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Radioactive Pollution in Lublin Region, April—May 1986

Skażenie promieniotwórcze okolic Lublina, kwiecień—maj 1986

Радиоактивные загрязнения в районе г. Люблин в апреле—мае 1986

1. INTRODUCTION

On April 28th 1986 in the laboratories of Institute an increase of the background in radiation counters was observed. During the next days the background of standard radiometers RUST-3 increased by factor 10-20 comparing with its normal level. That background rise proved to be the result of disaster in the nuclear power plant at Chernobyl (Ukraine).

To collect some information about the character of pollution we began the measurements with semiconductor detectors. At that time the Department of Nuclear Physics had to its disposal in Lublin one Ge(HP) probe with very good energy resolution, but of small active volume, thus of

low efficiency of gamma radiation detection. To speed up the data collection an older Ge(Li) detector 40 cm^3 was put into action, its resolution was a bit below the standards.

The gamma spectra of air and soil were observed. The spectra were registered using 4096-channel ICA-70 analyser and then stored on floppy discs in the data processing station PSPD-90, connected directly with the analyser. The relative abundance of various isotopes manifesting themselves in the gamma spectra was determined by measuring the area under selected, well resolved, gamma peaks, applying the correction for the detector efficiency and extrapolating back to the April 26th, 1986, 1^{00} GMT. Calibration procedure was performed with a set of standard sources. For easier identification of gamma lines the tables of isotopes [1] and tables of gamma spectra [2] were used. The positron annihilation line 511 keV seen in the spectra is the laboratory background: at the distance 2m from the detector there was ^{22}Na source (~$4x10^5$ Bq) placed in another experimental setup. The source was kept on that place, because the 511 keV line could serve in this situation as a local standard of intensity. The abundance of ^{106}Rh was determined by the intensity of 512 keV transition. Thus to avoid the systematic error in that case, the ^{22}Na annihilation sources were removed from the measurement room.

First measurement with germanium detectors was performed on April 30th.

2. MEASUREMENTS and RESULTS

a. Air pollution

We measured the air radioactivity, but not the radiactivity of aerosols in air, thus no sucking and filtering devices were used. The spectra were measured with the Ge(Li) detector without any point sources, in a closed compartment. The result is shown in Fig.1. In this conditions volatile fission products dominated in the spectra, i.e. ^{131}J, ^{132}J, ^{132}Te, also the gamma peaks of

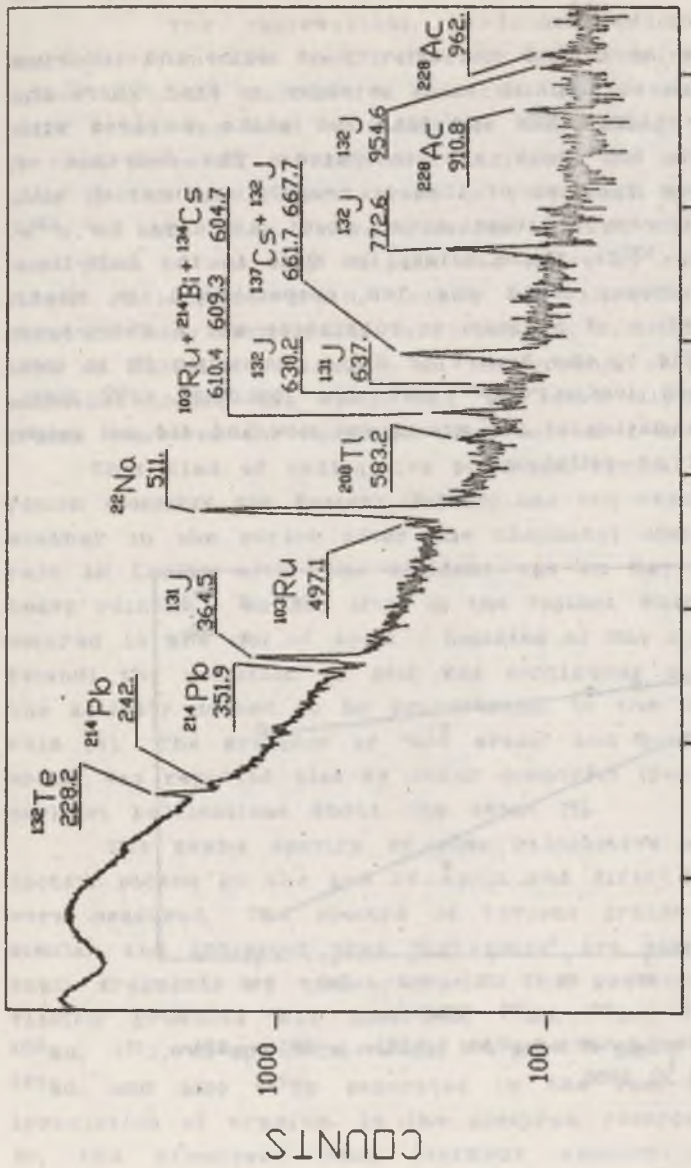

Fig.1. Gamma spectrum of air measured on April 30, 1986 by using Ge(Li) detector

^{134}Cs, ^{137}Cs, ^{103}Ru were seen. At longer measurement time weak peaks of natural radioactive elements ^{214}Pb, ^{208}Tl, ^{228}Ac became visible.

