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Abstract

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RADIOISOTOPE ANALYSIS OF ROCKS AND SOILS BY THE ENERGY COMPOSITION OF GAMMA RAYS IN THE NEAR-SURFACE ATMOSPHERE

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1. The energy and angular distribution of γ-rays in space is uniquely determined by the properties of the radiation sources and of the absorbing media. In a certain sense one may say that the γ-field contains information about the emitting and absorbing properties of the medium. This information will be the more complete, the more detailed the data we have on the spectral-angular function of the γ-rays. Data on the energy composition of γ-rays obtained with differential spectrometers have so far been used only for quantitative laboratory determinations in rock samples (1). The present article describes experience in radioisotope analysis of rocks and soils *in situ* from observations of the spectrum of γ-rays in the near-surface atmosphere, both directly at the earth's surface and at a height of several tens of meters (measurements from an aircraft). Such *in situ* analysis may be useful in geological-geochemical investigations of territories.

2. The fundamental possibilities of radioisotope analysis of rocks and soils from the spectrum of γ-rays follow from the fact that among the natural radioactive elements (the uranium and thorium families, radioactive potassium) there are radioisotopes that differ in the energy of the emitted γ-rays.

In observations in the near-surface atmosphere, the following should be borne in mind: the absorbing media—air, rocks, and soils—are formed by elements with small atomic number, and in the γ-ray energy interval 0.1-3.0 MeV their absorbing properties are determined by the variation of the mass density of matter $l = l(z)$, where z is the distance from the plane boundary between the atmosphere and the soil; the emitting and absorbing properties of the media, and consequently the γ-field, remain unchanged during the time of observation; the radiation sources are monoenergetic, and their concentration is $\varepsilon_i = \varepsilon_i(z)$. Under the stated assumptions, where the last of these is not strictly necessary if one operates with mean values of the field over the observation area, the energy composition of the γ-quanta is described by a function of two variables, $n = n(E, z)$.

Since for energies $0.1 < E < 3.0$ MeV and relaxation lengths x up to $3 \div 5$

Fig. 1. Instrumental spectrum of the γ-field over chernozem soils of Ukraine. Flight altitude $h = 25$ m

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the shape of the spectrum of scattered γ-rays depends only weakly on x and on the initial energy E_{0i}, as characteristics of the spectrum at the observation point we obtain $2m$ functions of the type $I_{0i}(z)$ and $I_{\mathrm{sc}\,i}(z)$, which determine, for each of the m lines E_{0i}, the decrease with distance of the energy intensity of the unscattered and scattered γ-rays, respectively.

When $z = z_0$ is fixed, the complete (maximum) amount of information on the energy composition of γ-rays at a given point is limited to $3m$ numbers $(E_{0i}, I_{0i}, I_{\mathrm{sc}\,i};\ i = 1, 2, \dots, m)$. These numbers can be used to identify radioisotopes, to obtain information on changes in their concentration with depth, or to determine the properties of the absorbing media.

Often a given radioisotope emits several monochromatic lines, for example $E_{01}, \dots, E_{0k}$. Since $\alpha_1 \varepsilon_1(z) = \dots = \alpha_k \varepsilon_k(z)$, the functions $I_{01}(z)$, $I_{\mathrm{sc}\,i}(z)$, etc., are not independent. In the indicated case it is possible to obtain more detailed (or more reliable) information on the sought parameters of the source and of the absorbing medium.

The possibilities and specific methods of radioisotope analysis are considered by us using particular examples (see §§ 3 and 4). Here, merely as an illustration, Fig. 1 gives a spectrogram obtained in measurements of the γ-field over soils (see Table 1, section 3) at an aircraft flight altitude of 25 m; similar spectrograms were also obtained in ground observations. In the measurements a scintillation γ-spectrometer was used (number of channels 100, NaJ crystal, 80 mm in diameter and 60 mm high; energy resolution 13%). It should be noted that analogous spectrograms had earlier been obtained by E. V. Shitov with the aid of a scintillation spectrometer of the gray-wedge type.

