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The distribution of tritium in aquatic environments, Lithuania

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ABSTRACT

The aim of this study is to investigate mobile radionuclide tritium (^{3}H or T) activity dynamics in aquatic environments related to Ignalina NPP (INPP) site and water bodies located in remote areas unaffected by the INPP. The ^{3}H excess in the INPP environment was analyzed and compared to the variable ^{3}H background level over the period of operation of the INPP (end of 1983 – end of 2009) and during the initial stage of decommissioning (2010–2017). ^{3}H in the INPP vicinity has been studied in the water of artificial channels related to operation of the INPP and site drainage, in natural surface water bodies and, at a smaller scale, in unconfined groundwater. This study presents an extensive ^{3}H data set extending back to 1980, i.e. before INPP operation started. To assess the contribution of global sources to ^{3}H dynamics, monthly precipitation was also studied, along with water from the Baltic Sea, Curonian Lagoon and Nemunas River were studied as well, all three of these located in the Lithuanian maritime zone. The ^{3}H activity concentration in water was measured using liquid scintillation counting (LSC) techniques (direct counting and counting after enrichment). During the period of INPP operation, ^{3}H from liquid effluent could be clearly observed in discharge channels, occurring in rather low diluted conditions, as well as in Lake Druksiai, the cooling basin, at an even more diluted level. The highest ^{3}H activity concentration in Lake Druksiai was observed in 2003 and reached 201.3 ± 1.3 TU at a time when ^{3}H activity concentrations in background water bodies was 9.2 ± 3.5 TU. After the closure of the INPP, the ^{3}H liquid effluent rate reduced by approximately two orders of magnitude (from 10^{12} Bq in 1991 to 10^{10} Bq in 2016) and when decommissioning activity commenced then the ^{3}H activity concentration fell to that approaching the background level (19–27 TU) that can still be observed in industrial discharge and rainwater drainage channels. ^{3}H as a result of leakage from the INPP can be observed in groundwater only in direct proximity to the INPP site near the radioactive waste storage zone.

1. Introduction

The operational history of the Ignalina Nuclear Power Plant (INPP) in Lithuania was very short, this being for different reasons, but mainly political. During the 26 years of operation, the INPP produced 307.9 billion kWh of electricity (Unit 1–136.9 billion kWh and Unit 2–170.2 billion kWh), this accounting for 80% of Lithuania's electricity generating capacity during certain periods. The total amount of electricity sold was 279.8 billion kWh (http://www.iae.lt). The INPP consists of two RBMK-1500 reactor units, Unit 1 and Unit 2 (Almenas et al., 1998). The "1500" refers to the designed electrical power in units of megawatts (MW). Its designed thermal rating is 4800 MW. The nominal thermal power is 4250 MW, and the nominal electrical power is 1300 MW. The RBMK is a graphite-moderated boiling water channel-type reactor with the principle of electricity generation the same as for boiling water reactors (BWRs). Unit 1 was in operation from December 31, 1983 to December 31, 2004, and Unit 2 from August 31, 1987 to December 31, 2009. The INPP is sited in the north-eastern part of Lithuania near the borders of Belarus and Latvia on the southern coast of the Lake Druksiai (Fig. 1). The biggest cities close to the INPP are Vilnius and Daugavpils (Latvia), these being at distances of 130 km and 30 km respectively. The INPP uses Lake Druksiai as a natural reservoir for cooling water and for accommodation of drainage water discharged from the industrial site. Lake Druksiai is a flow-through lake with six small creeks flowing in and one river, with a water regulation dam, flowing out.

Tritium (^{3}H or T) is one of the radioactive components of liquid and gaseous discharges of nuclear power plants (NPPs). ^{3}H is a radioactive isotope of hydrogen with a half-life of 12.32 years decaying to ^{3}He by emitting low energy beta radiation with an average energy of 5.7 keV and a maximum energy of 18.6 keV (Lucas and Unterweger, 2000). In nuclear power reactors, ^{3}H is produced in many reactions with rates dependent on the type of the reactor and its capacity (IAEA, 2004). Additionally, ^{3}H is also produced by nuclear reactions that occur

