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NEW MEASUREMENT OF THE ^{81}Kr ATMOSPHERIC ABUNDANCE

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ABSTRACT. We have determined the ^{81}Kr activity to be (0.067 ± 0.003) decay/min.l krypton. Using this activity in conjunction with our new measurement of the $^{80}\text{Kr}(n,\gamma)^{81}\text{Kr}$ reaction cross-section of (12 ± 4)b, we infer that the ^{81}Kr activity is 1.5-2.0 times that which would be calculated using the current cosmic ray flux. This implies that the average cosmic ray intensity in the atmosphere during the ^{81}Kr life-time was greater than the current intensity, a fact that has implications for the ^{14}C time scale.

INTRODUCTION

In order to explain ^{14}C results, it is necessary to have reliable data regarding the cosmic ray flux during the last several ten thousand years. There is evidence of a relative constancy of the cosmic ray flux in the solar system during the recent hundreds, thousands and millions of years. These data are obtained mainly from the studies of the different cosmogenic isotopes in meteorites. However, the study of the cosmic ray flux on meteorites gives information about mean flux at the distance of 1-4 a u only. The main part of the cosmic ray flux passing through the earth's atmosphere is subject to the effect of the geomagnetic field. For energies $<10^{11}$eV, therefore, the cosmic ray flux not only differs from the flux in the solar system but may vary significantly with changes in the magnetic field even if the galaxy and solar cosmic ray fluxes are stable.

This paper deals with the cosmic ray flux falling on the earth during the last several hundred thousand years. Detection of the abundance of radioactive ^{81}Kr present in the atmospheric krypton was the basis of the experiment. Although the ^{81}Kr half-life (2.13×10^5y) is much larger than that of ^{14}C, data concerning cosmic ray intensity obtained with ^{81}Kr can be very useful in the identification of ^{14}C variations.

^{81}Kr is produced in the earth's atmosphere by cosmic ray nucleons from stable krypton isotopes by spallation reactions: $^{82}\text{Kr}(p,pn)^{81}\text{Kr}$, $^{83}\text{Kr}(p,p2n)^{81}\text{Kr}$, $^{84}\text{Kr}(p,p3n)^{81}\text{Kr}$ and $^{86}\text{Kr}(p,p5n)^{81}\text{Kr}$, and by secondary thermal neutrons by the reaction, $^{80}\text{Kr}(n,\gamma)^{81}\text{Kr}$. Because of the long half-life and inertia of ^{81}Kr its total quantity stored in the atmosphere during the last hundred thousand years can be known. Almost total ^{81}Kr accumulates in the atmosphere. We can define the average cosmic ray flux intensity over a time approximately equal to ^{81}Kr half-life or more, if we know cross-sections of the spallation and (n,γ) reactions and the ^{81}Kr atmospheric abundance.

It is rather difficult to detect ^{81}Kr decay (electron capture, $E_K^{Br} = 13.475$ keV) because of the presence of ^{85}Kr (β-decay, $E_\beta^{max} = 0.67$ MeV, $T_{1/2} = 10.6$ y) in atmospheric krypton. ^{85}Kr concentration is permanently increased in the postwar period due to nuclear explosions and reactors. There are approximately 10^3 decays of ^{85}Kr per second in one liter of krypton gas today. The expected ^{81}Kr decay rate produced by cosmic rays is nearly equal to 10^{-3}/s.l krypton. Therefore, for the detection of ^{81}Kr produced in the atmosphere by cosmic rays, it is necessary to have old "prebomb" krypton. We used krypton, produced in 1944, which does not contain any noticeable ^{85}Kr.

METHOD

The experiment was carried out in the underground (660m water equivalent) low background laboratory of the Baksan Neutrino Observatory (Kovalchuk and others, 1977). A wall-less cylindrical proportional counter with inner guard proportional counters in anticoincidence with the main counter was used (fig 1). The cathode of the main counter was made of tungsten wires, which simultaneously were cathodes of the guard counters. The outer cathode of the guard counters is a copper tube (fig 1).

The diameter of anode wires is 24μ and that of the cathode wires is 51μ. During the measurement, the main counter operated in anticoincidence with the guard counters. This design permits significant decrease in the inner residual ^{85}Kr background and outer radioactive background. The counter was shielded against surrounding radioactivity with 4cm tungsten, 4cm copper, 6cm lead, and 50cm low-radioactivity concrete (fig 1). The shielding decreases the surrounding γ-ray radioactivity flux by several hundred times. For example, the total counting rate of the Geiger counter ($V = 340cm^3$, $P = 620mmHg$, 90 percent Ar + 10 percent C_3H_8)

