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RESEARCH ARTICLE

Environmental impact assessment of radioactive water pipe leakage at NPP Paks

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Abstract

Environmental impact of the leakage of radioactive water into the soil from a subsurface-pipeline on the site of NPP Paks was studied to assess the size of the contaminated area and to estimate the potential migration of radioisotopes. For this aim a comprehensive analysis study was performed on soil samples taken from the contaminated area. The activity concentration of representative radionuclides (such as ^{137}*Cs,* ^{134}*Cs,* ^{60}*Co,* ^{54}*Mn,* ^{7}*Be,* ^{3}*H and* ^{90}*Sr), the composition of soil, the distribution of radioactivity in different grain size fractions and the pH of soil were determined. Dissolving experiments with synthetic acidic rain water was also carried out.*

Results of the activity-measurements show that the average activity concentrations in the case of all isotopes are below the exemption limit given by the authority. The short-term migration of radioisotopes has been found to be negligible. Since pH of the soil is about natural and the radionuclides are very strongly bounded to the soil, the risk of the long-scale groundwater contamination is low.

Keywords

pipeline · leakage · radionuclides · soil · pollution · migration

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1 Introduction

On the site of NPP Paks in Hungary, sinking of soil was observed along a subsurface-pipeline transporting low- and intermediate level waste water. Since the preliminary quick screening indicated that the pipe had lost its compactness in more spots and radioactive water was leaking into the soil, the operation of the pipe was immediately stopped. For the assessment of the possible contamination of the vicinity of the pipeline large number of representative soil samples were collected and analyzed.

Soil studies carried out for the discharge assessment of the operation of nuclear facilities in general focus on the determination of the horizontal and vertical distributions and the possible migration of the radioisotopes [1]-[6]. For the surface studies the soil samples are usually taken from max. 0.5 meter depth from surface. Since the source of the leakage in our case was subsurface in nature, soil from deeper regions had to be sampled as well.

In this paper we present the results of the comprehensive study performed for the assessment of the extension of the contaminated area and its stability on the short term by repeating the analysis scheme several months later. In the first stage the activity concentrations of radionuclides (such as ^{137}Cs, ^{134}Cs, ^{60}Co, ^{54}Mn, ^{7}Be, ^{3}H and ^{90}Sr) in soil samples collected from the identified leaking spots were measured. The characteristic composition of soil, the distribution of activity of individual radioisotopes in different grain size fractions of soil and pH of soil were also determined. In addition, leaching experiments with synthetic acidic rain on samples of larger activity concentration were also carried out for the assessment of the mobility of isotopes in soil [6].

From the measured activity concentrations, maps were constructed showing the horizontal distribution of radionuclides near the leaking spots. These maps may help for the estimation of contaminated soil volume and migration of isotopes on a longer time scale.

2 Materials and methods

2.1 Sampling

During the preliminary screening, leakage was identified in three spots with approximately 50-50 meter distance found from each other. At each leaking spot 22 samples were collected, 11 samples from soil surface and 11 from depth of 2 meters (Fig 1). For the estimation of migration of radionuclides the sampling and the analysis were repeated three months later (Fig. 2). By the repeated sampling 32 samples were collected. The sampling was carried out by a soil-exploratory instrument (BORRO type), the mass of individual samples was about 2.5 kg.

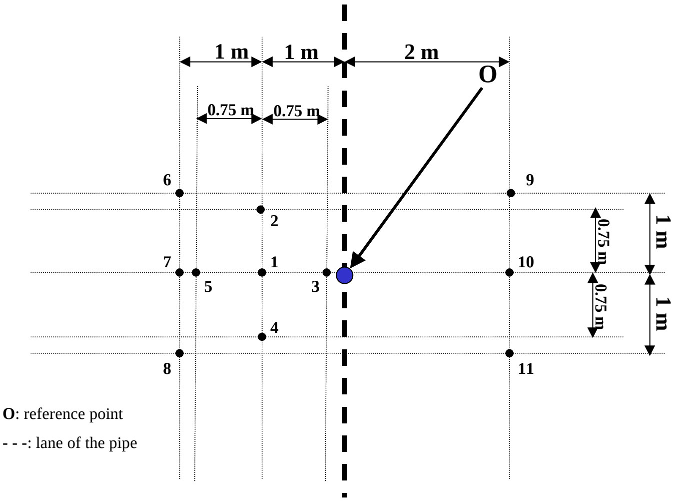

Fig. 1. Sampling scheme of the contaminated area near a leaking spot.