Fig. 1. Instrumental spectrum of the γ-field over chernozem soils of Ukraine. Flight altitude $h = 25$ m

In the spectrogram of Fig. 1, six peaks are recorded independently, belonging to RaC, RaC$'$, ThC$''$, K^{40}. In the experiments carried out, the concentrations of K^{40}, ThC$''$, and RaC$'$ were determined. The content of K^{40}, apparently, makes it possible to judge with sufficient reliability the total concentration of all potassium isotopes; it may be assumed that ThC$''$ and RaC$'$ are in equilibrium with all decay products in the segments ThX → ThC$''$ and Rn → RaD, respectively. In the remaining parts of the uranium and thorium families, a shift of equilibrium is possible.

The observed depth is comparable with the relaxation length for γ-rays and, for soils and rocks, depending on the energy E_{0i}, amounts to 10–20 cm.

Fig. 2

Figure 2: Fig. 2

3. In ground observations the exposure time was 15-20 min, which ensured good statistics. The method for determining the concentrations of K^{40}, ThC″, and RaC′ from the spectrogram is as follows: a) in four nonoverlapping energy intervals (1.12-1.46; 1.46-1.60; 1.60-2.3 MeV and above 2.3 MeV), as well as in two photopeaks (K^{40} 1.46 MeV; ThC″ 2.62 MeV), the number of recorded pulses is determined; b) the values obtained are used to determine, from six linear equations, the concentrations of K^{40}, ThC″, and RaC′ and three coefficients controlling the change in concentration with depth; c) the coefficients in the equations are determined from measurements of the γ-field over sites with known constant content of radioelements and from calculations of the effect on the detector according to (2).

It has been established that, in the observation areas, the variation with depth of the concentrations of K^{40}, ThC″, and RaC′ does not exceed 10-15%; the final results give the mean values of the concentrations.

The accuracy of determining the concentration *in situ* is 10-12% for K^{40}, 12-15% for ThC″, and 15-25% for RaC′.

The results of ground observations are illustrated by the example of the study of an area with a complex geological structure (Kazakhstan). The geological profile (see Fig. 2) passes through a zone of tectonic contact between acidic (albitophyres) and basic effusives (labrador porphyrites), between which a small intrusion of quartz porphyries was emplaced. The latter are sericitized and carbonatized, locally silicified (point 22). At the contact between the intrusion and the acidic effusives, a thick zone of brecciated and silicified rocks is traced (points 24, 25, 26, 27, 28). Relicts of the enclosing rocks in this zone occur in the mass of quartz; corrosion and replacement by quartz of the rock-forming minerals in fragments of rocks are observed everywhere. According to the field measurements, Fig. 2 gives graphs of changes in the contents of K^{40}, ThC″, and RaC′, the latter

are given in weight percent of the equilibrium amounts of the parent radioisotopes corresponding to them.

Let us note the features in the distribution of radioisotopes in rocks and the reasons which, in our opinion, may explain the spatial changes in the concentrations of these radioelements.

Fig. 2. Distribution of radioisotope contents in a zone of hydrothermally altered rocks:
a —quartz porphyry, *b* —albitophyre, *c* —labradorite porphyrite, *d* —zone of brecciated and silicified rocks, *e* —zone of sericitization and carbonatization and weak silicification

First of all, it should be pointed out that the concentrations of both RaC′ and ThC″, as well as K^40, decrease from acidic to basic effusive rocks, which agrees with known data on the distribution of radioelements in magmatic complexes; changes in the contents of individual radioisotopes in the rock varieties considered do not exceed 10–15%.

Further, in weakly hydrothermally altered quartz porphyries of a small intrusion, an increase in the contents of all three radioisotopes is observed, with substantial (more than 30%) fluctuations in concentration, which is apparently connected (a) with the generally higher Clarke values of Ra, Th, and K in quartz porphyries of small intrusions, noted for this area of work; (b) with the influence of postmagmatic processes on the redistribution of the indicated isotopes, as is evidenced, for example, by the slight introduction of uranium during sericitization and carbonatization processes (the presence of small amounts of the uranium-bearing secondary mineral sericite of carbonate composition), as well as the removal of RaC′, ThC″, K^40 during silicification (point 22).

The process of silicification is also associated with a sharp decrease in the contents of all three radioisotopes in the zone of brecciated and silicified rocks (points 24, 25, 26, 27, 28).

