GROUND DEPOSITION OF LONG-LIVED GAMMA EMITTERS IN POLAND FROM THE CHERNOBYL ACCIDENT

By K. Królas, M. Kubala and T. Ściężor

Institute of Physics, Jagellonian University, Cracow*

(Received December 22, 1986)

Activity composition was measured for soil contaminated with the fallout from the Chernobyl accident. The soil samples were collected at various areas of Poland. A map showing the ^{137}Cs deposit distribution was drawn for the most contaminated southern part of Poland.

PACS numbers: 28.90.+i

1. Introduction

The radioactive emission from the Chernobyl accident was spread over large areas located up to thousands of kilometers from the place of the event. Soon after the accident it appeared that the fallout deposit was rather nonuniform and strongly depended on meteorological conditions at the period of the radionuclide release. First maps summarizing ground-level radiation measurements were completed and published in some countries in May, 1986 [1, 2]. Long-term exposure and transfer of radioactive substances in the human body is determined by the amount of long-lived isotopes deposited on the ground. The information on the level and distribution of ^{137}Cs deposit is particularly important since ^{137}Cs has a long half-time ($T_{1/2} = 30.2$ y) and it enters vegetables and other plants via the root-uptake.

In the present work the measurements of the ground-level deposits were performed for several isotopes. Samples of soil were collected in many locations in Poland and we are able to complete a map of ground contamination. At the time of measurement, a few months after the Chernobyl accident, the bulk of short-lived isotopes was beyond the detection level but the activity of the long-lived ones remained significant. From among about 20 isotopes observed soon after the accident [3], we could get information on the ground-level distribution of 7 long-lived isotopes decaying with gamma ray emission.

* Address: Instytut Fizyki, Uniwersytet Jagielloński, Reymonta 4, 30-059 Kraków, Poland.

2. *Measurements and analysis*

Samples of soil were collected in about 150 locations in Poland by a few students during their holidays in July and August. As a rule, the soil was taken from open and flat areas preferably not covered by plants. A 1 cm thick upper layer of soil was cut from a surface of about 50 cm^2, corresponding to about 150 g of the soil. From every location 1 to 5 samples were taken and at some places the soil was taken additionally at different depths.

For ^{137}Cs activity determination a $3'' \times 3''$ NaJ(Tl) scintillator with 11% energy resolution was applied. Soil samples were placed in a standard container and were measured for at least 100 minutes each. The absolute efficiency calibration was performed using the non-radioactive soil sample contaminated with a ^{137}Cs source of known activity.

Later, 40 selected samples were examined using a 50 ccm Ge(Li) detector with energy resolution 4.5 keV for 662 keV. Each spectrum was recorded for 16 hours. A typical example of the obtained gamma spectrum is displayed in Fig. 1. Gamma lines of 4 isotopes:

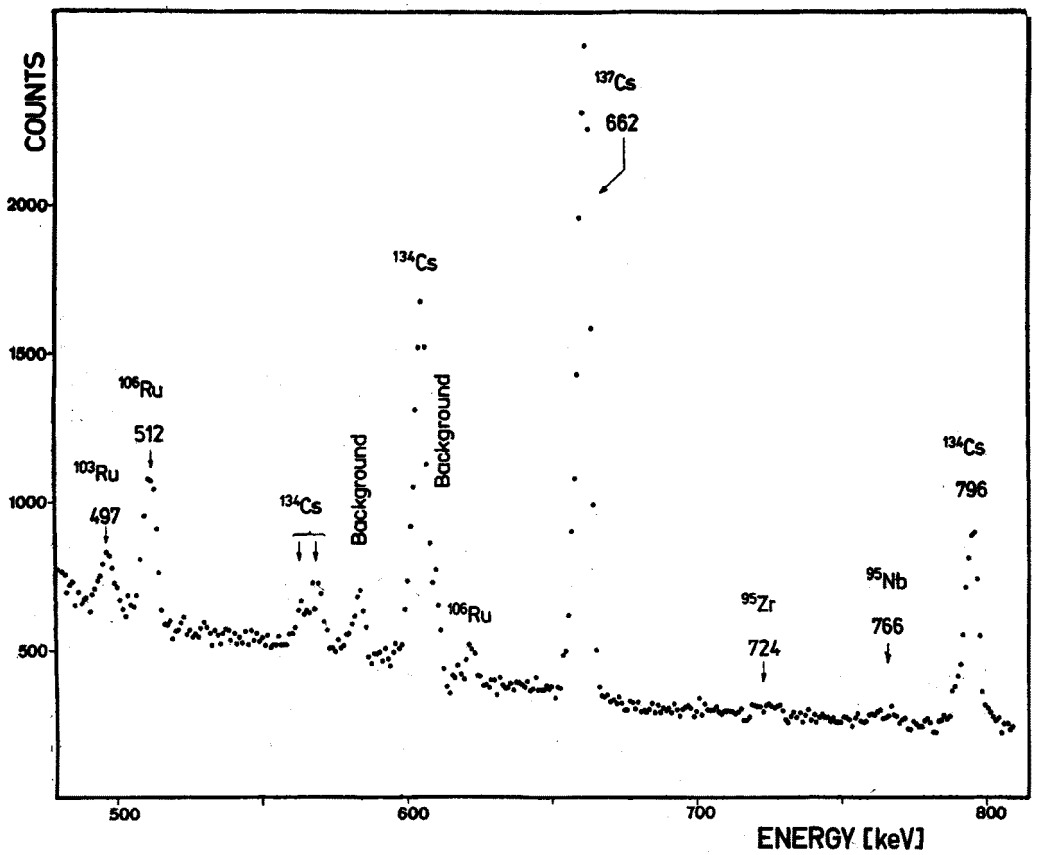

Fig. 1. Gamma spectrum of the soil samples taken at Skawina. The activity values were determined basing on intensities of the lines denoted with the gamma ray energy

^{103}Ru, ^{106}Ru, ^{134}Cs and ^{137}Cs are very well seen. Besides, for 7 samples, gamma rays of ^{95}Zr, ^{95}Nb and ^{144}Ce were also observed. For activity determination the following gamma lines were used: 497 keV for ^{103}Ru, 512 keV for ^{106}Ru, 662 keV for ^{137}Cs, 724 keV for ^{95}Zr, 766 keV for ^{95}Nb and 796 keV for ^{134}Cs. They are indicated in Fig. 1.

As indicated above the efficiency callibration takes into account selfsorption of ^{137}Cs 662 keV gamma rays. The effect for other lines used is estimated to be similar within 3%. The only exception is the 134 keV line of ^{144}Ce for which the absorption is very serious. Although we clearly observe a small activity of ^{144}Ce the meaningful activity value could not be determined.

The measured activity was corrected for decay which took place since May 1 to the time of measurement. The obtained activity values for 12 samples coming from different regions of Poland are listed in Table I.

3. Results and discussion

^{134}Cs($T_{1/2}$ = 2.06 y) and ^{137}Cs($T_{1/2}$ = 30.2 y)

The most complete data were obtained for ^{137}Cs activity and its distribution over different areas in Poland. The ^{137}Cs content in the upper layer of soil varied in a broad range from 0.05 Bq/g to 2.55 Bq/g. It appears that the most important factor was a rainfall which deposited activity on the ground at the end of April. Fig. 2 shows the activity of ^{137}Cs in the soil taken from areas with dry and wet fallout depositions. The activity of ^{137}Cs is relatively low and it varies from 0.05 Bq/g to 0.21 Bq/g for all the 16 locations at which no rain occurred from the April 28 to the May 2 [4] i.e. in the time of radioactive cloud passage above Poland [5]. On the contrary, the ground level activity due to the wet deposition, as a rule, is rather high. From among 150 samples, 14 samples were collected at the places where the stations of the State Hydro-Meteorological Institute registered a rainfall during 24 hours since 7 a.m. on April 30 to 7 a.m. on May 1 ranging from 0.5 mm to 10 mm. The activity of ^{137}Cs for these locations is drawn in Fig. 2 as a function of the rainfall amount. The general tendency is an increase in activity with increasing rainfall. Obviously, the rainfall was the very efficient means of depositing radioactive isotopes on the ground.

All results of the activity measurements may be used for constructing a map of the ground level activity of ^{137}Cs in Poland. In view of a rather limited number of the examined locations two additional indications were taken into account. First, a closer inspection of the maps of the rainfall [4] helped us to determine the borders of the rainfall zones and therefore, we believe, the borders of highly contaminated areas. Secondly, some information was obtained from the first rough measurements of total ground deposition in early May. An example of such measurements is presented in Fig. 3 which shows the ground level contamination measured on May 2 at successive railway stations along the railway from Cracow to the east direction.