Since April 30th the activity of iodine and tellurium in air began to diminish as it is shown in Fig.2, where the intensities of 364.5 keV and 228.2 keV peaks compared with constant 511 keV peak are indicated. The decrease of activity as a function of time is roughly exponential, with the half-decrease constant equal about 9.9 days for ^{131}J and 64h for ^{132}Te. These values lie close to the half-lives of these isotopes (8.04d and 78h respectively). It means that the inflow of radioactive substances was a short-term one (according to the Report of State Commission [3] it took place between 1986.04.27 0^{00} GMT and 1986.04.27 12^{00} GMT), and the translation of air masses was slow and did not cause the removal of pollutions.

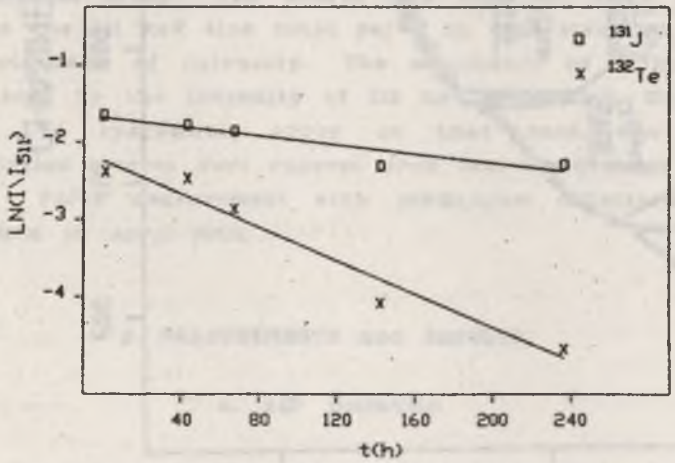

Fig.2. The activity of iodine and tellurium in air, time–zero April 30, 1986.

b. Soil surface pollution

The observations, performed using standard radiometers on the open, relatively smooth surfaces, have shown that the radioactive pollution is predominantly of point type. The head of radiometer moved along the surface of roof, pavement, parking area indicated the activity level close to the background, except of very small areas spaced 1-2 m to each other. Taking a sample of dust, sand etc. from that place it was easy to pick and select by elimination a single radioactive grain, usually one thenth of mm in diameter. Many such grains were found in Lublin and in the area of Lublin voivodship - on the ground, on the leaves, sometimes inside the flats. At the zero distance those grains delivered the radiation doses between 1 and 25 mR/h.

This kind of radioactive pollution, typical for Lublin region (possibly for Eastern Poland) one can explain by dry weather in the period after the Chernobyl accident. First rain in Lublin after the accident was on May 10th, first heavy rainfall - on May 12th. In the regions, where the rain occured in the end of April - begining of May (e.g. Southern Poland) the pollution of soil was continuous and uniform, the activity seemed to be proportional to the intensity of rain [4]. The presence of "hot areas" and point-like "hot spots" was reported also by other countries (Sweden) in the earliest publications about the event [5].

The gamma spectra of some radioactive grains ("hot spots"), picked in the end of April and first days of May were measured. The spectra of various grains were very similar and indicated that "hot spots" are essentially the small fragments of nuclear fuel. The presence of many fission products was observed: ^{95}Nb, ^{95}Zr, ^{99}Mo, ^{103}Ru, ^{106}Ru, ^{131}J, ^{132}J, ^{132}Te, ^{133}Xe, ^{140}Ba, ^{140}La, ^{141}Ce, ^{144}Ce, ^{147}Nd, and also ^{239}Np generated in the fuel by neutron irradiation of uranium. In the spectrum recorded on April 30, the strongest peak (without application of the correction for counter efficiency) belonged to the ^{239}Np activity, disappearing fast with the half-life 56.3h.

Typical gamma spectrum of "hot spot" measured on May 3th, 1986 is shown in Fig.3a and 3b, while the spectrum of the same particle recorded on May 28-29 is shown in Fig.4abc. In the spectrum in Fig.3. the group of peaks ^{239}Np-^{239}Pu in the vicinity of 100 keV is still well visible, while on the Fig.4. only the long-lived species are present, particulairly the peaks of ^{134}Cs (half-life 2.06y) and ^{137}Cs (half-life 30.2y) become discernible above the background. The gamma lines of ^{214}Bi and ^{40}K seen in Fig.4. represent the natural contribution. No gamma lines of ^{51}Cr were observed, contrary to the results given in [5]. Relative content of gamma emitters in two selected "hot particles" is shown in Table 1.

Table 1. Percentage activity of gamma emitters in selected "hot particles".

