Environmental Radio-Analytical Research in the Field of a New LILW Disposal Facility in Hungary - 12401

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ABSTRACT
The new Hungarian National Radioactive Waste Repository was established in the granite of the Mórágy Block Mountains (Bátaapáti) to store low and intermediate level radioactive waste originating from Paks Nuclear Power Plant. Before the start of the operation of the facility the environmental characteristics of the site and its vicinity, i.e. the so-called zero level was determined. The determination of the zero level is inevitable as the evaluation of the measurement data in the course of the operation of nuclear facilities should mainly be related to this zero level. In the course of the monitoring activity environmental elements, i.e. air, soil, water (springs, streams, precipitation, fall-out, wash-out, plant and animal samples were investigated from several points of view. From the samples radiocarbon, tritium, Sr-90, gamma emitters, gross alpha, gross beta, and field gamma measurements were carried out. Results reported show that the state preceding the operation of the facility can approximately be considered as the Hungarian background data.

INRTODUCTION

The question of the permanent disposal of low and intermediate level radioactive wastes (LILW) generated in the course of the operation of Paks Nuclear Power Plant became nowadays an important and urgent task. The granite of the Mórágy Block Mountains proved to be the most proper solution from the long-term disposal's point of view. To fulfil this task the National Radioactive Waste Repository (NRWR) was built in Bátaapáti (46.22225° N, 18.60028° E). The central and technology building of the facility began its operation in the autumn 2008. The requirements of the long-term safe operation of the facility, a way of the environmental monitoring and the rules regarding the radioactive emissions to the atmosphere and to the hydrosphere, as well as their control are regulated by the decree 15/2001.(VI.6.) KöM of the Ministry of Environmental Protection [1]. The task of the environmental monitoring is to observe the changes and tendencies in the extent of the activity level of radionuclides in the environmental elements (soil, water, precipitation, air, plants and animals) by continuous sampling and measurements. It should be certified by continuous measurements whether radioactive contamination detectable by the most state-of-the-art and most sensitive measurement process was emitted from the facilities of the NRWR. To the radiation protection monitoring of the emissions 5 permanent sampling points with so called "A"-type monitoring stations (containers) were installed (Fig. 1.).

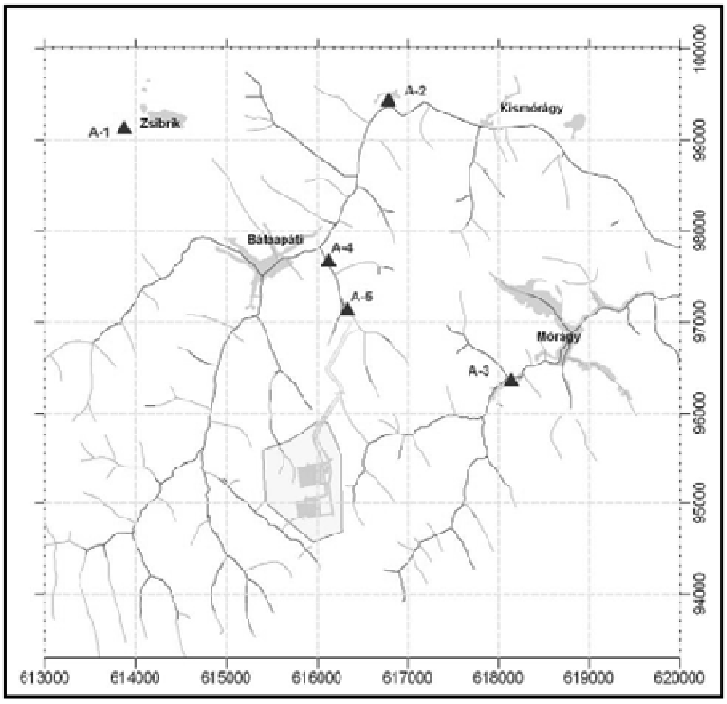

Fig. 1. Locations of the „A”-type monitoring stations in the vicinity of the NRWR.

At these locations continuous C-14, H-3, fall-out/wash-out, aerosol and temporal soil and plant samplings are performed. In the region of the facility spring and stream waters were sampled for radio-analytical measurements.
The determination of the natural background is only possible before the beginning of the storage of the wastes, therefore, the environmental state of the facility and its vicinity was investigated prior to its operation in autumn 2008. During the operation period measurement data should be related mainly to this zero level.

