Investigations of a potential enrichment of radionuclides in the environment of the Paks Nuclear Power Plant[1]

Introduction

In our age the monitoring of the contamination effect exerted by large industrial objects cannot be neglected even if this environmental impact is estimated to be at a minimum level. In the course of normal operation of nuclear power plants quantities of radionuclides emitted into the atmosphere usually are so small that their contribution to the natural background radiation, the cosmic radiation etc. is undetectable even in the close surroundings of the power plant.

On the other hand, chronic and long-lasting processes might occur which promote the accumulation of radionuclides emitted during operation by plants-indicators of specific geomorphic facies.

A survey of places of enrichment in isotopes is important not only for revealing chronic contamination but these areas also are expected to increasingly accumulate radionuclides in the case of an accidental fallout.

The above mentioned problems and demands were considered during environmental radiation protection monitoring and geomorphological and geo ecological surveying the start of operation of the Paks Nuclear Power Plant in 1982. In this period a radioactive isotope originating from atmospheric emission and deposited on the soil surface was ^{110m}Ag. This isotope with a half-life of T = 250 d is a by-product of corrosion during neutron activation.

Based on preliminary calculations this radiosilver with an annual release of 1.0–1.5 GBq and in a form of aerosols is deposited on the soil surface at an annual rate of 1 Bq/m^2 in the vicinity of 1–3 km radius. These presumptions became supported by control measurements conducted by the power plant service itself showing concentration of 1 μBq/m^3 of ^{110m}Ag in the atmospheric aerosol within a distance of 1.5 km from the power plant while other radionuclides could not be revealed.

Land form characteristics

The results of the geomorphological investigations and mapping achieved in the environs of the Paks Nuclear Power Plant demonstrate, that the geomorphological environment of the power plant with the radius of 10 km is very various and rich in many different landforms *(Figure 1).*

Geomorphic features of the surroundings of the Paks Nuclear Power Plant with radius of 5 km are determined by fluvial erosional and accumulational landforms. So the immediate impact zone is to be regarded as being of plain relief type *(Figure 2).*

According to the altitudes a.s.l. the area can be subdivided into three levels: among them the lowest is the flood bed with living meanders being permanent change of channel configuration; an intermediate position is occupied by the low flood plain with cut-off meanders in various stages of evolution; the uppermost level is represented by high flood plains of sporadic distribution.

