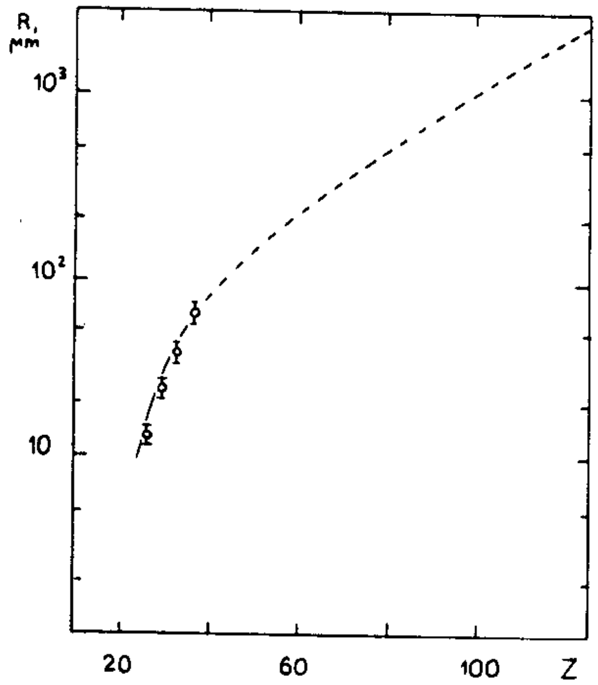


Fig. 3. Total etchable track lengths in olivine as a function of the atomic number Z. Experimental data for accelerated Fe, Zn, Ge and Kr ions are shown. Dotted curve represents the extrapolation based on the Katz and Kobetich model[18].

ting in a rather flat track lengths distribution. Therefore it is not surprising at all to observe only one possible gap in the histogram in the length interval 740-780 μm, which also is not statistically significant. Then for want of something better we shall assign the track length interval 780-900 μm to the longlived nuclei of the uranium group ($Z = 90-96$).

With this assumption then we are led to assign track lengths intervals to approximate charge groups as follows: 160-280 μm to $Z \sim (52 \div 62)$; 280-440 μm to $Z \sim (63 \div 74)$; 440-640 μm to $Z \sim (75 \div 83)$ and 640-740 μm to $Z \sim (75 \div 83)$, the last intervals being ascribed to Pt and Pb peaks broadened by different partial annealings. In view of the various uncertainties and the presently necessary guess work to predict the recordable track lengths for high Z nuclei, we have confined ourselves to discussion of results in terms of broad charge groups only.

Table 2 (column 3) give the abundances of groups of elements with $Z > 52$ relative to Fe group ($20 < Z < 30$) and the related numbers of tracks. They are calculated from Fig. 2b histogram on the basis of the extrapolation based on the Katz and Kobetich /18/ model (Fig. 3). To obtain the abundances various parameters and some corrections were taken into account: i) for partial annealing of tracks (shortening effects are <15% of track lengths); ii) for fragmentation and showing down of VVH nuclei in pallasitic matter (an exponent $\gamma = 2.5$ was assumed for the kinetic energy spectrum); iii) for crystals not previously

TABLE 2

Abundance of cosmic rays with $Z>52$

Charge groups and abundance ratios	Kinetic energy (GeV/amu)	Abundance using Katz and Kobetich model/18/ $\frac{N(Z_i)}{20<Z<30}$	Abundance if track lengths 780–900 μm belong to $90 \leq Z < 96$ $\frac{N(Z_i)}{20<Z<30}$	Solar system abundance/8/ $\frac{N(Z_i)}{20<Z<30}$
$52 < Z < 62$	>1.05	—	0.8×10^{-5} (643)*	2.0×10^{-5}
$57 < Z < 62$	>1.05	2.8×10^{-6} (231)*		3.2×10^{-6}
$63 < Z < 74$	>1.15	8.7×10^{-6} (572)	4.0×10^{-6} (312)	2.0×10^{-6}
$75 < Z < 83$	>1.3	4.6×10^{-6} (292)	3.1×10^{-6} (196)	8.7×10^{-6}
$90 \leq Z < 96$	>1.45	9.7×10^{-7} (64)	1.4×10^{-7} (9)	1.0×10^{-7}
90–96/63–83		7.3×10^{-2} +	2.0×10^{-2}	0.9×10^{-2}
$\frac{90-96}{52-83}$			0.9×10^{-2}	0.3×10^{-2}
$\frac{90-96}{57-83}$		6.2×10^{-2}		0.7×10^{-2}

* Numbers of tracks are in parentheses.

\+ A value of 7.9×10^{-2} for the ratio 90-96/65-83 was found by Price and Shirk /36/ in the Skylab experiment.

irradiated with the xenon beam the probability is lower for shorter tracks (160-280 μm) to be revealed by the etchant pervading fissures or cracks inside the volume. It was estimated from observations that faults and/or slits were homogeneously distributed with spacing of about 300μm. Corrections were also made considering the "edge effects", namely for the fact that the probability for registering a complete track is smaller near the borders: a factor of 1.4 was estimated for this effect.