Of considerable interest is the fact that the ratio ThC″/RaC′ for weakly altered albitophyres fluctuates only slightly (from 7 to 9), which corresponds to the average value of this ratio for unaltered rock varieties of similar composition in the area. At the same time, in the zone of brecciation and intense silicification this ratio changes substantially (from 6.2 to 11.8), which is probably connected with the redistribution of radioisotopes during the process of silicification. Let us also note that in the silicification zones the ratio ThC″/K^40 is almost 4 times lower than in quartz porphyries and albitophyres, which is due to the greater migration capacity of potassium. Such zones of intense rock alteration, apparently, can be detected from changes in potassium content.

The analysis carried out does not claim to be complete, but it indicates the possibility of using the information obtained to elucidate individual geological-geochemical processes in the investigated areas.

4. On the basis of the results of aircraft observations, we determined the concentrations of K^40 and ThC″ in the soils of some regions of the European part of the USSR. In the calculations it was assumed that K^40 is distributed uniformly with depth in the soil; the change with depth of the ThC″ concentration was monitored by the value of the ratio of the photopic area $E = 2.62$ MeV to the number of pulses for γ-rays with energy greater than 2.3 MeV. With the exposure we used—20 min—and a flight altitude of 25–35 m, the accuracy of the concentrations found for K^40 and ThC″ is estimated at 15 and 20%, respectively.

The results obtained for several soil types are given in Table 1,

where the content of ThC″ is given in units of Th in equilibrium with it, and K^{40}

for the sum of all potassium isotopes. The data characterize the average content of the elements in the near-surface horizons of soil sections and are calculated in grams per gram of all substances contained in the upper soil layer.

Table 1

No. of sections	Soil type	Geographic position of the survey route	Content, wt. % ThC″ $\cdot 10^{-4}$	Content, wt. % K^{40}
1	Light-gray and gray forest podzolized	Area of the city of Kazan	5.0	1.6
2	Sod weakly and moderately podzolic	Sosnovskoye village, 60 km SW of the city of Gor’ kii; Krasnaya Gorbatka village, 60 km SE of the city of Kovrov	4.0	0.8
3	Southern chernozems, low-humus, medium-thick (solonetzic)	Ukrainian SSR, Kakhovka city—Bug estuary, 30 km south of the city of Nikolaev	7.0	1.5
4	Dark-chestnut	Ukrainian SSR, Bug estuary, 30 km south of the city of Nikolaev —Koblevo village (Tiligul estuary)	7.2	1.6

Table 2

Rocks	α	β
I. Fresh varieties		
Coarse-grained granites; Variscan age	0.85–1.1	0.75–1.0
Aplitic granites; Variscan age	0.85–0.95	0.8–1.0
Vein aplites; Variscan age	0.80–1.0	0.8–1.0
II. Hydrothermally altered varieties		
Albitophyres, weakly sericitized; Devonian	0.95–1.0	0.90–0.95
Quartz porphyries, sericitized, carbonatized, and weakly silicified; Devonian	0.80–0.95	0.50–1.0

Comparison of the potassium data in Table 1 for dark-chestnut soil with the results of work ([3], Table 29) showed satisfactory agreement. The obtained concentration of ThC$''$ for the Kazan area agrees with the data of ([4]).

5. Let us present information on the ratio of the concentrations ThC$''$/ThX $= \alpha$ and RaC$'$/Ra $= \beta$ (the contents of ThX and Ra were obtained by radiochemical analysis of samples; ThC$''$ and RaC$'$ by γ-spectrometry *in situ*). In Table 2, α and β for equilibrium states are taken as unity.

The value of α, both for fresh and for altered varieties of rocks, is close to unity, since the deviations obtained lie within the limits of observational errors. At the same time, the value β, while remaining close to unity for fresh rocks, is characterized by considerable fluctuations for hydrothermally altered quartz porphyries. The latter is probably connected with the increased emanation of these rock varieties. Thus, in fresh varieties of igneous rocks in the area of the work, equilibrium in the chains ThX $\rightarrow$ ThC$''$ and Ra $\rightarrow$ RaC$'$ practically exists.

In conclusion, we note that the ratio of the concentration of K^{40} (γ-spectrometry *in situ*) to the total potassium content (flame photometry of samples) for 50 samples of igneous and sedimentary rocks gives no deviation greater than $±$8%, which is within the limits of measurement errors.

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Note: Figure translations are in progress. See original paper for figures.