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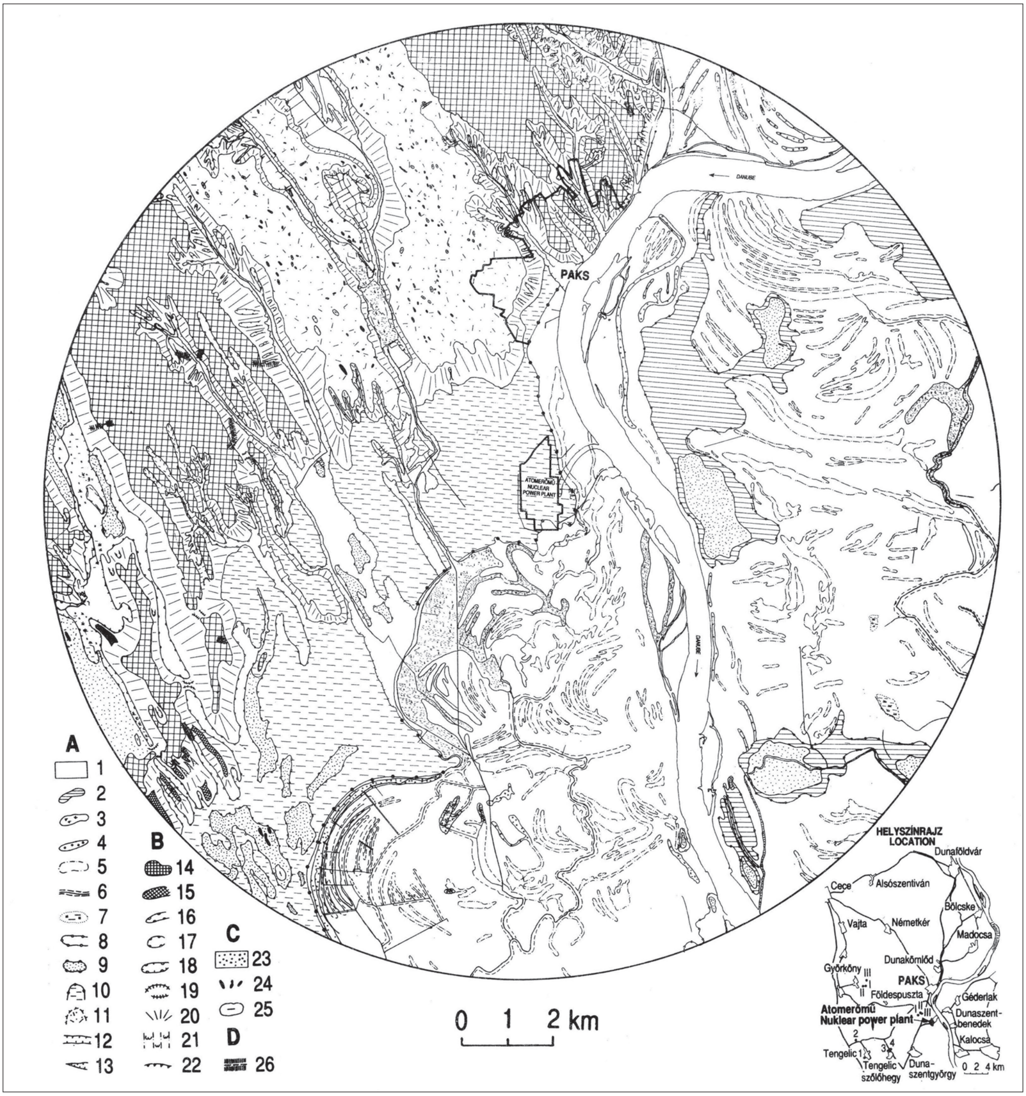

Fig. 1. Geomorphological map of the environs of the Paks Nuclear Power Plant (compiled by Balogh, J. *et al.* 1994). – A = Fluvial, erosional and accumulational landforms: 1 = low flood plain level; 2 = former, cut-off or abandoned meanders; 3 = former, upfilled meanders, intermittently inundated, with water vegetation; 4 = former, cut-off or abandoned meanders in flood-plain forest; 5 = former, upfilled meander, cultivated; 6 = former, upfilled meander, chanellized; 7 = alkali flats, frequently covered by water; 8 = high flood plain; 9 = former terrace island on the flood plain; 10 = fluvial terrace; 11 = flat alluvial fan; 12 = broad and flat erosional valley; 13 = gully; B = Landforms of complex genesis: 14 = loess plateau; 15 = low interfluvial ridge; 16 = derasional dry valley; 17 = derasional niche; 18 = erosional-derasional valley; 19 = derasional col; 20 = slope, undistinguished; 21 = slope with stabilized fossil slump; 22 = unstable bluff; C = Landforms of deflation: 23 = sand blanket; 24 = sand forms (longitudinal dunes, blowout, residual ridges); 25 = deflation hollows; D = Man-made landforms: 26 = sunken road