Nuclide	Particle 1	Particle 2
^{95}Nb	6.2	
^{95}Zr	6.4	
^{99}Mo	6.2	
^{103}Ru	5.3	78.9
^{106}Ru	1.1	21.1
^{131}J	0.7	
^{132}J	5.1	
^{132}Te	4.9	
^{134}Cs	—	
^{137}Cs	—	
^{140}Ba	7.0	
^{140}La	8.0	
^{141}Ce	8.3	
^{144}Ce	5.9	
^{147}Nd	2.4	
^{239}Np	32.5	

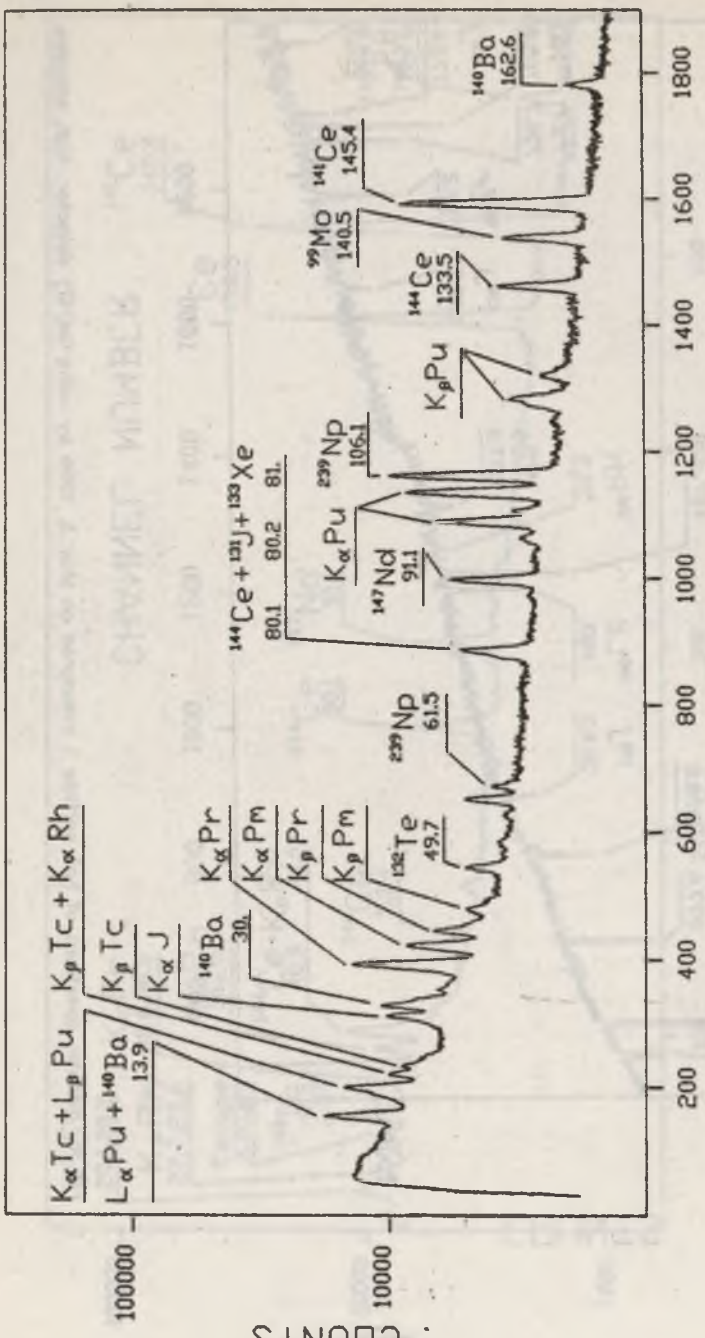

Fig.3a. Gamma spectrum of hot particle 1 measured on May 3, 1986 by using Ge(HP) detector, low energies.