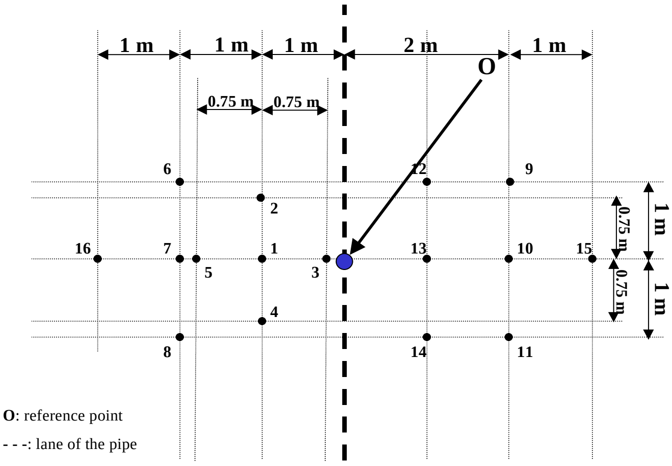

Fig. 2. Sampling scheme for the second analysis performed three months after the first study.

2.2 Activity-concentration measurements

The ^{137}Cs, ^{134}Cs, ^{60}Co, ^{54}Mn and ^{7}Be content of soil was determined by high resolution (HPGe) gamma-spectroscopy. The samples were dried at 60 C° for 4 hours and 100 g dried samples were measured (for at least 10 000 s measuring time). The characteristic range of activity-concentrations obtained for the individual isotopes in the samples are listed in Table 1 from which it can be seen, that the magnitude of the ^{137}Cs contamination could be considered to be the most significant, therefore for this isotope the activity-concentration distribution map was constructed. From the groundwater contamination point of the view, the possible change in the distribution of isotopes at the underground level is very important, thus the 2 meter deep distribution of ^{137}Cs around the most contaminated spot is illustrated by Figs. 3 and 4.

Tab. 1. Range of gamma-spectroscopic measurements

Origin of samples	^{7}Be	^{54}Mn	^{60}Co	^{134}Cs	^{137}Cs
		(Bq/kg dry mass*)			
I S	0-2.8	0-1.9	0-8.8	0-15.5	2-81
II S	0-4.9	0-3.6	0-13.2	0-73	7-375
III S	0-4.5	< 0.8	0-8.6	0-3.3	2-16
I D	0-2.7	< 0.3	0-7.1	0-2.4	0-14.4
II D	0-2.5	< 0.9	0-3	0-4.5	0-24
III D	0-4,8	0-3.3	0-14.1	0-8.4	0-35
3 months later repeated measurements					
I S	0-5*	0-1.7	0-4.7	0-6.5	1.3-30.6
II S	0-6.6	0-2.1	1-8	1.5-23	8-109
III S	0-2.3	< 0.7	0-3.7	0-4.2	1-18.6
I D	0-4.2*	< 0.6	0-7	0-10	0-38
II D	0-2	< 0.7	0-3.6	0-6.7	0-31
III D	0-2.3	< 0.8	0-7.6	0-2.5	0-10.5

*: water content of soil samples 5-15%

I, II, III: numbers of the leaking spots

S: surface samples; D: samples from 2 meter depth

For the ^{90}Sr measurements 3-4 samples were selected and analyzed at each spot using a crown ether extraction method [7]. The activity of ^{90}Sr was determined using liquid scintillation counting technique, the results are shown in Table 2.