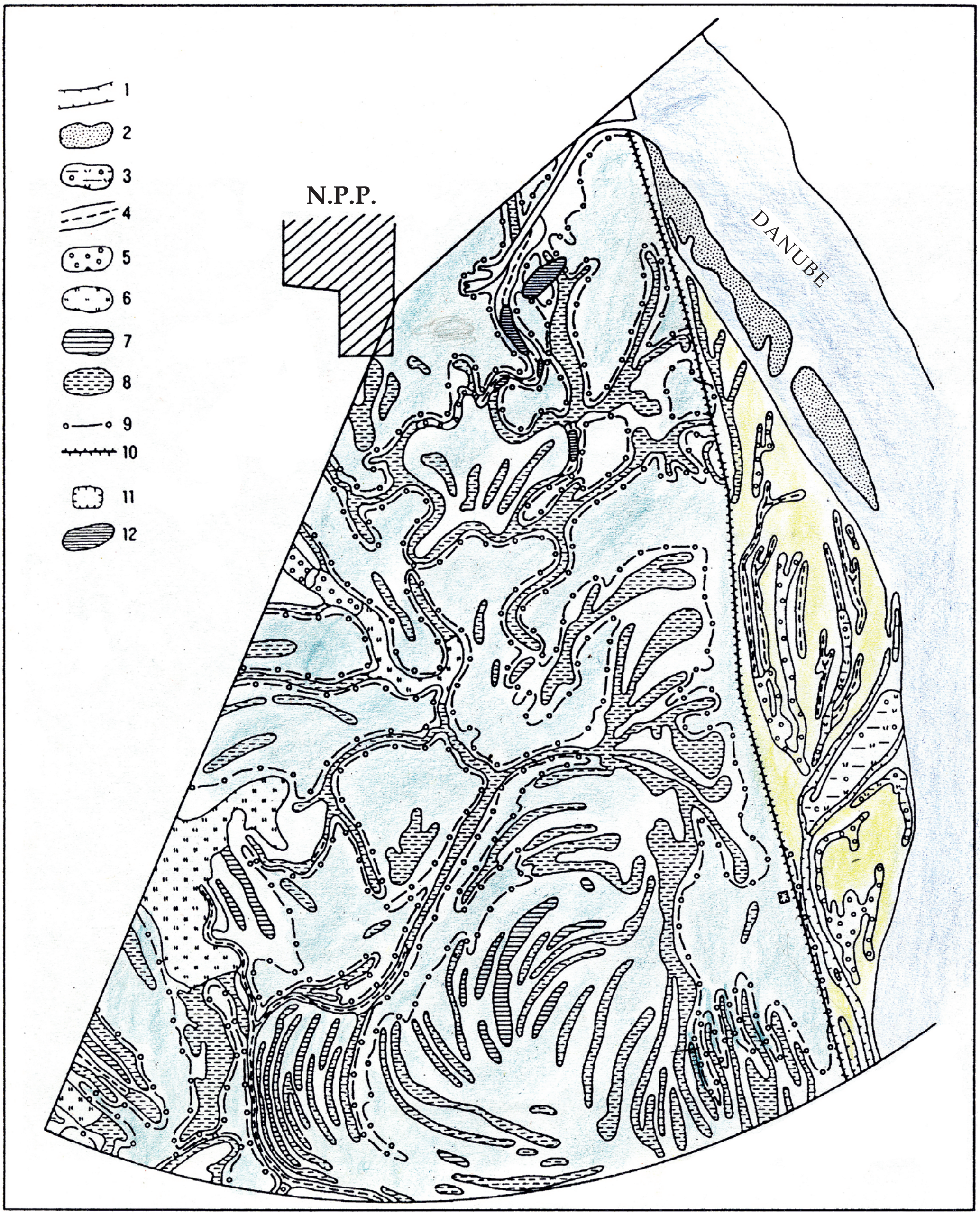

Fig. 2. Map of geomorphic groups of facies (compiled by Schweitzer, F., Balogh, J., Juhász, Á. 1991). 1 = living branches on the flood plain; 2 = point-bars; 3 = intermittent waterlogged depressions on the flood bed covered by sedge, reed and forest; 4 = intermittently waterlogged depressions; 5 = cut-off meander remnants with gallery forests; 6 = meander remnants covered by meadow; 7 = former point-bars on the low flood plain; 8 = meander under cultivation; 9 = high flood plain; 10 = dyke flood control; 11 = pit; 12 = lake

Description of the environment of the selected control points

Based on the meteorological data the prevailing wind direction is of NW–SW. This means that emitted radionuclides tend to accumulate SE and S of the nuclear power plant. It was a basic consideration when control measurement points within the area of the sector of 34 km^2 were selected. Actual and potential future measurement points were indicated on the map according to sectors of wind direction. Measurement (control) points were shown by Roman numbers with sector numbers as denominators and distances from the centre of the nuclear power plant were indicated, too *(Figure 3)*.