Fig. 4 shows a map of southern Poland only because here the radioactive deposits were the highest. In the northern and the central part of Poland the ^{137}Cs activity did not

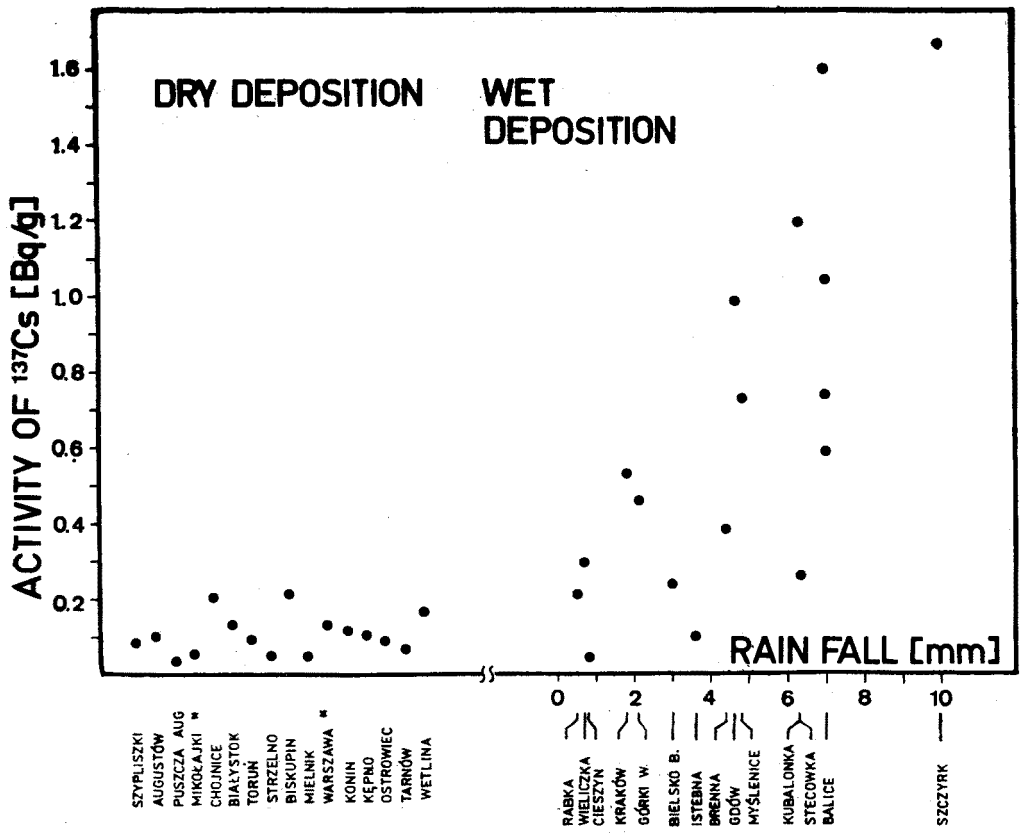

Fig. 2. The ^{137}Cs activity in soil samples from different locations in Poland. First part: the locations with dry fallout deposition ordered with respect to decreasing geographical altitude. *Data from Ref. [6]. Second part: activity against amount of the rain falling since 7 a.m. on April 30 to 7 a.m. on May 1 [4]. The average activity value is drawn for each location except Balice (rainfall 7 mm) for which 4 points denote activities of 4 different samples to show activity value irregularity

exceed 0.22 Bq/g and the average value was equal to 0.12 Bq/g. Thus we found that about 80% of the area of Poland was rather weakly contaminated with ^{137}Cs and that most of it was deposited in the South.

In southern Poland the activity is rather nonuniform, relatively high, and reaches up to 2.55 Bq/g at the Jałowiecka Saddle (East from Żywiec). Two characteristic elongated zones of the highest activity extent to the south-west from the points situated 30 km East and 10 km West from Cracow. Isolated areas of the high activity were also found in the Tatra and the Bieszczady Mountains. In view of the limited number of sampling points it is not certain whether all heavily contaminated areas were found out and are included in the map. Nevertheless, we believe that the map depicts the real situation since the meteorological stations did not report heavy rainfall at noninvestigated places. The average activity of ^{137}Cs in the South of Poland was estimated as equal to 0.4 Bq/g.

