A method of determining the content of α-emitting plutonium isotopes according to the characteristic $L_x$ radiation line emitted from uranium in samples taken from the environment is proposed. Comparing the method described and the standard radiochemical determination of plutonium isotopes for soil samples taken from a 5-km zone around the Chernobyl nuclear power plant showed that the results are in good agreement with one another. The concentration of plutonium and americium in the zones with high contamination with transuranium nuclides is measured. It is established that the migration rate of americium is the same as that of plutonium. It is shown that about 90% of the α-emitting nuclides are located at depths of 5–10 cm.

The standard determination of the content of plutonium isotopes in samples requires a long and costly radiochemical procedure. This greatly limits the study of the behavior of plutonium in the environment, where it is necessary to perform measurements on a large number of samples with approximately the same and known ratio of its isotopes. This problem arises in, for example, the study of the plutonium and americium contamination in the 30-km zone around the Chernobyl nuclear power plant [1].

In the present paper, methods for studying α-emitting $^{238–240}$Pu and $^{241}$Am according to the characteristic and γ radiation are described and the results of an investigation performed by these methods of certain sections of a 30-km zone around the Chernobyl nuclear power plant are presented. The content of plutonium isotopes is determined on the basis of the characteristic $L_x$ emission of uranium, using semiconductor spectrometers with a beryllium window.

Two sections with the most intense fallout of radionuclides were chosen for these investigations. On one section (a pine forest in the closest 5-km zone around the Chernobyl nuclear power plant), the radionuclide concentration was so high that all trees in it died. This section is labeled on topographical maps of the exclusion zone as “faded red-brown colored forest.” The second section was chosen near Lake Glubokoe, located in the region of the northern track of the radioactive emissions. The sections were chosen on the basis of the fact that they contain almost all types of soils characteristic for this territory.

It is known [2] that the low-energy states of uranium isotopes with energies 44.51 and 45 keV, respectively, are excited as a result of the α-decay of $^{238–240}$Pu with a probability of about 25%. These energies are less than the binding energy 116 keV of the K electrons in uranium. These states decay primarily by internal conversion of γ radiation on the L-shell electrons followed by the emission of the characteristic x-ray radiation with energy in the range 13–23 keV. Figure 1 shows a
fragment of the spectrum of a soil sample from a section of the pine forest in the 5-km zone, measured using a 1 cm³ detector consisting of ultrapure germanium and possessing a beryllium window and resolution of 300 eV on the 17 keV line.

The high resolution of the detector and the use of the “instrument line” method [3], where this line is measured using the same detector, have made it possible to separate with a degree of reliability (~5%) the $L$ group of neptunium, accompanying the decay of $^{241}\text{Am}$, and the $L$ group of uranium, accompanying the $\alpha$-decay of the plutonium isotopes. The probability that the low-energy state will be filled and the fluorescence yield coefficients of the x-ray radiation are well known. Using calibration sources, it is also possible to measure directly the yield of the $L_\alpha$ and $L_\beta$ groups of the plutonium isotopes. The computed and measured yields of the $L_\alpha$ and $L_\beta$ groups of uranium and neptunium showed that the intensity of the x-ray radiation of uranium per decay is $I(L_\alpha) = (3.9 \pm 0.2)\%$ and $I(L_\beta) = (5.2 \pm 0.2)\%$.

Thus, having measured the intensity of the x-ray radiation, the activity of the plutonium isotopes in the sample can be measured. If the measurements are performed only according to the yield of $L_\alpha$ radiation, then the sample must be thin so that the self-absorption of the x-ray radiation can be neglected. However, thin samples, a high-resolution detector, and the correspondingly low efficiency of the detection of $\gamma$ rays increase the measurement time. If a large number of identical samples must be analyzed, then the speed of the method described can be increased substantially by using thick samples and detecting $^{241}\text{Am}$ $\gamma$-rays. To account for self-absorption of the low-energy x-ray spectrum in “thick” samples correctly, it should be kept in mind that the samples investigated contain, as a rule, a substantial amount of $^{137}\text{Cs}$, whose decay results in the excitation of the 661 keV isomeric state of $^{137}\text{Ba}$ and subsequent emission of the characteristic 32–37 keV $K_\alpha$ radiation of barium. The self-absorption coefficient for $L_\alpha$ radiation of uranium can be calculated from measurements of the intensity of the x-ray $K_\alpha$ emission of uranium and the accompanying high-energy 661 keV $\gamma$ radiation, for which self-absorption is unimportant. The obtained self-absorption coefficient is used to determine the activity of plutonium isotopes from measurements of the intensity of the x-ray $L_\alpha$ emission of uranium.

It is known that the $\alpha$-decay of plutonium present in the plutonium from power production is accompanied by the accumulation of $^{241}\text{Am}$ which decays with the emission of a 59.4 keV $\gamma$-ray. The ratio of the intensity of this transition and the probability of the x-ray $L_\alpha$ emission of neptunium has been measured with an error of 2–3%. As one can see from Fig. 1, the shift of the $L_\alpha$ emission of uranium relative to neptunium is ~0.5 keV, so that its self-absorption can be treated as negligible. Thus, the activity of plutonium with a substantial decrease of the self-absorption fraction can be calculated from the ratios $L_\alpha(\text{U})/L_\alpha(\text{Np})$ and $L_\alpha(\text{Np})/59.4$ keV. The measurements performed with thin and thick samples yielded a correction for self-absorption for 59.4 keV with an error not exceeding 5%. It was found to be $f = [A(662)/A(32)]^{0.25}$, where $A$ is the activity of $^{137}\text{Cs}$ measured on the 662 and 32 keV lines, for all types of soils.

On this basis, large-volume detectors with a thin beryllium window can also be used effectively. Figure 2 shows a fragment of the spectrum of the same soil sample as in Fig. 1, measured with a 150 cm³ detector consisting of ultrapure germanium.