Control points were selected that radioactivity determinations were carried on different geomorphic facies. These were as follows:

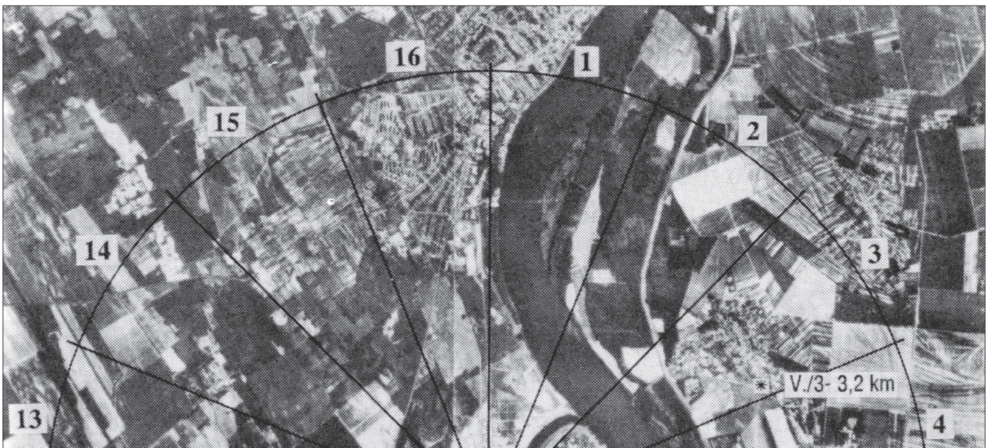

Fig. 3. Description of the environment of the selected control points (Compiled by Kanyár, B., Schweitzer, F. 1993)

I. Channelized meander (surrounded by willow-grassy vegetation and arable land);

II. Former filled-up meander under cultivation (along a dirt road flanked by a row of poplars);

III. Inter-meander point-bar (flat surface, place of a former farmstead) *(Photo 1)*;

IV. Filled-up, intermittently waterlogged meander (with reed-sedge vegetation) *(Photo 2)*;

V. Flat surface on the high flood plain (used as meadow and pasture).

The first control point (I/8 – 4.9 km) was fixed in a bending in E–W direction meander of 1 km length, 200–300 m width and 1.5 m depths *(Photo 3)*.

This meander represents the former river bed, now regulated and protected from floods. Excess water is drained by a 1.5 m deep canal. Parent materials are clay and sand covered by a meadow soil of 20–60 m thickness. About 70 per cent of the meander is cultivated, the canal is flanked by remnants of a willow grove forest.

The second control point (II/7 – 4.1 km) is situated in an non-drained former meander stretching in NE–SW direction with 500–600 m length, 150–200 m width and 1.5 m depth. The control point was set up immediately in the axis of the meander which is agriculturally cultivated, actually under winter wheat. As a result of sheet wash following intense rainfalls there is an abundant deposition along the central axis of the meander, hence the selection of this control point.

The third control point (III/7 – 2.7 km) was located on a flat flood plain surface, on a point-bar, in a geomorphological position, essentially different from the above environments. This surface at 91.8 m a.s.l. covered by meadow chernozem soil under cultivation now (a couple of years ago there was a stockyard on the spot).

Photo 1. Floodplain of the Danube protected by low embankments and natural leeves. (Photo: Schweitzer, F.)

Photo 2. Detail of the today channeled Danube between the nuclear power plant and Dunaszentgyörgy on the protected floodplain, this section was a living river bed before the flood control works (1860s). (Photo: Schweitzer, F.)

Photo 3. One of the islands surrounded by upfilled meanders and covered by 20–40 cm fluvial sand, South from the nuclear power plant (Photo: Schweitzer, F.)