Of course, the activity of ^{137}Cs is always accompanied by the activity of ^{134}Cs. Within

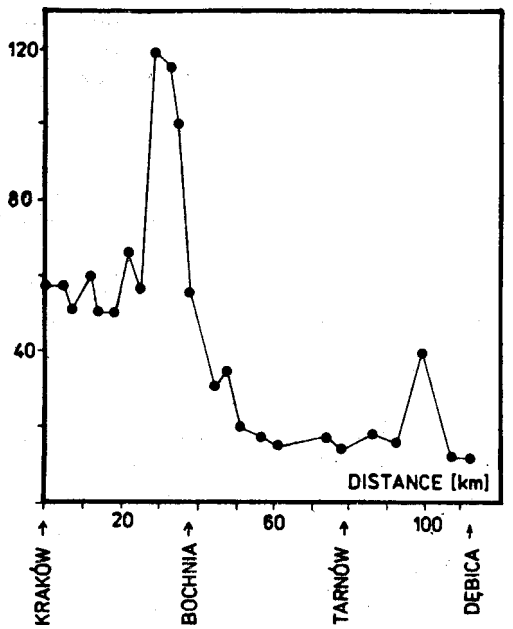

Fig. 3. Total ground activity measured on May 2 at successive railway station along the railway from Cracow into the eastern direction. (Arbitrary units)

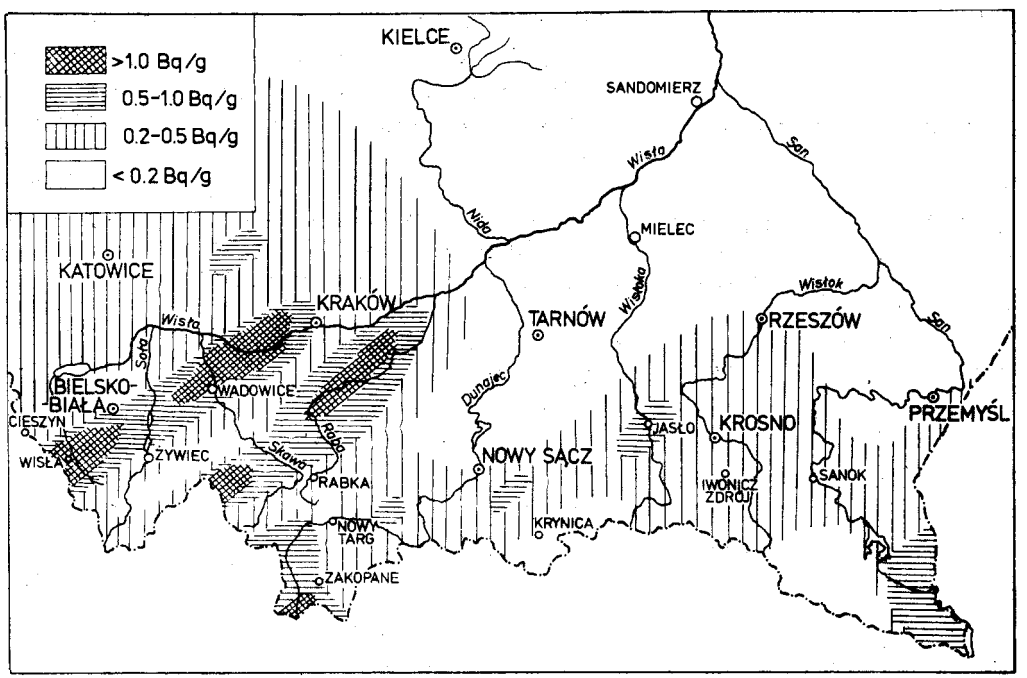

Fig. 4. Map of ^{137}Cs activity in upper layer of soil in Southern Poland. Activity for the whole remaining area of Poland is lower than 0.2 Bq/g or, in limited areas, it exceeds 0.2 Bq/g a bit

limits of statistical error the activity ratio $^{134}Cs/^{137}Cs$ was the same for each sample. The average value equaled 0.57(2) for activities extrapolated to May 1. Consequently, the map presented in Fig. 4 is also valid for the ^{134}Cs distribution with appropriate changes in the activity scale.