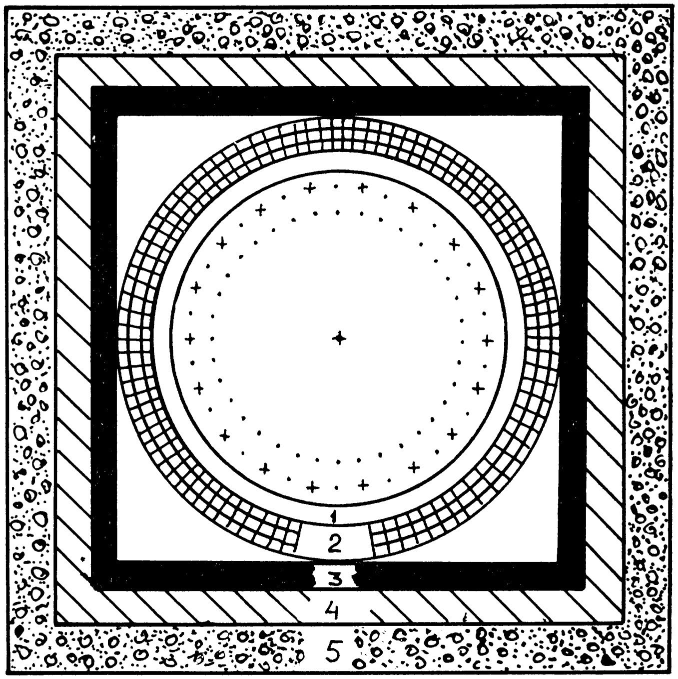


Fig 1. The experimental installation — the proportional counter inside the shield: 1) plexiglass, 2) tungsten, 3) copper, 4) lead, 5) low-radioactivity concrete.

TABLE 1

Parameters of proportional counters

Material of the outer cylinder	Length (mm)	Diameter of the main counter (mm)	Volume of the main counter (cm^3)	Total Volume (cm^3)	Pressure (mm Hg)
I Copper	437	71.5	1754	3350	1047
II Quartz	460	34	417	905	620

at the surface laboratory without shielding is (4.88 ± 0.07)s^{-1} and that at the underground laboratory inside the shield is (7.1 ± 0.2) × 10^{-3}s^{-1}. Parameters of this counter (I) are presented in table 1. The gas mixture of old krypton (82.8 percent), xenon (7.2 percent) and methane (10 percent) was employed. The multiplication factor was near 5000.

There are two calibration windows along the outer guard tube. The calibration was constantly produced with a ^{109}Cd γ-source (88 and 22 keV).

RESULTS

The passive and active shield of the main counter decreases its total counting rate to 0.3 p/min above 4 keV threshold. Total measurement time of the old krypton specimen is 26,625 min. A statistical precision of our experiment is higher than that of former experiments (Loosli and Öeschger, 1969; Barabanov and others, 1973; Barabanov, Golubev, and Pomansky, 1974; Barabanov and Pomansky, 1977). A background at the ^{81}Kr peak (11-16 keV) region can be approximated by a smooth curve passing through points at left and at right of the peak (fig 2), although the intensity of the left points can be partly conditioned by the ^{81}Kr decay. Therefore, the true background curve may lie slightly lower.

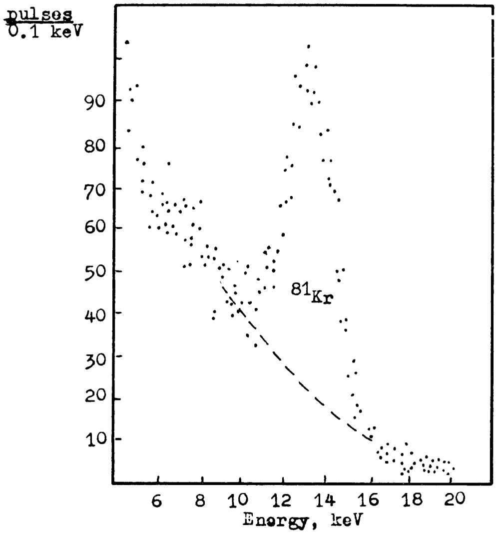
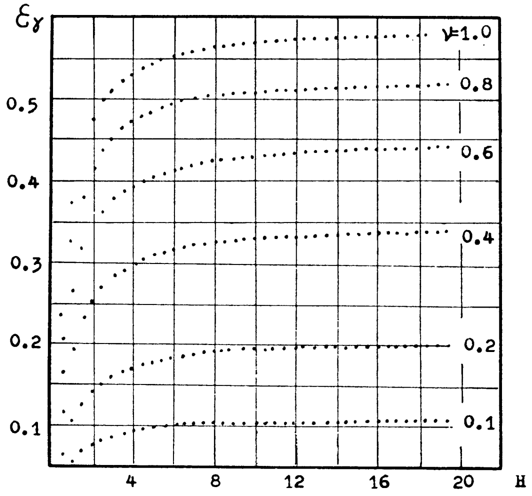
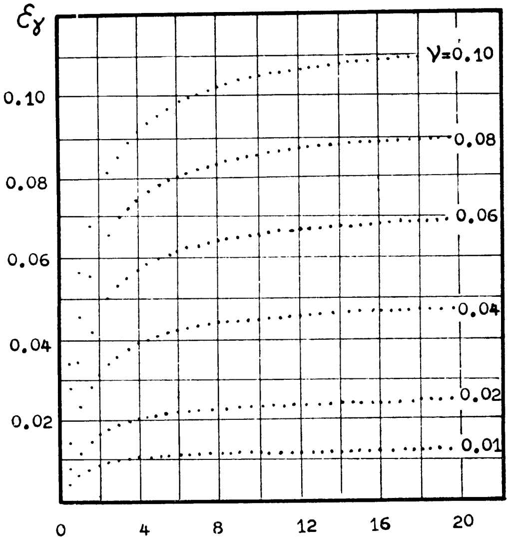