The fourth control point (IV/7 – 1.4 km) is situated in the immediate neighbourhood of the power plant ('Hotwater canal'). A geo-ecologically characteristic place was chosen since the non-drained, 50–100 m wide former meander of N–S orientation represents a facies of intermittently waterlogged meanders. Its vegetation being close to a natural one is reed with high sedge and cultivated land is to be found in its surroundings *(Figure 4).*

For the studied period a 10 per cent probability of the prevailing directions is characteristic and high velocities are related with these values. In the immediate impact zone (with a radius of 5 km) multi-annual average of stormy days (with squalls of 15 m/s and more) amount to 25.

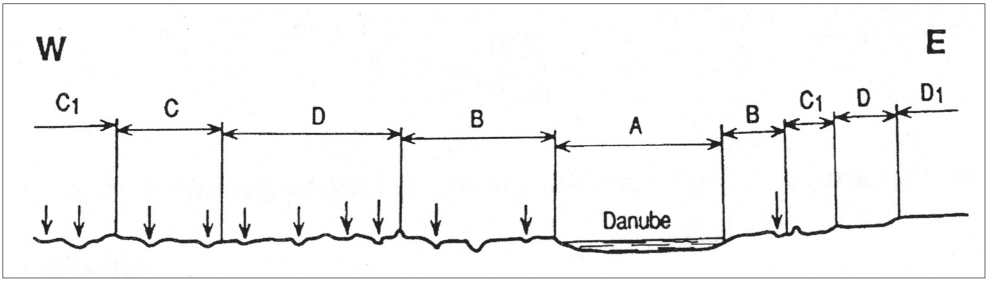

Fig. 4. Theoretical profiles of geomorphic groups of facies in the vicinity of the Paks Nuclear Power Plant with potential places of the accumulation of radionuclids (after Juhász, Á. 1991). A = actual bed of the Danube; B = flood bed with living branches; C = low flood plain with high ground water; C_1 = low flood plain with non drained meanders; D = high flood plain with meander remnants; D_1 = elevations of the high flood plain due to wind blow sand; Black arrows = potential places of accumulation of contaminants

Wind directions

They result from a rather complex interaction of two factors. The major one is a basic current determined by the general atmospheric circulation while the minor component is the modifying effects stemming from the local topographic conditions. The latter can be considered to exert a permanent influence in the impact zone of the power plant.

The spatial pattern of a potential contamination is affected eventually by characteristic north-westerly and northerly currents. Direction probabilities were elaborated for the first quarter of 1991 (the period of sampling – see *Figure 5*) which illustrate a relative high probability of occurrence of southerly and north-easterly winds, too. The medium wind velocity shows high values characteristic for the late winter and springtime, while the lowest velocities were measured in early autumn.

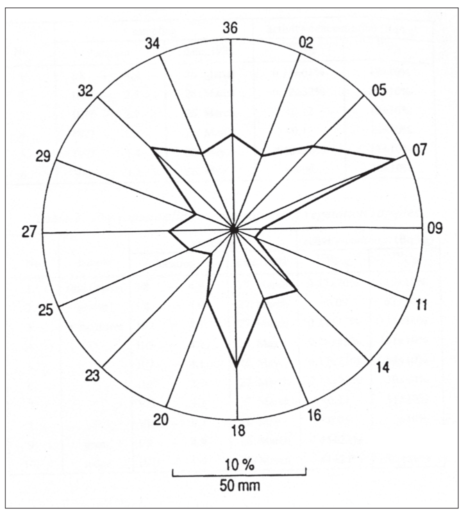

Fig. 5. Frequency of wind directions (in per cent) at Paks in the first quarter of 1991.