$^{103}Ru(T_{1/2} = 39.4\ d)$ and $^{106}Ru(T_{1/2} = 368\ d)$

Activity of ruthenium isotopes was observed for all 40 samples investigated with the Ge(Li)-detector. The determined value of isotopic ratio $^{106}Ru/^{103}Ru$ equaled 0.21(3) on May 1.

The distribution of the ruthenium isotopes did not differ significantly from that for the caesium isotopes. The initial activity ratio $^{103}Ru/^{137}Cs$ ranged from 1.6(1) to 2.6(1), but it was similar for neighbouring sampling points. We have not found a common feature for these locations where the ground deposit was enriched with the ruthenium isotopes with respect to ^{137}Cs. Anyway, ^{103}Ru and ^{106}Ru always accompanied volatile isotopes like caesium and some short-lived isotopes of tellurium and iodine. This suggests that ruthenium, with its high melting point, was released from the reactor in its volatile oxide form.

$^{95}Zr(T_{1/2} = 64\ d)$, $^{95}Nb(T_{1/2} = 35\ d)$ and $^{144}Ce(T_{1/2} = 285\ d)$

The gamma rays emitted in ^{95}Zr, ^{95}Nb and ^{144}Ce decays were detected in all samples collected in East and North-East Poland but in 2 samples only from southern Poland.

The ^{95}Zr activity extrapolated to the "May 1" value ranged from a trace amount to 0.7 Bq/g. As it was already mentioned the ^{144}Ce activity could not be determined due to strong selfabsorption of the 134 keV line in the samples. The ^{95}Nb is a daughter product of ^{95}Zr decay and independently of the starting values of their activities the $^{95}Nb/^{95}Zr$ activity ratio of 2.2 is practically reached after a few months of their decays [7]. Indeed, a similar activity ratio was observed in our samples. The initial activities of ^{95}Nb and ^{144}Ce, however, could be suggested basing on the constant ratio $^{95}Nb/^{95}Zr$ and $^{144}Ce/^{95}Zr$

TABLE I

Radioactive composition in the upper layer of soil taken at 12 different locations in July and August. Activity values in [Bq/g] are extrapolated to May 1, 1986. Statistical error equals about 5% while the activity for two samples taken at the same location differed up to 50%

Location	^{137}Cs	^{134}Cs	^{103}Ru	^{106}Ru	^{95}Zr
Puszcza Aug.	0.05	0.03	0.1	0.03	0.4
Szypliszki	0.08	0.03	0.1	0.05	0.6
Białystok	0.12	0.07	0.1	0.02	0.3
Przemyśl	0.15	0.11	0.4	0.08	—
Kielce	0.18	0.10	0.43	0.10	—
Gliwice	0.23	0.13	0.52	0.08	—
Kraków	0.45	0.26	0.92	0.18	—
Mszana D.	0.47	0.26	0.76	0.12	—
Balice	1.15	0.65	1.9	0.46	0.1
Andrychów	1.55	0.88	2.0	0.46	—
Szczyrk	1.65	0.90	2.5	0.58	—
p. Jałowiecka	2.55	1.35	4.0	1.0	—

observed for the hot particles separated from the radioactive fallout following the Chernobyl accident [7]. One can assume that the initial activities of ^{95}Zr, ^{95}Nb and ^{144}Ce were equal with 30% accuracy.

It has to be emphasized that the distribution of the discussed above isotopes is completely different from the ^{137}Cs distribution in Poland. In South, there was practically no activity of ^{95}Zr, ^{95}Nb and ^{144}Ce in contrast to the usually high ground contamination with ceasium and ruthenium isotopes. In the samples taken from North-East of Poland the activity of ^{95}Zr was relatively high while the activity of ^{137}Cs was low (see Table I).

Total fallout deposit

The radioactivity deposited on the ground surface at the end of April penetrates into deep layers of soil with a rate depending on soil properties. Fig. 5 shows the ^{137}Cs activity as a function of depth at which the soil was taken. In November 1986, more than 90%