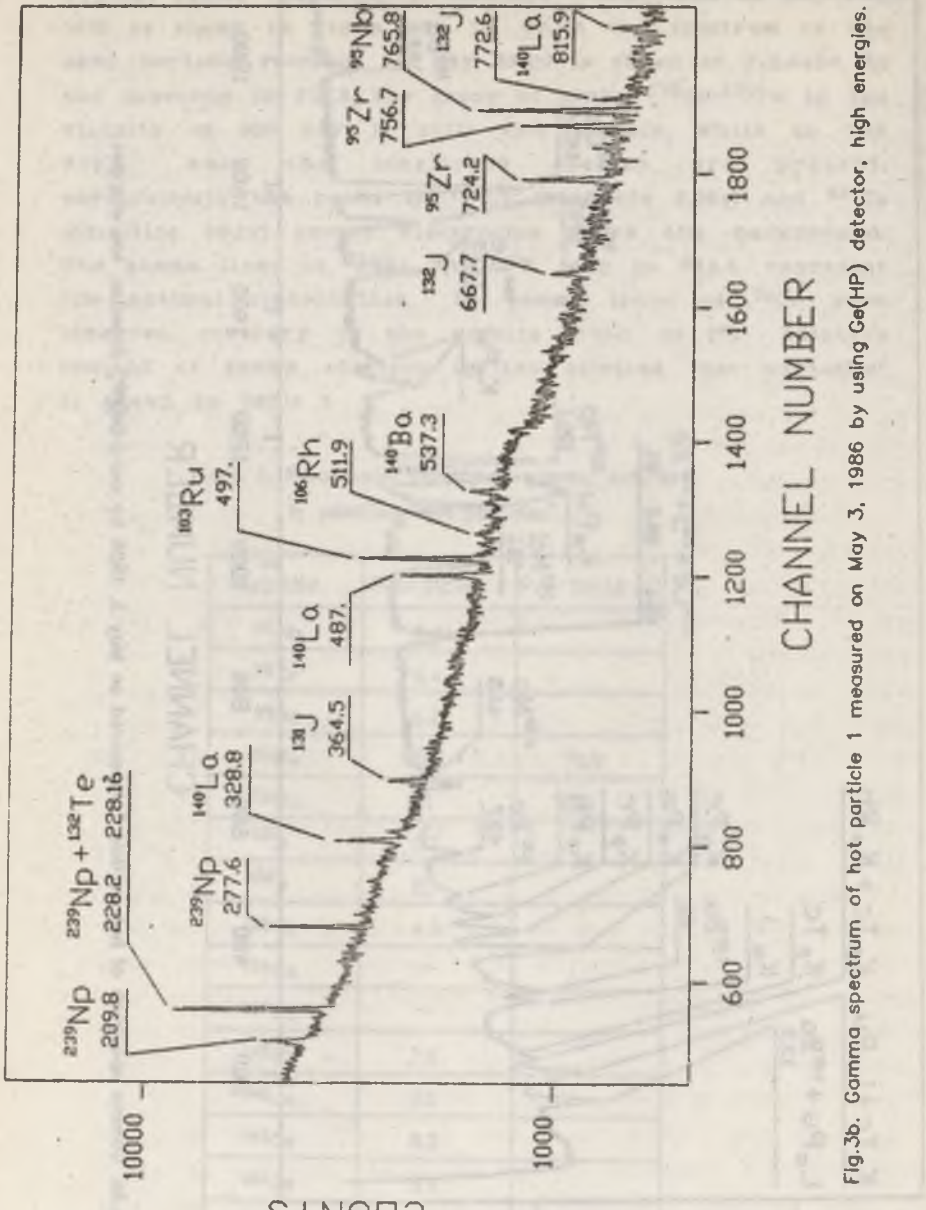

Fig.3b. Gamma spectrum of hot particle 1 measured on May 3, 1986 by using Ge(HP) detector, high energies.

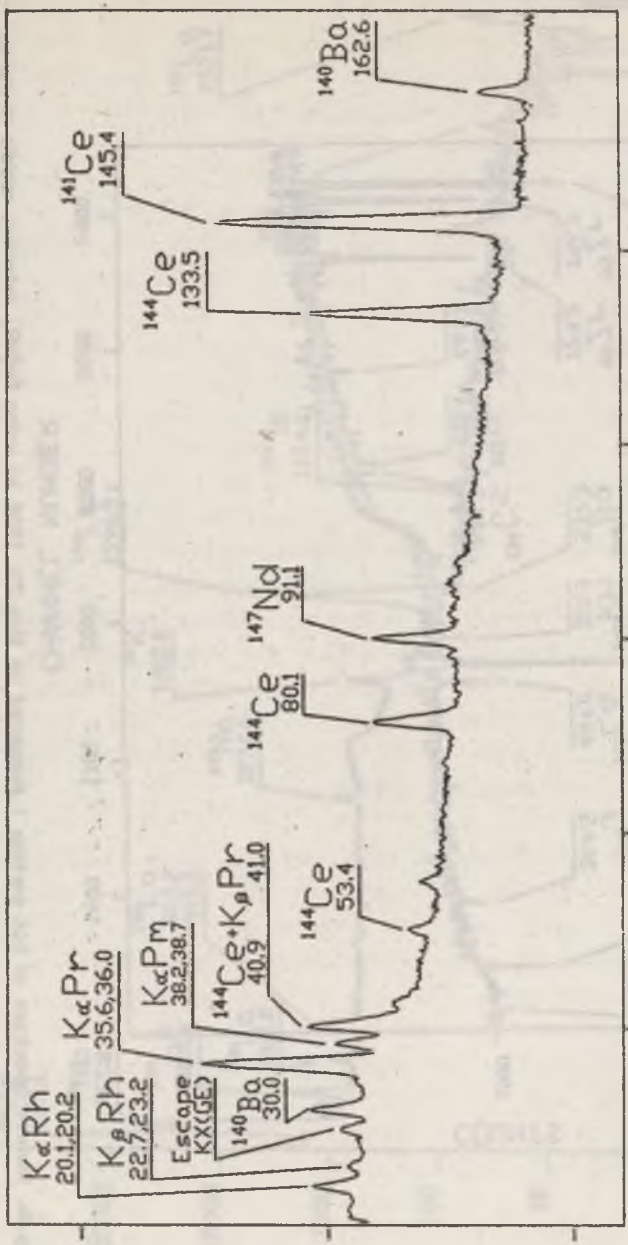

Fig.4a. Gamma spectrum of hot particle 1 measured on May 29, 1986 by using Ge(HP) detector, low energies.