Determination of the radioactivity

The reaionuclide contamination of the prepared environmental samples has been determined by gamma spectrometry equipped with semiconductor detector of HpGe and multi-channel analyser type of Canberra S-35t. The relative efficiency of the detector was 20 per cent and the energy resolution 1.8–2.2 keV depending on the energy of the gamma-rays. The radioactivity of ^{110m}Ag was counted mostly by the gamma-ray of energy 885 keV and abundancy of 74 per cent. The gamma-ray with energy of 658 keV (abundancy 100 per cent) was less effective because of the close peak of ^{137}Cs-contamination (662 keV) from the Chernobyl fallout. In most of the cases the counting time was 1 day and the minimal detection level 0.1–0.5 Bq depending on the sample geometry used.

To decrease the detection limits following the chemical separation of the silver, the activity was determined in addition to gamma spectrometry by a low background (0.5–1.0 count/min) GM-tube.

For monitoring the surface activity 'in situ' gamma spectrometry of Canberra type was used. The detection limit of it was 200 Bq/m^2, therefore, only the radiocesium from the Chernobyl fallout and the natural radionuclides have been measured and not the radiosilver.

Results

Topsoil sampling and measurements have supported the assumptions based on the emission by the power plant and subsequent distribution of radionuclides about an annual surface deposition of 1 Bq/m^2. *Table 1* shows Ag concentrations achieved by measurements using low background GM-tube following the silver separation and ^{137}Cs concentrations obtained by gamma spectrometry in the topsoil samples of 1–2 mm thickness down from the surface. It is very likely that ^{110m}Ag was emitted by the Paks Nuclear Power Plant while ^{137}Cs originated from the Chernobyl fallout.

For radiosilver determination the error amounted to 30–60 per cent and half of the cases were out of measuring range, data, thus, obtained are scanty, of low accuracy and insufficient to characterise geographical distribution. Concentrations are heavily affected by the thickness of soil samples and there are ambiguities due to the thin layer of sampling.

In situ gamma spectrometry for control points I to IV resulted in surface concentrations 1,500–2,340 Bq/m^2 of ^{137}Cs for land under crops. Control point V was located E of the Danube on an non-cultivated soil (meadow). The concentration measured here was 3,640±370 Bq/m^2. The $^{134}Cs/^{137}Cs$ ratio varied between 0.07–0.12 having been 0.5 in May 1986 following the Chernobyl accident. Change of this ratio since then is due to the shorter half-period for ^{134}Cs.

Radiosilver fixed by aerosol particles is absorbed by moss vegetation easily i.e. moss accumulates radiosilver too. Therefore, sampling program included mossgrow rinds result contained in *Table 2* support the assumption on a higher accumulation of radiosilver and radiocesium contamination by mossgrown rinds compared to that that by rinds devoid of moss. Similar to measurements of soil samples radiosilver found in mossgrown rind is most likely to have originated from the Paks Nuclear Power Plant while radiocesium is a residue of the Chernobyl fallout.

Table 2 testifies on measurable quantities of ^{110m}Ag radionuclide. Nevertheless, much more measurements are necessary to investigate correlation and other interrelationship between geographical pattern of distribution, prevailing wind directions and contamination. On the other hand, emission of ^{110m}Ag into the atmosphere was the highest between 1989 and 1991 and has declined since then (1989: 2.10; 1990: 1.11; 1991: 1.26; 1992: 0.62; 1993: 0.26 GBq) so detection of radiosilver has became more difficult.

The initial target of nuclear investigations was to establish the radiation exposure of the local population living in the impact zone of the nuclear power plant due to the presence

Table 1. The radionuclide concentrations of the soil samples

No.	Sampling		Activity-concentration, Bq/kg	
	location	date, 1991	^{110m}Ag	^{137}Cs
1.	I/8 – 4.9 km	26 March	0.11±61%	19±10%
2.	III/7 – 2.7 km	26 March	0.12±32%	21±10%
3.	III/7 – 2.7 km	26 March	<0.12	12±10%
4.	IV/7 – 1.4 km	26 March	<0.12	12±10%
5.	IV/7 – 1.4 km	28 May	0.10±36%	11±10%
6.	V/3 – 3.2 km	26 May	<0.06	30±10%