METHOD

Soil samples

Combined bulk soil samples were taken from the upper 10 cm layer from many locations in the vicinity of five “A”-type stations for laboratory gamma and Sr-90 activity measurements. Soil samples were air dried, homogenised and sieved at 2 mm hole diameter. Gamma measurements were performed directly on the sieved fraction by a GC10021 HPGe detector (Canberra) [2], [3]. Strontium for the Sr-90 activity measurements were extracted from the soil samples by crown ether separation method combined with oxalate co-precipitation. The activity of the Sr-90 was measured by an ultra low background Canberra Packard 3170 TR/SL liquid scintillation counter [4].

Spring and stream samples

As the waste will be disposed below ground level, by the investigation of spring and stream waters it can be checked whether radioactive contamination gets to the

environment from the facility. Altogether, sampling of 11 springs and streams were performed. Water samples were taken to determine the activity of H-3 Sr-90 and gamma emitters, and to the conventional water chemistry and ICP-AES measurements. Parallel with sampling the temperature, the pH and the conductivity of the waters were measured.

Atmospheric radiocarbon, tritium and fall-out/wash-out samples

Sampling units suitable for sampling atmospheric C-14 and H-3 were installed on the "A"-type stations. These units are able to take radiocarbon samples from CO_2 and carbohydrates and H-3 samples from vapour and carbohydrates. The sampling periods take two months [5]. C-14 measurements were performed by gas proportional technique (GPC) [6], while H-3 measurements by an ultra low background Canberra Packard 3170 TR/SL liquid scintillation counter [7]. With the help of fall-out/wash-out sampling it is possible to simultaneously collect radionuclides got to the surface from the atmosphere by dry settling and wet deposition. Sampling occurred by a funnel with the area of 1 m^2 prepared for this purpose. Samples collected by the continuously operating fall-out/wash-out sampling units were gathered monthly. From the samples collected gamma measurements were performed.

Plant samples

As a first step the most proper monitoring plant advantageous for both the sampling and for the laboratory investigations was selected. Dandelion (Taraxacum officinale) proved to be the best solution. It can be found practically everywhere and identified with ease. It sprouts every year, therefore, it shows information about the given year. It accumulates alkali earth metals including strontium well, thus, better detection level can be reached compared to plants accumulating alkali earth metals in a less extent. Sampling occurred in the vicinity of the "A"-type stations. Washed and unwashed samples were taken at each station. After preparation gamma, gross-α and gross-β, and after combustion Sr-90 measurements were made.

Animal samples

Radionuclides from the facility can get into the higher organisms including humans via the food chain, therefore, the examination of the haslets of a young wild boar (Sus scrofa scrofa) from within a range of 5 km of the facility was also performed. The determination of the Sr-90 activity was fulfilled from bones [8] and that of the gamma activity from the haslets and bones. At the examination of the haslets two significantly different organs were prepared. Liver is an organ isolated physically from the outside world, however, a significant part of the nutrients are processed in it. The other organ examined is the lung being in a direct connection with the outside world. Some of the aerosols in the atmosphere adsorbs irreversibly when got into the lung giving an inner radiation load from the atmospheric isotopes.

RESULTS

Results of soil samples

Data from measurements in the area of the NRWR, Bátaapáti were compared to the average values of Hungary. Activity concentrations of the radionuclides of natural origin in the soil samples do not differ significantly from the Hungarian values (Fig. 2.).

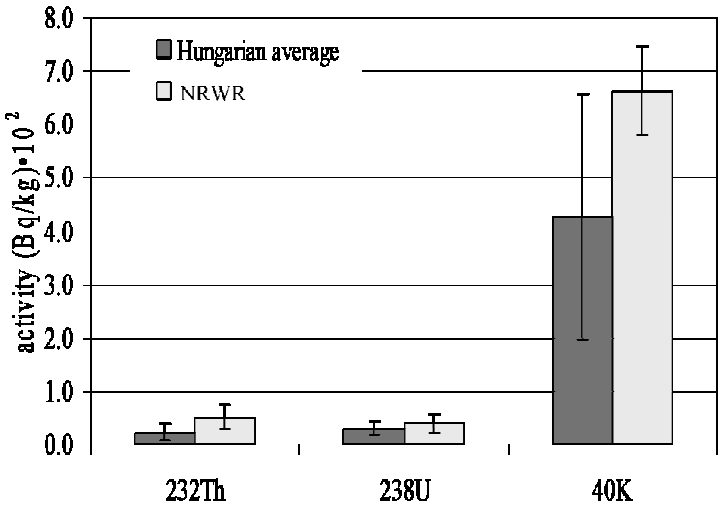

Fig. 2. Comparison of the gamma emitter isotopes of natural origin measured in the NRWR with the Hungarian average values.