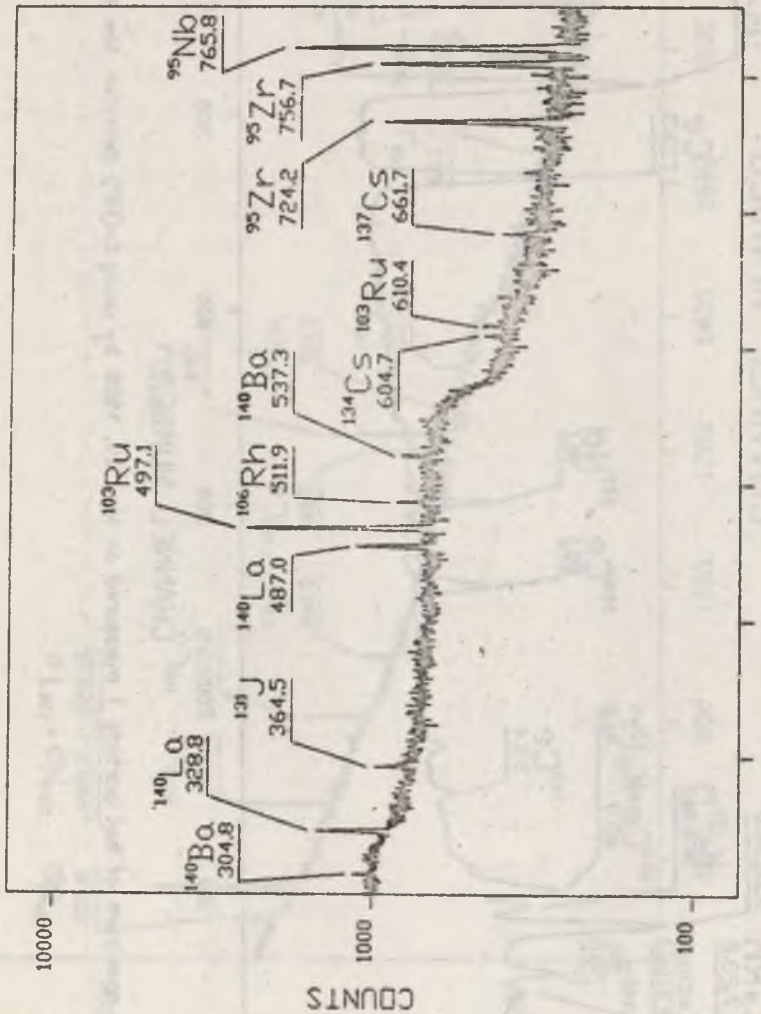

Fig.4b. Gamma spectrum of hot particle 1 measured on May 29, 1986 by using Ge(HP) detector, middle energies.

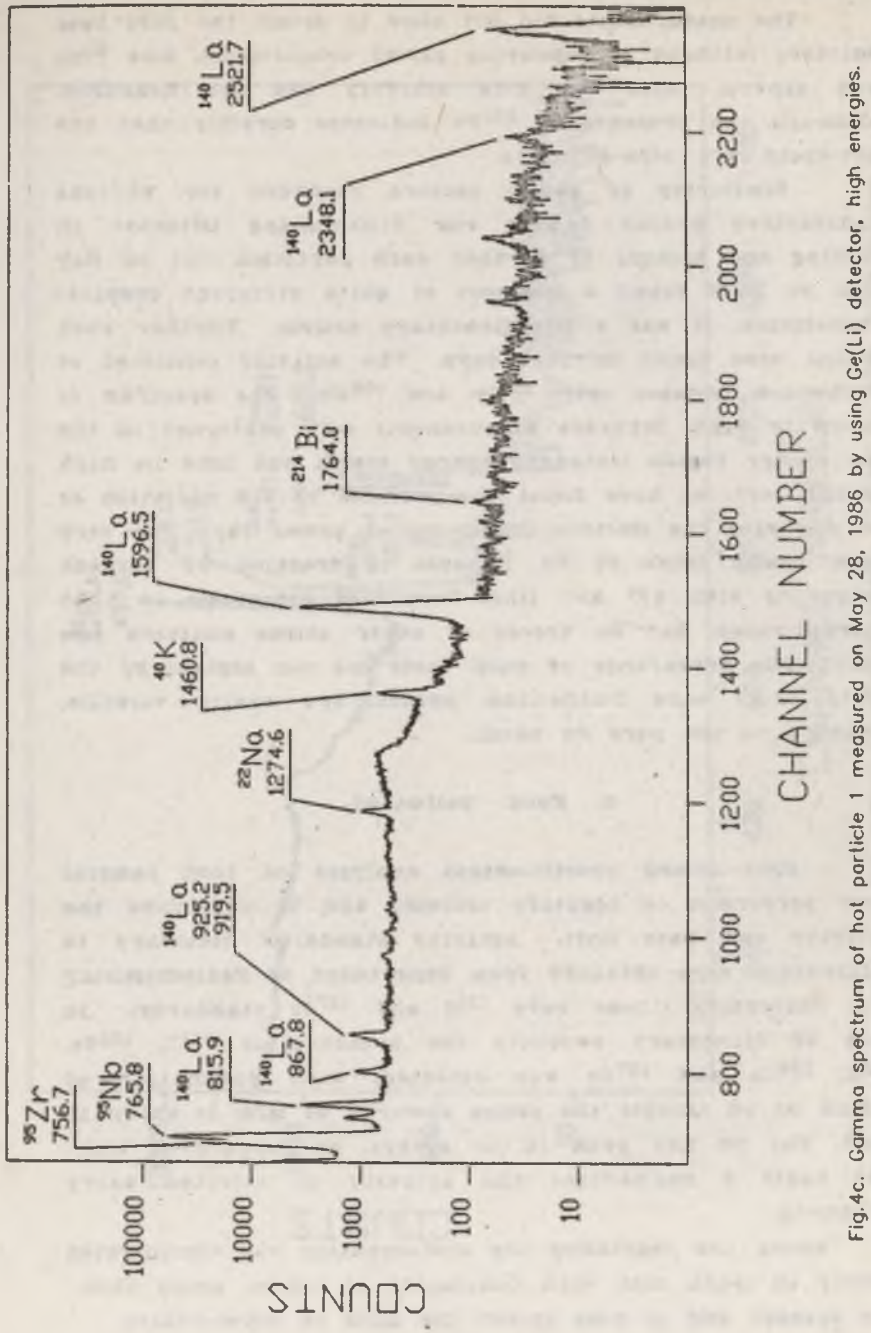

Fig.4c. Gamma spectrum of hot particle 1 measured on May 28, 1986 by using Ge(Li) detector, high energies.