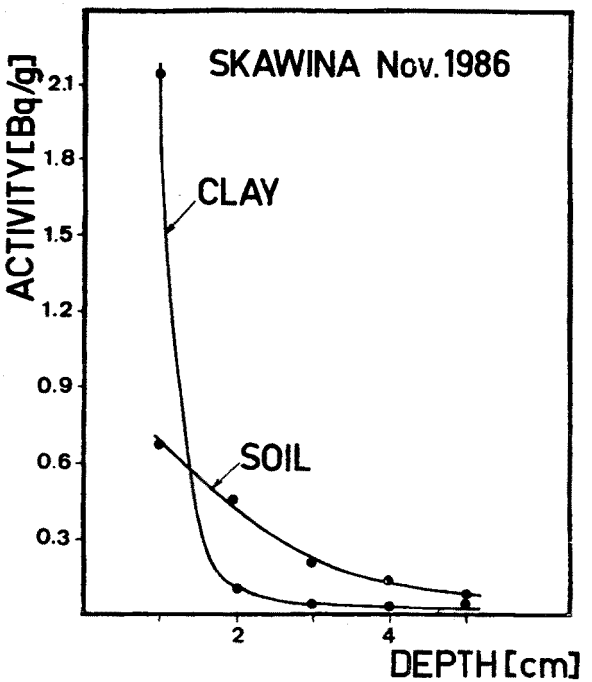

Fig. 5. Activity dependence on depth for two types of soil (clay- and sand-like soil) taken in November, 1986 at Skawina near Cracow

of the total activity stayed in the upper one centimeter thick layer of clay-like soil while 40% of the total activity remained near the surface in the sand-like soil. Much lower penetration was observed for the samples collected in July. It was estimated that, in average, about 70% of the total deposited activity of ^{137}Cs was still in the upper layer of soil collected in July and August to investigate the activity distribution.

The total deposit of ^{137}Cs in Poland was estimated as follows: the average activity

0.12 Bq/g assumed for 80% of the Polish territory corresponds to the ground surface activity $0.12\ \text{Bq/g} \times 30000\ \text{g/m}^2 \times (0.7)^{-1} \approx 5\ \text{kBq/m}^2$.

In the same way, one could obtain the ground surface activity equal to 20 kBq/m^2 for the remaining territory. This gives 2.5×10^{15} Bq of ^{137}Cs as the total deposit all over Poland. It is roughly half of the total deposit of ^{137}Cs in Sweden [8] and about 2 times more than the total activity of ^{137}Cs deposited in this country during the nuclear weapon atmospheric test period [8].

Among 7 identified long-lived gamma emitters, the activities of ^{137}Cs and ^{134}Cs reached the level at which their contributions to the annual radiation doses should be considered. Annual external dose calculated for the most contaminated area (100 kBq/m^2 of ^{137}Cs) is equal to 300 mrad per year assuming 24 hours exposure every day. This value is about 2 times larger than the average annual dose from natural background radiation, however, individual doses depend on how much time people have spent outdoors and will be significantly low. It is much more difficult to estimate the internal dose due to ceasium isotopes which enter body with food. We believe that present determination of the highest activity deposit area will stimulate new studies on ^{137}Cs behaviour in the food-chain and its abundance in the food produced in contaminated zones.

Annual doses from the content of the other identified isotopes are much less. First, their halflives are relatively short and secondly they accumulate neither in plants nor in human body. However, there is an evidence that the activity of ^{95}Zr and ^{144}Ce is often accompanied by alpha emitters [9]. Our results indicate that while the fallout of ceasium isotopes was mostly deposited in Southern Poland the north-eastern-part may be weakly contaminated with some alpha emitters.

The authors are grateful to R. Broda and W. Waluś for many useful discussions and to R. Kmieć for a consent to publish data shown in Fig. 3. Warm thanks to L. Józefowski, T. Włodek and S. Wróbel who helped us to collect samples.

REFERENCES

[1] T. Bennerstedt, M. Holmberg, L. Lindborg, *Chernobyl-fallout, measurements and consequences*, SSI-report 86-13, Stockholm 1986.
[2] C. Hohenemser, D. Deicher, A. Ernst, G. Lindner, E. Recknagel, Environment 28, No 5 (1986).
[3] R. Broda, J. Grębosz, J. Wrzesiński, Report No 1330, Institute of Nuclear Physics, Cracow 1986.
[4] Archive of the State Hydro-Meteorological Institute, Warsaw.
[5] Report of the Government Council, Warsaw, June 1986.
[6] J. Jastrzębski, to be published.
[7] R. Broda, *Acta Phys. Pol.* **B18**, 935 (1987).
[8] J. O. Snihs, *Consequences in Sweden of the Chernobyl accident*, SSI-report 86-21, Stockholm 1986.
[9] R. Broda, Z. Szeglowski, K. Zuber, to be published.