The average Cs-137 activity concentration in the soil examined is 5.68 Bq/kg, which is approximately half as high as the country average according to the data of the National Environmental Radiological Monitoring System (13.7 Bq/kg) [9]. This activity is due to the former atmospheric nuclear weapon tests and the reactor accident in Chernobil. Taking into consideration that the contamination of Chernobil origin was higher in the northern part of Hungary, the average value of the soil samples from the vicinity of the NRWR can be accepted as zero level.

The Sr-90 activity concentration of the soil varies between 0.80-1.54 Bq/kg. Measurement data were compared with data of samples taken from the vicinity of Paks Nuclear Power Plant and Püspökszilágy Radioactive Waste Treatment and Disposal Facility. The average Sr-90 activity concentration of soils in Paks was recently 0.6 Bq/kg [10], and in Püspökszilágy 0.52 Bq/kg [11]. Sr-90 activities measured in the vicinity of Bátaapáti were similar to the values in Paks.

Results of the spring and stream samples

Activity of the gamma emitter radio isotopes detectable in the water samples is identical with the natural conditions. The Sr-90 activities of the spring waters (B34, B35, B40, Roc.) are below detection level of 0.5 mBq/dm^3, however, in the surface waters the activity concentrations were 1.56-7.08 mBq/dm^3 (Fig. 3.). Spring waters got below the surface relatively long time ago, therefore, they are contaminated with Sr-90 isotope of

artificial origin falling out from the atmosphere in an undetectable or only extremely low extent.

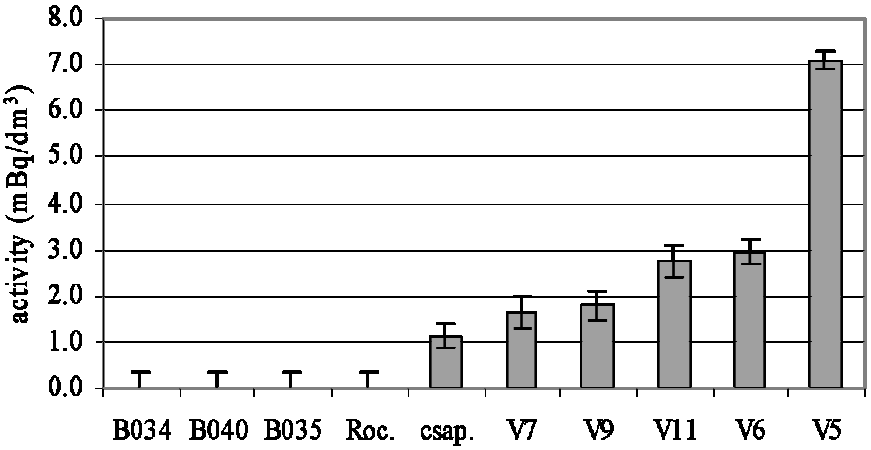

Fig. 3. Sr-90 activities of the spring and stream waters

The gross-β activity exceeds the detection level 2.76·10-1 Bq/dm^3 in only few cases. The average gross-α activity 1.4·10-2 Bq/dm^3, which can be considered as natural, as well. The H-3 activity is higher in the surfaces water [(5.7±0.1)·10^{-1} Bq/dm^3] than in the spring water [(3.0±0.1)·10^{-1} Bq/dm^3]. This is a natural phenomenon, as H-3 is of atmospheric origin and has both natural and artificial sources. The half-life of H-3 is 12.3 years, thus older spring waters have lower H-3 activity than stream waters mixed with newly fallen precipitation. The H-3 activity of stream waters is identical with the natural background level [9].

Results of the atmospheric radiocarbon and tritium samples

According to the results of the samplers both the atmospheric radiocarbon [(4.21-4.36)·10^{-2} Bq/m^3 air] and the atmospheric H-3 activities [8.0·10^{-3}-2.99·10^{-2} Bq/m^3 air] are identical with the background measured in Hungary. As a comparison the B24 background station of Paks in Dunaföldvár showed activity concentrations of C-14 to be [(4.0-4.4)·10^{-2} Bq/m^3 air] and H-3 to be (7.2·10^{-3}-3.1·10^{-2} Bq/m^3 air). The radiocarbon and H-3 in the carbohydrate fractions, and HT do not add a surplus to the C-14O_2 (CO_2) and HTO activity [10,11]. The gross gamma activity of the fall-out/wash-out samples varies between 0.6-8 Bq/dm^3; the minimum value was measured in winter. Nearly the 90% of the activity is made up by the Be-7 generated as an effect of the cosmic and solar background radiation.