The measurements did not allow to detect the pure beta emitters (without accompanying gamma transitions), like ^{90}Sr and others. Also the alfa activity was not measured, although the presence of ^{239}Pu indicates directly that the hot-spots are alfa-emitters.

Similarity of gamma spectra recorded for various radioactive grains caused our diminishing interest in seeking and picking of further such particles, but on May 12th we have found a hot-spot of quite different chemical composition. It was a monoelementary sample. Further such grains were found in next days. The activity consisted of ruthenium isotopes only: ^{103}Ru and ^{106}Ru. The spectrum is shown in Fig.5. Separate measurements were performed in the low energy region (extended energy scale), but like in high energy part, we have found the presence of K-X radiation of Rh following the electron conversion of gamma rays. The very weak gamma lines of Ru isotopes (a fraction of percent comparing with 497 keV line) were also detectable in that energy range, but no traces of other gamma emitters (see Fig.6.). The appearance of such spots one can explain by the facts that some ruthenium oxides are easily volatile, contrary to the pure Ru metal.

c. Food pollution

Some gamma spectrometric analyses of food samples were performed to identify isotopes and to determine the activity per mass unit. Activity standards necessary to calibration were obtained from Department of Radiochemistry MCS University (those were ^{131}J and ^{137}Cs standards). In most of alimentary products the presence of ^{131}J, ^{132}Te, ^{132}J, ^{134}Cs and ^{137}Cs was detected, with domination of iodine. As an example the gamma spectrum of milk is shown in Fig.7. The 511 keV peak is, as always, an "apparatus line". The table 2 summarizes the activity of selected dairy products.

Among the vegetables the contamination was concentrated mainly in leafs, also with domination of iodine, among them the spinach and to some extent the leafs of horse-radish

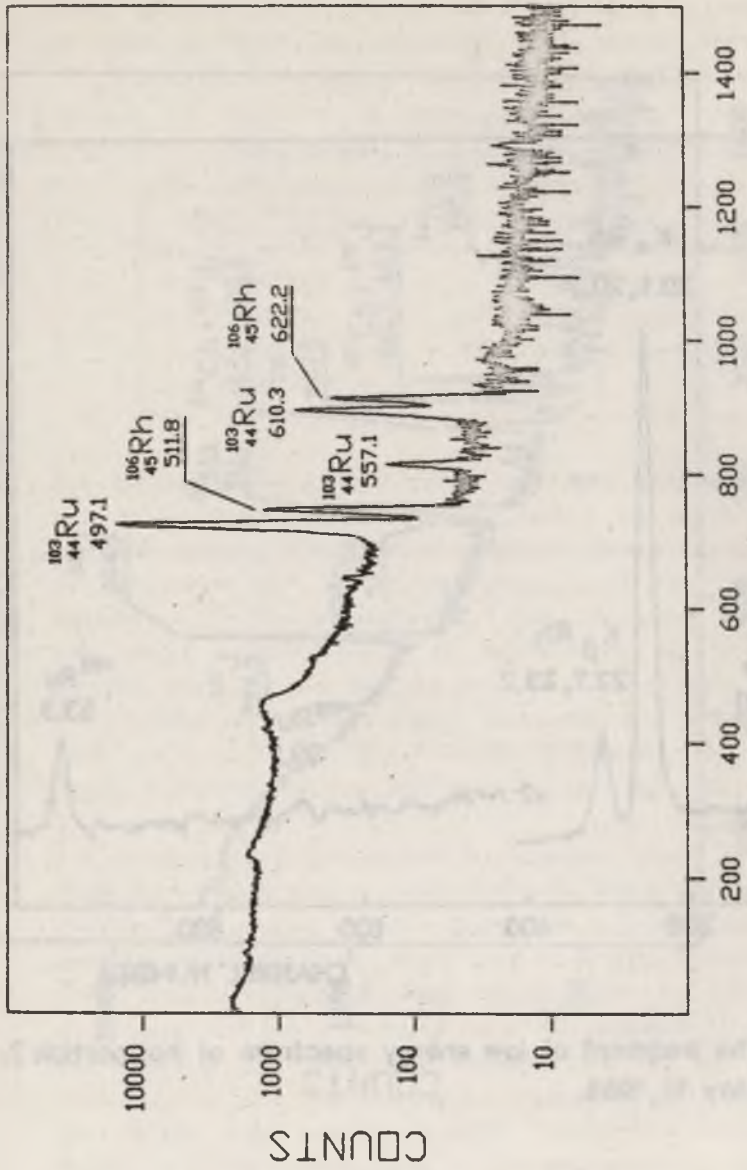

Fig.5. Gamma spectrum of hot particle 2 measured on May 12, 1986 by using Ge(Li) detector.

