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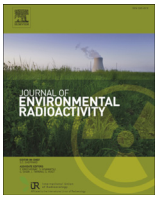

Monitoring of tritium, ^{60}Co and ^{137}Cs in the vicinity of the warm water outlet of The Paks Nuclear Power Plant, Hungary

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ABSTRACT

Danube water, sediment and various aquatic organisms (snail, mussel, predatory and omnivorous fish) were collected upstream (at a background site) and downstream of the outlet of the warm water channel of Paks Nuclear Power Plant. Gamma emitters, tissue free-water tritium (TFWT) and total organically-bound tritium (T-OBT) measurements were performed. A slight contribution of the power plant to the natural tritium background concentration was measured in water samples from the Danube section downstream of the warm water channel. Sediment samples also contained elevated tritium concentrations, along with a detectable amount of ^{60}Co. In the case of biota samples, TFWT exhibited only a very slight difference compared to the tritium concentration of the Danube water, however, the OBT was higher than the tritium concentration in the Danube, independent of the origin of the samples. The elevated OBT concentration in the mollusc samples downstream of the warm water channel may be attributed to the excess emission from the nuclear power plant. The whole data set obtained was used for dose rate calculations and will be contributed to the development of the ERICA database.

1. Introduction

The Paks Nuclear Power Plant is situated 1800 m from the Danube River in south-central Hungary (46°34′N; 18°51′E). The power plant is operating by four pressurised-water reactor (PWR) blocks. Danube water provides the cooling water supply for the reactors, and the spent cooling water returns to the Danube through the warm water channel ($4\cdot10^5$ m^3/h). The treated wastewater of the facility is also discharged into the Danube near the outlet of the warm water channel (60 m^3/h).

According to measurements of the Joint Environmental Radiation Monitoring System during 2010 (NRIRR, 2011), ^{3}H (tritium), ^{14}C, ^{54}Mn, ^{58}Co, ^{60}Co, ^{106}Ru, ^{110m}Ag, ^{124}Sb, ^{125}Sb, ^{131}I, ^{134}Cs and ^{137}Cs have been detected in the liquid discharge. In addition to this previous monitoring work we collected samples from fluvial living organisms and sediments for different isotope analyses in September 2010. The isotope concentrations obtained were then used for dose-rate calculations and the impact of the nuclear power plant on the aquatic environment.

As summarized by Smith et al. (2006), effluent released to a river disperses due to hydraulic processes (transport and diffusion), while geochemical processes influence the interaction of dissolved radionuclides with suspended matter and bottom sediments. Sedimentation and resuspension are of importance for controlling the two-way migration of radionuclides from the water column to the bottom sediments and vice versa. These processes have been studied and modelled based on fission and activation products such as ^{54}Mn, ^{60}Co, ^{90}Sr and ^{137}Cs by several authors (Benes and Cerník, 1997; Monte et al., 2005; Smith et al., 2006). Similarly, a number of models exist for the description of processes controlling the migration of radionuclides from the abiotic to the biotic components of river systems (Monte et al., 2005; Smith, 2006). In practice, if the uptake rate of a radionuclide is assumed to be in equilibrium with the combined rate of excretion and decay, then concentration ratios (CR, usually in l/kg) can be assessed for different species–radionuclide combinations (Beresford et al., 2008; Hosseini et al., 2008). The knowledge of CRs – along with the corresponding distribution coefficients (K_d, also in l/kg) – is essential in non-human biota radiation-protection assessment tools like the ERICA

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(Environmental Risk from Ionising Contaminants Assessment and Management) Integrated Approach (Beresford et al., 2007).

A large amount of tritium is released into the biosphere as a result of anthropogenic activity, which can increase the natural tritium concentration locally and in fact, regionally, as well. Currently, the main source of the anthropogenic tritium emission is from nuclear power plants (Okada and Momoshima, 1993; Galeriu and Melintescu, 2010).

Tritiated water emitted with the liquid discharges and washed out from the atmosphere can be taken up by biota. The level of contamination can be estimated by knowing the amount of the accumulated organically-bound tritium (OBT), the dose affecting the biota and the extent of the atmospheric tritium (McFarlane et al., 1979; Hisamatsu et al., 1987; Kim and Han, 1999; Pointurier et al., 2004).

Tritium may occur in three different forms in biota, which affect its subsequent mobilisation. The first is tissue free-water tritium (TFWT), the water molecules filling in the interspace of the tissues and cells. This fraction mainly characterises the tritium concentration of the medium and quickly exchanges with it. Tritium, which is combined with organic materials, can be divided into two other groups. Exchangeable organically-bound tritium (E-OBT) is bound to sulphur, nitrogen or oxygen that can be removed by washing with tritium-free water. The other one is the non-exchangeable organically-bound tritium (NE-OBT) is bound to carbon atoms and therefore exhibits the longer tritium exposure of the biota. (Sweet and Murphy, 1984; Guénot and Bélot, 1984; Diabate and Strack, 1997; Jean-Baptiste et al., 2007; IAEA, 2008).