The ^{3}H-activity of bounded water content of soil samples was determined by a standard method. Water content of 2-2.5 kg soil sample was removed by a special vacuum-system and the frozen water was detected using liquid scintillation counting (Table 3).

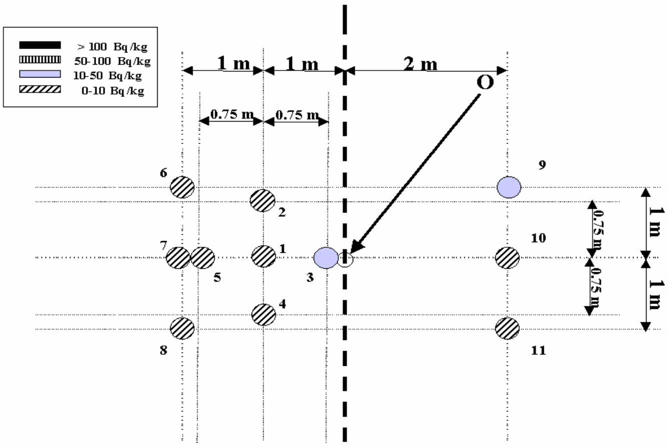

Fig. 3. The distribution of ^{137}Cs at the most contaminated leaking spot (marked with II) in depth of 2 meters

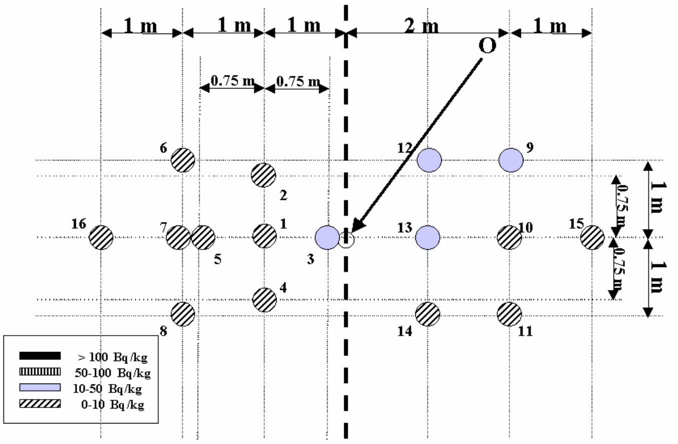

Fig. 4. The distribution of ^{137}Cs in the most contaminated spot II in depth of 2 meters, 3 months later

Tab. 2. Activity-concentration of ^{90}Sr in soil samples

First measurements		3 months later repeated measurements	
Origin of samples*	^{90}Sr (Bq/dry mass)	Origin of samples*	^{90}Sr (Bq/dry mass)
I S	0.93	I S	0.96
II S	7.73	II S	0.78
III S	0.95	III S	1.96
I D	1.46-1.50	I D	0.70-0.93
II D	0.69-2.22	II D	0.58-1.12
III D	0.77-1.25	III D	0.80-0.87

*: At each spot sample was collected at the same position relative to the reference point of sampling (see Fig. 1 and Fig. 2)., data here represent the maximal values.

Tab. 3. ^{3}H Activity-concentration of water content of soil samples

First measurements		3 months later repeated measurements	
Origin of samples*	^{3}H (kBq/L)	Origin of samples*	^{3}H (kBq/L)
I S	4.56	I S	2.01
II S	3.48	II S	0.24
III S	10.8	III S	0.54
I D	0.25-117.4	I D	3.5-184.7
II D	2.05-117.6	II D	11.8-107.9
III D	2.25-10.6	III D	0.52-4.99

*: At each spot sample was collected at the same position relative to the reference point of sampling (see Fig. 1 and Fig. 2), data here represent the maximal values.

2.3 Determination of soil composition

The composition of soil samples was determined by grain size distribution analysis where clay, silt, sand and coarse fractions were distinguished (Table 4).