Results of plant samples

Regarding the gamma activity concentration of plant samples there is no significant difference between the washed [(1.95±0.28)·10^3 Bq/kg dry mass] and the unwashed [(1.99±0.40)·10^3 Bq/kg dry mass] samples (Fig. 4.).

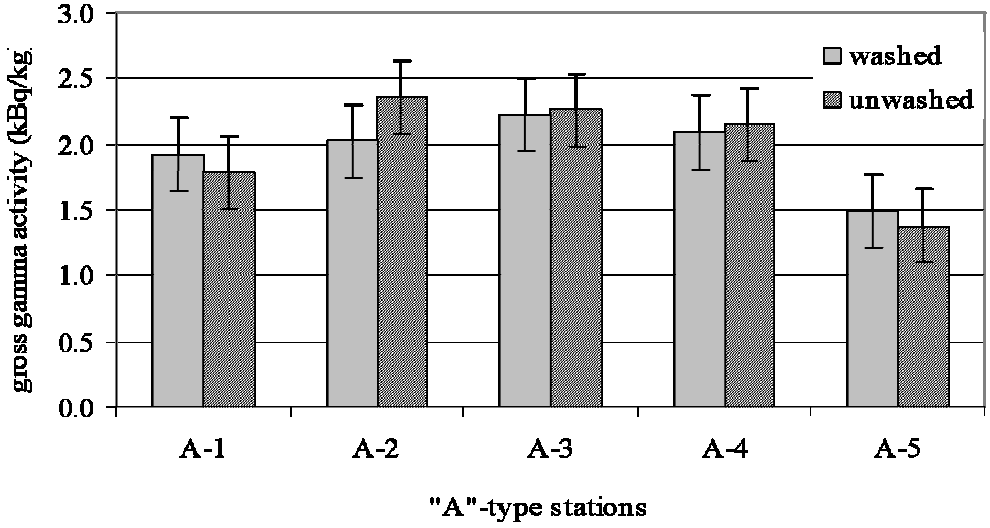

Fig. 4. Comparison of the gross gamma activity of the washed and unwashed plant samples.

The reason of the phenomenon is that the majority of the activity of plants comes from the K-40. Among the natural isotopes activities of Be-7, K-40, Ra-226 and Th-232 could be detected. The average Sr-90 activity concentration is 2.61 Bq/kg dry mass. Average value of Hungary is unknown, only a value in the vicinity of Paks Nuclear Power Plant is given in the literature. In the basis of these data, the average Sr-90 activity concentrations in plant samples varied between 1.5-4.2 Bq/kg dry mass between 1988-1998 [10]. These activities are due to the global contamination in the 60's and the Chernobil accident, and they have decreased in a small extent since then. The gross-β and the K-40 gamma activity concentrations [$(1.0\text{-}1.56)\cdot 10^3$ Bq/kg dry mass] are in good accordance with each other, and show that in plants there is no detectable artificial beta emitter isotope except the uptake from the soil contaminated by the Chernobil fall-out.

Results of animal samples

Cs-137 activity concentrations measured in the animal bone and haslets are close to the detection level (0.06-0.1 Bq/dm^3). The Sr-90 activity was determined from the bone of the animal as strontium similarly to calcium is bone seeker and builds into the organism in a similar way. Sr-90 activity concentration was found to be 23.7 Bq/kg dried bone. This value is an order of magnitude higher than the activities in the environmental elements proving the significant strontium accumulation ability of the bone. K-40 is responsible for the majority of the activity 19.1±1.1 Bq/kg in bone to 96.1±1.1 Bq/kg liver. In the electrolyte homeostasis sodium plays a significant role in animals, therefore, the mainly cosmogenic Na-22 gave a detectable activity.

DISCUSSION

The determination of the zero level is inevitable as the evaluation of the measurement data in the course of the operation of nuclear facilities should mainly be related to this

zero level. In the course of the monitoring activity environmental elements, i.e. air, soil, water (springs, streams, precipitation), fall-out, wash-out, plant and animal samples were investigated from several points of view. From the samples radiocarbon, H-3, strontium-90, gamma emitters, gross alpha, gross beta, and field gamma measurements were carried out. Results reported show that the state preceding the operation of the facility can approximately be considered as the Hungarian background data.

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