During an examination of the Rhône River in France, the tritium concentration measured in the fluvial mud was several orders of magnitude higher than in the river, probably due to the organic paint particles with large tritium concentration originating from watch factories (Jean-Baptiste et al., 2007). The sediment is supposed to accumulate tritium from particles containing tritium. However, organic substances in the sediment have been shown to absorb tritium from tritiated water (Turner et al., 2009).

2. Experimental

2.1. Sampling method

To examine the effect of the liquid discharge of Paks NPP water, mud, snails, mussels and two types of fish samples were collected from the Danube beyond the mouth of the warm water channel. Background samples of the same types were also collected upstream of the warm water channel outlet.

Samples were obtained 200 m downstream from the mouth of warm water channel at the right riverbank of the Danube (marked with black star in Fig. 1.) (46°34′N; 18°52′E). Control samples were also collected upstream on the Danube, 20 km north of the Paks NPP (46°44′N; 18°58′E). All samples were collected in September, 2010.

Water samples were collected in 2000 ml plastic (polyethylene; PE) bottles and stored in a dark and cool place until preparation. The sediment samples were taken from a water depth of 30–50 cm and stored in plastic bags until preparation.

In parallel with the samplings above, snail (*Viviparus acerosus*) and swollen river mussel (*Unio tumidus*) samples were also taken from a depth of 30–140 cm. Samples were cleaned from mud (with Danube water) and stored in plastic bags and in hermetically closed pots at −18 °C until preparation. Fish samples were also collected. The sampled species were the carnivorous zander (*Sander lucioperca*) and the omnivorous ide (*Leuciscus idus*). These samples were also stored in plastic bags and in hermetically closed pots at −18 °C until preparation.

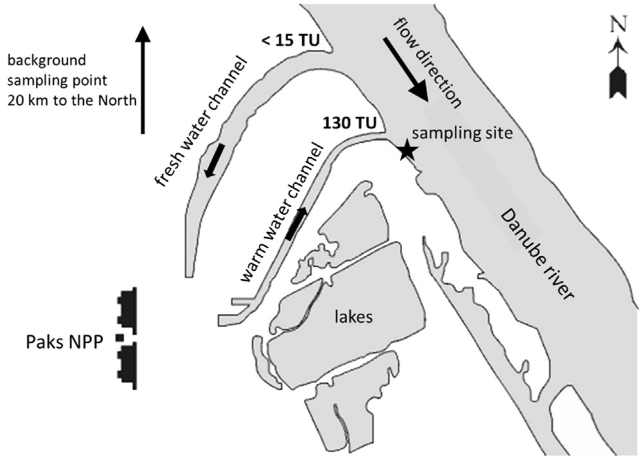

Fig. 1. Map of the sampling area (Paks NPP: Paks Nuclear Power Plant).

2.2. Sample processing

Biological samples were thawed to a semi-frozen state for processing. Snail and mussel samples were divided into shell and soft body components. The whole bodies of the fish samples were cut into small pieces. Sediment and shell samples were dried at 90 °C, in a drying oven. Then the samples were disaggregated/ground and passed through a 2 mm sieve. Macroscopic plant remnants (from sediment samples) were removed during processing.

To separate the OBT and TFWT fractions of the biological samples, freeze-drying method was most frequently used (Garland and Ameen, 1979; Kim and Baumgartner, 1988; Jean-Baptiste et al., 2007). After homogenization, the samples were placed into a lyophilisation desiccator constructed in our laboratory (Fig. 2).

Two water traps were attached to the desiccator in series. The significant part of the water released is collected by the first isopropyl-alcohol trap frozen at −60 °C using a Cryocool$^{®}$ device. The second trap is cooled to −196 °C with liquid nitrogen, and is used for trapping traces of water which pass through the first trap. The vacuum system was pumped by a rotary vane pump.

In all cases, 1–1.5 kg samples were prepared. After drying, the desiccator was heated to 70 °C for 4 h to ensure that all the TFWT was released. During vacuum distillation, 0.8–1.3 L water was obtained from each sample and the entire amount was used for measurement. The dry organic matter remaining in the desiccator was finely ground and stored in a hermetically closed PE pot for gamma-spectrometry and T-OBT measurements.

Sample processing was performed in a low-tritium background laboratory (<10TU), as previous studies report that the tritium content from the humidity of the laboratory can contaminate the samples (Garland and Ameen, 1979; Garland and Cox, 1982; Kozak, 1982; Rank, 1987).

2.3. Determination of the hydrogen concentration of the organic material

In order to compare the tritium concentration of the river water and TFWT with the tritium concentration bound in the biota tissues, the specific hydrogen concentration of each sample was needed. To determine the specific hydrogen concentration, a CHN analyser is usually used (Jean-Baptiste et al., 2010). For OBT measurements with the LSC technique, organic samples are combusted in dry oxygen current and the activity of water generated is measured (Bogen et al., 1973; Takashima et al., 1987; Lockyer and Lally, 1993; Pointurier et al., 2003). In our case, the determination