Fig 2. Prewar krypton spectrum during 26,625 min.



A.



B.

Fig 3A. The efficiency of the end to end distributed γ-source registration in the cylindrical detector. H = length/radius, $\nu = \mu \times$ radius μ = linear coefficient of the γ-quantum energy absorbtion in the detector matter.

B. The efficiency of the end to end distributed γ-source registration in a cylindrical detector for other values of H and ν. H = detector length/detector radius, ν = production of μ and detector radius, μ is linear coefficient of the γ-quantum energy absorbtion in the detector matter.

A number of counts over our background in ^{81}Kr peak region is equal to (2064 ± 94).

In order to calculate ^{81}Kr activity, the efficiency of detection of the X-rays with energy 11.9 keV should be known. During K-capture ^{81}Kr releases Auger electrons with an energy of 13.5 keV in 38.6 percent, and X-rays of 11.9 keV plus Auger electrons of 1.6 keV in 61.4 percent of the cases. For an estimate of the detection efficiency of the 11.9 keV X-rays, we used the calculations of Galperin and others (1979). The data of this paper are presented in figure 3, A and B. Parameter H is the ratio of detector length to radius. Parameter ν is the product of the linear coefficient of the γ-quantum energy absorbtion in the detector matter (Storm and Israel, 1967) and radius. $H = 12.2$ and $\nu = 0.625$ for the described counter, and so according to figure 3A, $\epsilon\gamma = 0.445$. Thus the total efficiency of the ^{81}Kr decay registration is $\epsilon = 0.386 + 0.445 \times 0.614 = 0.659$. Taking into consideration this efficiency and data about L/K orbital capture ratio of ^{81}Kr, the total number of counts under the ^{81}Kr peak corresponds to an ^{81}Kr activity of (0.067 ± 0.003) decay/min.l krypton.

In order to calculate the total ^{81}Kr production rate in the atmosphere, the cross-section of the ^{80}Kr(n,γ)^{81}Kr reaction must be known. Former measurements of this cross-section have given the results: (12.5 ± 1.5)b (Reynolds, 1950), (95 ± 15)b (MacNamara and Thode, 1950) and (15.6 ± 1.9)b (Barabanov and others, 1972). We measured this value again. A quantity of krypton, irradiated with reactor thermal neutrons was put into a proportional counter similar to the one described above, but with an outer cylinder of quartz. An outer cathode for the guard counters was produced by tungsten wires strung along the quartz tube. Parameters of this counter (II) are presented in table 1. A gas mixture of 90 percent Ar + 10 percent CH_4 was used.

There is an accumulation of ^{85}Kr during the irradiation of krypton gas with the reactor thermal neutrons due to the reaction ^{84}Kr(n,γ)^{85}Kr. Electrons of its decay give a large background, which makes ^{81}Kr decay registration very difficult. The use of the guard proportional counters decreases this background by a factor of 8 on the range of ^{81}Kr decay (11-16 keV), because a large part of these electrons will pass through both the main and guard counters and are rejected by the anticoincidence design. Results obtained during 8100 min are presented in figure 4. The ^{81}Kr decay peak is seen in the region of 12-15 keV. The spectrum obtained by subtracting of the background under ^{81}Kr peak is presented in the top part of figure 4. An efficiency of the ^{81}Kr decay detection was determined as described above. Parameters H and ν of the quartz counter are: $H = 27$ and $\nu = 0.0825$. The cross-section of the reaction ^{80}Kr(n,γ)^{81}Kr according to our measurement is (12 ± 4)b. A comparison of this result with former values shows that the value (95 ± 15)b appears to be incorrect.