2.4 Measurement of activity in different particle size fractions of soil

In the case of a more active sample we have determined the distribution of nuclides, measurable by gamma-spectroscopy, in different soil fractions (Table 5). The chosen sample originated from the sampling place marked with *II S3*.

2.5 Synthetic acidic rain leaching experiments

In the case of sample *II S3,* leaching experiments were also carried out with synthetic acidic rain water. The pH of the solution was adjusted to 4.5 with ammonium-acetate buffer prepared by mixing of 0.1 M ammonium-acetate and 0.1 M acetic-acid solution in proportion of 1:1. Synthetic rain water solution and soil were mixed in weight ratio of 3:1, and after six hours mixing the two phases were separated by filtration. The liquid phase and the soil dried at 60 C° were measured by gamma-spectroscopy, the results are presented in Table 6.

2.6 pH measurements of soil

The pH of soil samples were determined by the soil-suspension method. The pH values we have found to be between 6.4 and 6.6.

3 Results and discussion

3.1 Isotope inventory

The quantity and spatial distribution of radionuclides in the individual contamination spots could successfully be determined. The greatest levels of contamination were found for ^{137}Cs and ^{3}H isotopes. In general, the surface activity of isotopes – except ^{3}H – was greater than in depth of 2 meters, which may be attributed to the up-stream of the waste water from the pipe. In the case of ^{3}H however, the surface activities are 1-2 order of magnitude lower than in 2 m deep. This can be explained with the washing effect of precipitate fallen since the leakage and/or the evaporation of soil water, because the mobility of ^{3}H – due to the fast isotope-exchange taken place between pore-water of soil and precipitate – is fairly large in the soil.

The total mass, volume, maximal isotope specific activity-concentrations and total activity of the contaminated soil at each spot are summarized in Table 7. In addition, the corresponding exemption limits are also indicated [8]. The contaminated soil volume was estimated to be around 40 m^{3} at each leaking spot, the mass of soil was calculated with the average density of sand (1.6 kg/m^{3}). It can be seen from the table that none of the activity-concentrations of the individual isotopes exceeded the limits. In the case of ^{7}Be and ^{54}Mn the total activity values calculated do not exceed the limit given by authority, but in the case of ^{60}Co, ^{134}Cs, ^{137}Cs, ^{90}Sr and ^{3}H the total activities are a little

Tab. 4. Characteristic composition of soil

Origin of samples	Fractions	Characteristic fraction size (μm)	Quantity (%)
surface	clay	< 50	5-10
	silt	50-250	20-25
	sand	> 250	65-75
	coarse fraction		5
samples from depth	clay	< 50	10-15
	silt	50-250	25-30
	sand	> 250	45-55
	coarse fraction		5

Tab. 5. Distribution of activity in different soil fractions

Sample code	Quantity (%)	^{7}Be	^{54}Mn	^{60}Co (Bq/dry mass)	^{134}Cs	^{137}Cs
II S3 sand	33.0	< 4.7	3.0±0.5	8.7±0.4	27.6±0.7	116.7±3.3
II S3 silt	62.9	< 6.1	2.7±0.7	9.2±0.6	22.0±0.7	95.0±2.9
II S3 clay	4.1	< 9.8	20.7±1.1	20.7±1.1	48.5±1.4	210±7
II S3 Total		< 4.8	3.6±0.6	10.7±0.8	23.1±0.8	104.9±3.4

Tab. 6. Results of the synthetic acidic rain water leaching experiment of sample *II S3*.

Sample code	^{7}Be	^{54}Mn	^{60}Co	^{134}Cs	^{137}Cs
Acidic rain (Bq/L)	< 3.3	< 0.2	< 0.2	< 0.2	< 0.2
II S after leaching (Bq/dry mass)	< 8.6	1.5±0.7	23.0±1.0	23.2±0.8	105.7±3.5
II S before leaching (Bq/dry mass)	< 4.8	1.6±0.6	20.7±0.8	23.1±0.8	104.9±3.4

larger than the corresponding limit. If the above calculations are made using the average activity-concentration values however, the polluted soil may be considered below the exemption limit.