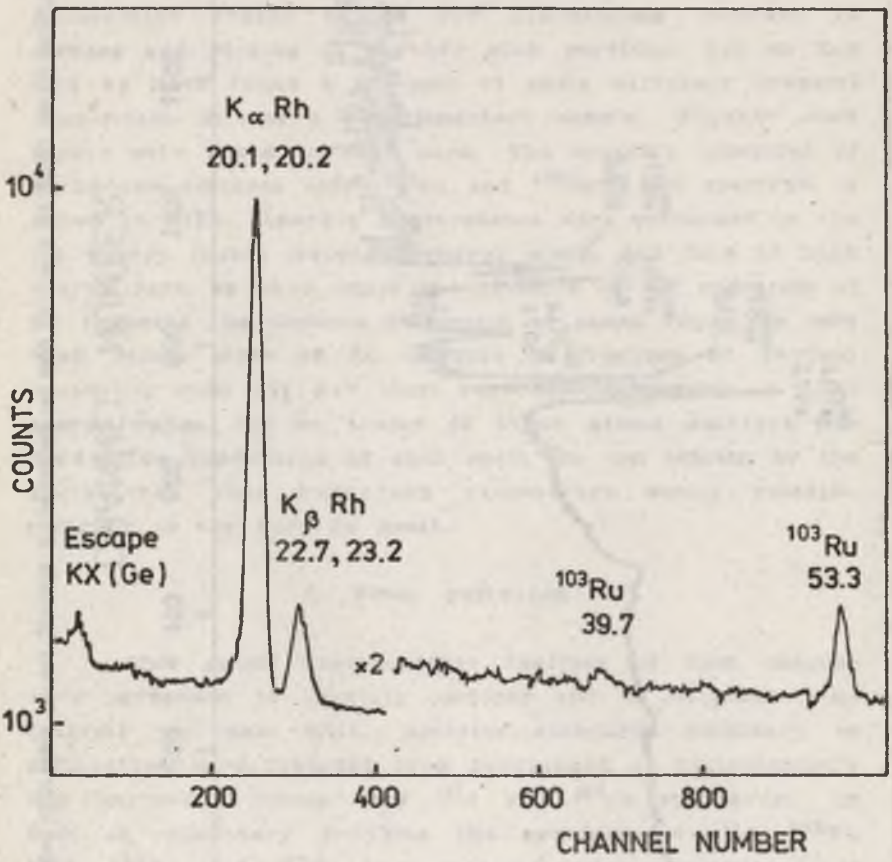

Fig.6. The fragment of low energy spectrum of hot particle 2, May 12, 1986.

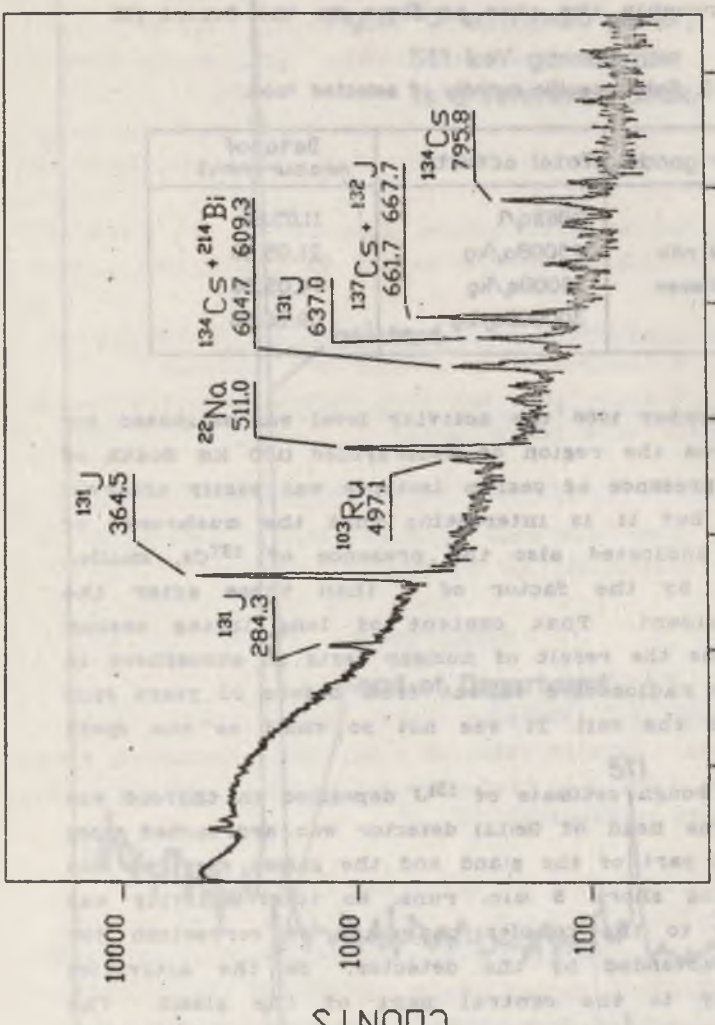

Fig.7. Gamma spectrum of milk measured on May 11, 1986.

stand out for its rich spectrum of radioactive isotopes. The content of radioactive substances in fruits and roots was decidedly smaller than in leafs. The measured level of activity is roughly the same as given in the Report [3].

Table 2. Total specific activity of selected food.