By using the second approach and after the appropriate corrections mentioned above, we obtain the abundances relative to Fe group as shown in Table 2 (column 4).

By comparing the two sets of data with the solar system abundances [8] we observe some agreements and disagreements. With the second approach, charge groups ~63- ~83 and 90-96 are close to the solar abundances within a factor of 1.4, whereas the estimated abundance of charge group ~52 - ~62 is lowered by a factor of 2.5. Furthermore, the 90-96/~63--~83 and 90-96/~52 - ~83 abundance ratios agree within a factor of 2 to 3 with the solar ratio. By contrast the first approach shows a drastic increase of uranium group nuclei by a factor of 10 compared to solar abundances, in qualitative agreement, however, with recent lexan detector data [35,40]. Charge groups ratios 90-96/63-83 and 90-96/57-83 are also overabundant by a factor of ~ 9 compared to solar abundances. In short the two approaches used give values which are in rather good agreement with the meteoritic abundances in the charge interval ~ 52-83 relative to Fe group; they differ drastically with each other with respect to the abundance of actinide (90-96) nuclei.

Although at this stage we have no convincing argument which allows us to choose between the two sets of data, we are inclined to state that the second approach to be more valid if one accepts that the 9 longest tracks recorded in the range 780-900 μm correspond to the narrow charge group 90-96. With this assumption, and taking into account all approximations and uncertainties, we observe no clear evidence that ultraheavy cosmic ray abundances are widely different from the solar (cosmic) abundances. The abundance ratios of Table 2 (the fourth column) is confirmed would give some support to an acceleration mechanism of electromagnetic nature taking place in a low density plasma of approximate solar composition, as suggested by Casse et al.[10]

The main emphasis in future work we believe should be to verify if the assignment of track lengths in the interval 780-900μm to actinides is justified because of the monitoring implications of the relative abundance to nucleosynthesis, as discussed in recent papers of Blake and Schramm [5], and Schramm [39]. From this point of view, olivines from Eagle Station appear to be the more promising samples for future work.

We may briefly comment on the implications of the present work to the recording of exotic particles like the magnetic monopole. It is possible to predict, on the basis of Katz and Kobetich [18] model, that in olivine a moving monopole of pole strength $g = n(137\, e/2)$ with $n \geq 4$ would produce along its path a large ionisation rate sufficient to lead to very long etchable tracks with no variation of etching rate along their length. Considering the exposure age of Marjalahti we obtain an upper limit of the flux of these monopoles corresponding to 10^{-18} cm^{-2} sec.sr since we did not detect any tracks of length >2-3 mm. This upper limit agrees with previous values of Fleischer et al.[14] obtained on terrestrial micas and obsidians. Recently Price et al.[37] reported on the detection of a magnetic monopole of strength $g = 137e$ with a corresponding abundance of 10^{-13} cm^{-2} sec.sr. Based on our experimental conditions the flux of monopoles with magnetic charge $n \geq 4$ should be at least 10^5 times lower.

In this attempt to determine the abundances of ultraheavy ($Z>50$) cosmic rays in olivine crystals from pallasites we obtained statistically significant data due to the very high collecting power of Marjalahti crystals. About 1200 tracks of $Z \geq 52$ were measured which corresponds to an increase by a factor of 3 of the total number of events registered in plastic and emulsion detectors to date[16,36]. However, some uncertainties, namely lack of adequate calibrations of the olivine detector, partial annealing during space exposure and nuclear interactions within the meteorite, prevent us at present from obtaining abundances for different elements; nevertheless it is possible, with some assumptions, to tentatively determine the abundances for the different charge groups (52-62 and 57-62, 63-74, 75-83, 90-96) relative to Fe group nuclei. Studying Marjalahti olivines was an

exciting prospect because the meteorite sweeped out large regions of the Galaxy and crossed the spiral arm. Therefore relatively large increases of the abundance of actinide elements produced by r -process synthesis may not be unexpected[5,38]. The present data do not exclude a large enrichment by a factor of 10, if the charge group assignment is based on the Katz and Kobetich [18] model. On the other hand, assignment of charge group $90 \leq Z \leq 96$ to tracks of length in the interval of 780-900 μm (the maximum track lengths observed) would correspond to an enrichment of actinide nuclei by a factor of only ~ 2. In this case the relative abundances would be consistent with a solar (cosmic) distribution, within the statistical uncertainties.

The authors are pleased to express their thanks to J.Audouze, M.Casse, G.N.Flerov, D.Lal, J.C.Lorin and H.Reeves for constructive remarks and helpful discussions.

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Received by Publishing Department
on March 23, 1976.