3.2 Short-scale mobility of the isotopes

From the data presented in Table 1 it can be seen that the activity-concentrations in depth of 2 meters did not significantly changed in 3 months period. This is also illustrated for the distribution of ^{137}Cs in depth of 2 meters for the most contaminated place in Fig. 3. and Fig. 4. Furthermore, it can be stated that acceptable correlation can be shown in activity-concentrations between ^{137}Cs and the other measured isotopes, thus the ^{137}Cs-profile on the activity-distribution map adequately indicates the spreading of the contamination.

During the three months waiting period some landscaping were performed by the operator near the contaminated spots I and II, which had some reducing effects on the level of the surface activity-concentrations of isotopes, which is also indicated in Table 1. This was not the case for spot III, for which it can be concluded that the surface activity-concentration values did not changed significantly. The surface activity-concentration values of ^{3}H however, show a marked decrease during this period for each cases (Table 3), which may be attributed to the washing effect of the precipitate and surface evaporation. Furthermore, it can be noted here that this effect was partly compensated by the surface layer mixing of the landscaping.

In general it can be concluded that the state of the contamination did not change during the observed time period, thus it can be announced that the NPP have successfully stopped the environment polluting activity. The negligible extent of radionuclides migration – excepted for ^{3}H – can also be proven by the results of the soil pH measurements and leaching studies. The natural pH of soil is not favorable for the migration of isotopes and the leaching investigations showed that the isotopes are strongly bounded to the soil (Table 6).

This assessment contains uncertainties due to the seasonal variations in climatic and hydrological conditions that can influence the migration. In addition, the greatest amount of activity is bounded to the finer components (fine sand and clay) of the soil (Table 5) thus the finer grain fraction of the surface soil may be spread by wind on the long term.

Tab. 7. Total mass, volume, maximal isotope specific activity-concentrations and total activities at the individual contaminated spots.

Leaking spot	Soil (m^3) (kg)	^{7}Be (Bq/kg) (Bq)	^{54}Mn (Bq/kg) (Bq)	^{60}Co (Bq/kg) (Bq)	^{134}Cs (Bq/kg) (Bq)	^{137}Cs (Bq/kg) (Bq)	^{90}Sr (Bq/kg) (Bq)	^{3}H (Bq/kg) (Bq)
I	40 $6.4 \cdot 10^4$	4.9 $3.2 \cdot 10^5$	1.7 $1.1 \cdot 10^5$	8.8 $5.5 \cdot 10^5$	15.5 $1.0 \cdot 10^6$	81 $5.0 \cdot 10^6$	1.1 $6.4 \cdot 10^4$	$1.8 \cdot 10^5$ $1.1 \cdot 10^{10}$
II	40 $6.4 \cdot 10^4$	6.6 $4.1 \cdot 10^5$	3.6 $2.2 \cdot 10^5$	13.2 $8.2 \cdot 10^5$	73.2 $4.6 \cdot 10^6$	375 $2.3 \cdot 10^7$	1 $6.4 \cdot 10^4$	$1.2 \cdot 10^5$ $7.8 \cdot 10^9$
III	40 $6.4 \cdot 10^4$	4.8 $3.0 \cdot 10^6$	3.3 $2.1 \cdot 10^5$	14.1 $9.0 \cdot 10^5$	8.4 $5.4 \cdot 10^5$	35 $2.2 \cdot 10^6$	2.0 $1.2 \cdot 10^5$	$1.1 \cdot 10^5$ $7.0 \cdot 10^9$
Exemption limit [8]		$1 \cdot 10^6$ $1 \cdot 10^7$	$1 \cdot 10^4$ $1 \cdot 10^6$	$1 \cdot 10^4$ $1 \cdot 10^5$	$1 \cdot 10^4$ $1 \cdot 10^4$	$1 \cdot 10^4$ $1 \cdot 10^4$	$1 \cdot 10^5$ $1 \cdot 10^4$	$1 \cdot 10^9$ $1 \cdot 10^9$

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