Consumer goods	Total activity	Data of measurement
Milk	500Bq/l	11.05.86
Powdered milk	2300Bq/kg	21.05.86
Cottage cheese	1000Bq/kg	14.05.86
Spinach	30000Bq/kg	09.05.86

In September 1986 the activity level was estimated for mushrooms from the region of Zwierzyniec (100 km South of Lublin). The presence of cesium isotopes was easily observed (^{134}Cs, ^{137}Cs), but it is interesting that the mushrooms of 1985 vintage indicated also the presence of ^{137}Cs, smaller approximately by the factor of 3 than these after the Chernobyl accident. That content of long living cesium isotope is thus the result of nuclear tests in atmosphere in the past. The radioactive fallout from before 20 years still is present in the soil. It was not so small as one could suppose [6].

A very rough estimate of ^{131}J deposited in thyroid was undertaken. The head of Ge(Li) detector was approached close to the central part of the gland and the gamma spectrum was collected during short, 5 min. runs. No total activity was estimated due to the complex character of correction for solid angle subtended by the detector. So the activities concern mainly to the central part of the gland. The measurements were performed approximately one month after the accident (28-29 May). Typical values were 100-120 Bq (for unknown reasons one of us has shown 5 times smaller ^{131}J contamination than all other persons, in spite of no administration of iodine preparates; see Fig.8.).

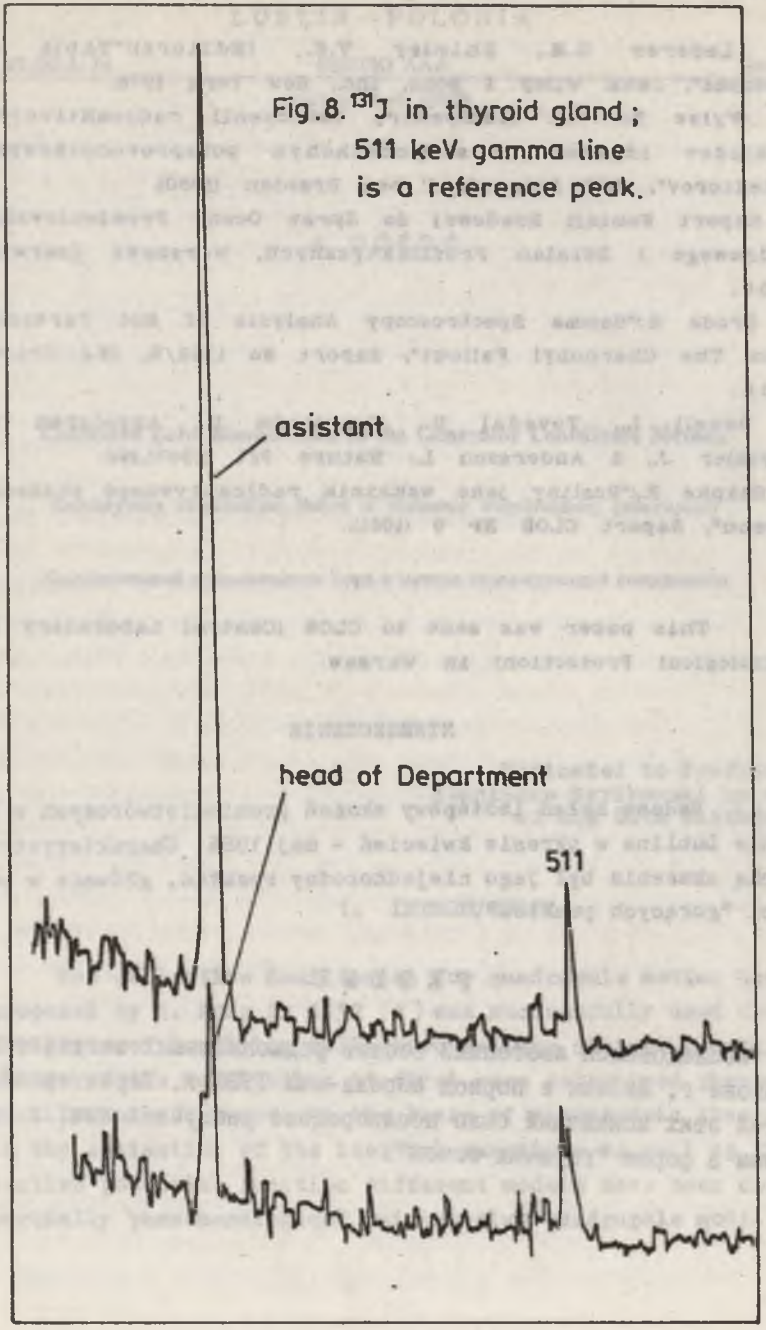

Fig.8. ^{131}J in thyroid gland; 511 keV gamma line is a reference peak.

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This paper was sent to CLOR (Central Laboratory of Radiological Protection) in Warsaw.

STRESZCZENIE

Badano skład izotopowy skażeń promieniotwórczych w rejonie Lublina w okresie kwiecień - maj 1986. Charakterystyczną cechą skażenia był jego niejednorodny rozkład, głównie w postaci "gorących punktów".

РЕЗЮМЕ

Исследовался изотопный состав радиоактивных загрязнений в районе г. Люблин в период апрель-май 1986 г. Характерной чертой этих искажений было неоднородное распределение, в основном в форме "горячих точек".