Table 2. The radionuclide concentrations ofvegetation samples

No.	Sample	Sampling		Activity-concentration, Bq/kg	
		location	date, 1991	^{110m}Ag	^{137}Cs
1.	rind (mossgrown)	I/8 – 4.9 km	27 March	0.32±20%	115±10%
2.	rind (mossgrown)	I/8 – 4.9 km	27 March	<0.09	62±10%
3.	mossgrown	II/7 – 4.1 km	27 March	0.31±17%	125±10%
4.	mossgrown	II/7 – 4.1 km	27 March	0.39±18%	121±10%
5.	mossgrown	II/7 – 4.1 km	28 May	0.12±23%	54±10%
6.	mossgrown	III/7 – 2.7 km	28 May	0.11±22%	30±10%
7.	mossgrown	V/3 – 3.2 km	27 March	<0.11	11±10%
8.	mossgrown	VI/13 – 0.7 km	28 May	<0.05	5±10%
9.	grass	I/8 – 4.9 km	26 March	0.45±23%	<5±10%
10.	sedge	IV/7 – 1.4 km	27 March	0.41±25%	<5±10%

of ^{110m}Ag. Emissions and spatial diffusion of radiosilver contributed to radiation load by 5 nSv for adults and 18 nSv for one-year old children at a distance of 3 km in 1989. The critical pathway is cattle feedstaff and milk consumption.

Summary and conclusion

Places of the enrichment in radioisotopes selected with taking the preceding geoecological survey into account were analysed by 'in situ' gamma spectrometry and laboratory measurements of the collected samples. Due to the extremely low amount of ^{110m}Ag isotope was extracted from the soil and vegetation samples using chemical methods to reduce threshold values of its detection.

Apart from the quantity of natural radionuclides the applied methods permitted to determine radiocesium of the Chernobyl fallout and radiosilver originating from the Paks Nuclear Power Plant.

1. In the case of a potential accident the various geoecological facies: flood beds, river channels, low flood plain and high flood plain are to be treated differently.
2. Surfaces particularly prone to the accumulation of radionuclides are the non-drained and abandoned river channels and meander remnants with no communication with the surrounding areas. In the non drained former meanders the accumulation is enhanced by permanent or intermittent waterlogging. This morphofacies behaves as a pollution trap.
3. An important aspect is the way of land use, seasonal or annual variations of the crop pattern, the circulation and accumulation of vegetation matter.
4. For the time being surveying the local isotope traps is in an initial stage and data are scanty, moreover, due to sampling and measuring errors (amounting to 20–70 per cent) an evaluation of deviations would be too early. At any rate, sensitive analytical methods allowed the detection of radiosilver

in the soil, namely 0.1 Bq/kg concentration in the uppermost mms. This corresponds to a nearly 1 Bq/m^2 surface contamination.

5. Mossgrown rind and grass and sedge collected from undisturbed places contained 0.4–0.5 Bq/kg (dry weight) radiosilver concentration.

6. Similar to the earlier reports it should be emphasized that contaminations from the NPP are negligible from a radiation hygienic viewpoint and the resulting load contribution is a mere one per ten thousand part of the natural background radiation fluctuating by 20–30 per cent. The experience, however, might be very useful for the elaboration of a sampling programme to be realised in the case of a potential accidental emission.

7. Activity of ^{137}Cs as a consequence of the Chernobyl fallout varied 1.5–3.6 kBq/m^2 at the five control points which value is below the country average.

Problems and suggestions concerning the future activities can be summed up as follows:

– The study of local enrichment in radioisotopes under natural conditions is hindered by agricultural activities for there are very few places with undisturbed topsoil where the superposition of contaminants of several years could be established.

– Measurements conducted up till now are not sufficient for the comparison of places of isotope enrichment with the contamination of other regions. Further and extended investigations are necessary.

– For a future mapping of radiosilver originating from the emissions by the Paks Nuclear Power Plant along with an earlier applied analysis of the tissue of liver of sheep grazing on the local pastures, measurements of contamination accumulated by vegetation, primarily that by moss should be involved.

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