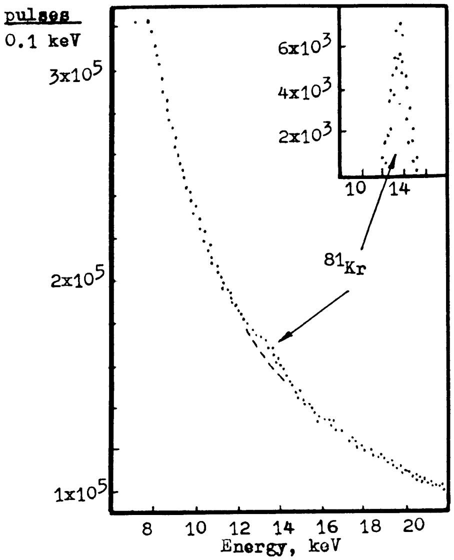


Fig 4. Reactor irradiated krypton spectrum during 8100 min.

DISCUSSION

Calculations show (Öeschger and others, 1970) that the cross-section value (12 ± 4)b leads to an ^{81}Kr atmospheric activity <0.02 decay/min.l krypton due to the ^{80}Kr(n,γ)^{81}Kr reaction. Besides that, ^{81}Kr is produced by spallation reactions which give approximately the same activity. Thus, the total calculated ^{81}Kr activity must be <0.04 decay/min.l krypton. Therefore, the measured ^{81}Kr activity appears to be 1.5 to 2.0 times above the calculated value.

Our measurement of the ^{81}Kr activity (0.067 ± 0.003) decay/min.l krypton is 1.5 times higher than that of Barabanov and others (1973; 1974; 1977) and approximately the same factor lower than that of the Bern University group (Loosli and Öeschger, 1969; Öeschger and others, 1970). We believe that the discrepancy is due to an incorrect estimate of the efficiency of the 11.9 keV x-ray absorbtion in the filling gas of proportional counters in former papers, especially in those of Barabanov and others (1973; 1974; 1977).

The higher atmospheric abundance of ^{81}Kr may be caused by solar cosmic rays produced during large solar flares or supernova flares that may be connected with neighboring pulsars. Both these explanations look rather doubtful. Our opinion is that the ^{81}Kr increase is caused by geomagnetic field variations. It is necessary to make new calculations of the ^{81}Kr production rate in the atmosphere and measurements of the spallation reaction cross-section to confirm the discrepancy between the experiment and the calculation.

REFERENCES

Barabanov, I R, Gavrin, V N, Golubev, A A, and Pomansky, A A, 1973, An intensity of cosmic rays in the atmosphere during the last 10^5-10^6 years and ^{81}Kr content: Izvestija Akad Nauk SSSR, ser Phys no. 6, p 1186-1188.

Barabanov, I R, Golubev, A A, and Pomansky, A A, 1974, Geomagnetic field and content of ^{81}Kr in the atmosphere: Geomagnetizm i Aeronomija, v 14, p 924-926.

Barabanov, I R, and Pomansky, A A, 1977, Atmospheric abundance of ^{81}Kr and cosmic ray intensity, *in* International conference on low-radioactivity measurements and application, The High Tatras, Czechoslovakia: Bratislava, Solvenskè Pedagogické Nakladatel'stvo, p 405-408.

Barabanov, I R, Pomansky, A A, and Rjasny, V G, 1972, A measurement of the cross-section of the reaction ^{80}Kr(n,γ)^{81}Kr: Pisma JETPh, p 643-646.

Galperin, M D, Kuzminov, V V, and Striganov, P A, 1979, Calculations of the efficiency of γ-quanta absorbtion in different materials: Preprint, INR.

Kovalchuk, E L, Kuzminov, V V, Pomansky, A A, and Zatsepin, G T, 1977, Deep underground laboratory for low-radioactivity measurements, *in* Internatl conf on low-radioactivity measurement and application, The High Tatras, Czechoslovakia: Bratislava, Slovenské Pedagogické Nakladatel'stvo, p 23-27.

Loosli, H H, and Öeschger, Hans, 1969, ^{37}Ar, ^{39}Ar and ^{81}Kr in the atmosphere: Earth and Planetary Sci Letters, v 1, p 67-75.

MacNamara, J and Thode, H G, 1950, The relative neutron capture cross-sections of the isotopes of krypton and xenon: Physics Rev, v 80, p 296.

Öeschger, Hans, Houtermans, J, Loosli, H H, and Wahlen, M, 1970, A constancy of cosmic rays following from the study of different isotopes in meteorites and at the earth, *in* Olsson, I U, ed, Radiocarbon variations and absolute chronology, Nobel symposium, 12th, Proc: Stockholm, Almqvist & Wiksel, p 471-487.

Reynolds, J H, 1950, A new long-lived krypton activity: Physics Rev, v 79, p 886.

Storm, E and Israel, H, 1967, Photon cross-sections: Los Alamos Scientific Laboratory rept, June, 1967.