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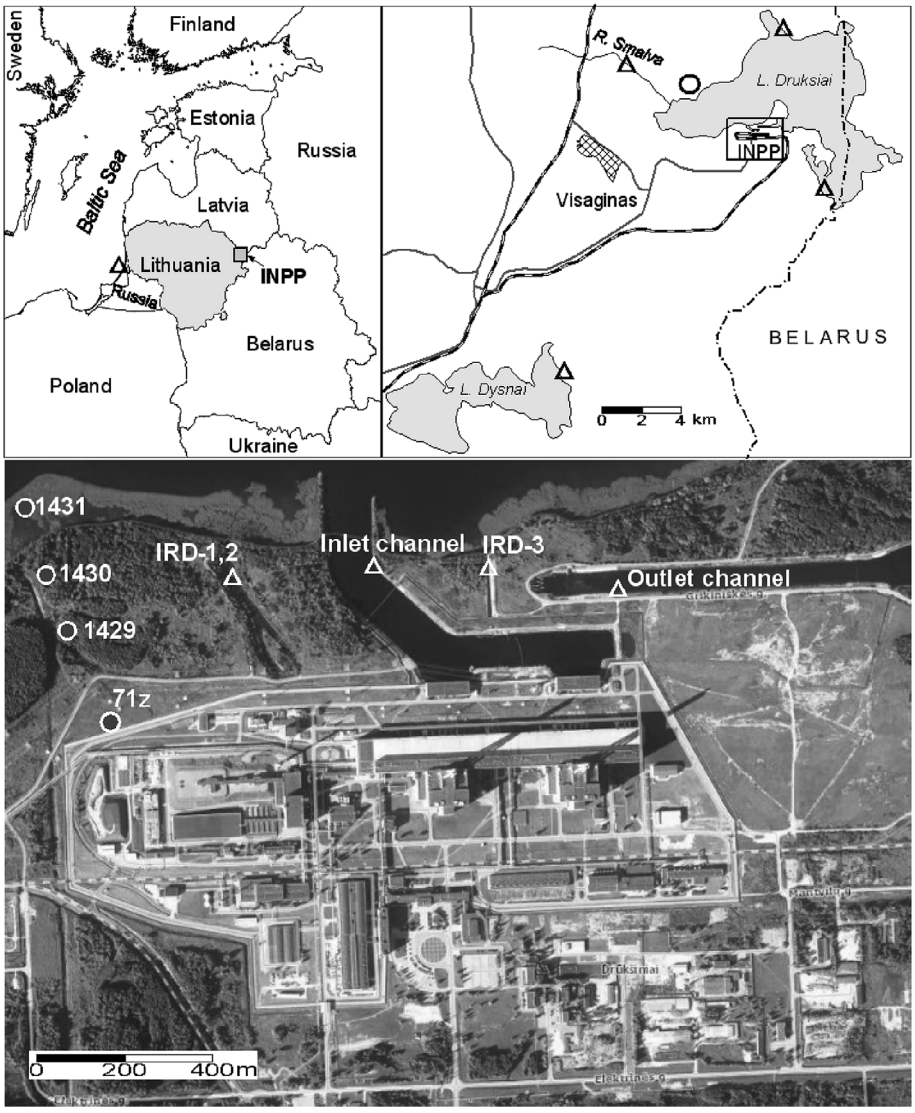

Fig. 1. Sampling locations (triangles in the top schemes represent the approximate position of surface water sampling points in the Baltic Sea, Curonian Lagoon, the lower reaches of the Nemunas River and in background water bodies in the INPP vicinity; the circle in the top right scheme represents the position of the background groundwater well No 17, Budiniai; the bottom scheme represents part of the INPP industrial site with the positions of groundwater wells and sampling points in channels as reworked from http://maps.lt/map/default.aspx).

naturally in the environment and in nuclear weapon testing. Natural ^{3}H is continuously generated by the interaction of high energy cosmic rays with oxygen and nitrogen atoms in the upper atmosphere. These processes produce most of the world's natural ^{3}H, which is subsequently included in very mobile chemical forms as HTO vapour, gaseous HT and CH_3T. A significant part of the natural ^{3}H converts into water and reaches the Earth's surface as atmospheric precipitation. An estimated production rate of 1.48×10^{18} Bq/year results in a world steady state natural inventory of 2.59×10^{18} Bq (Grimes et al., 1982; NCRP Report No. 62, 1989). Following the nuclear weapon tests of early sixties, there was a significant release of artificial ^{3}H into the atmosphere and the ^{3}H content of precipitation in the northern hemisphere increased 1000-fold (Begemann and Libby, 1957; Martell, 1963). The input of ^{3}H to the environment was about 2.96×10^{20} Bq, most of which has now decayed, but approximately 1.85×10^{19} Bq is still present in the environment, mainly in a much diluted state in the world oceans (IAEA, 2004).

Since 1963, the ^{3}H content of precipitation has decreased to a natural level during winter and about twice the natural level during summer. Since that time, the local anthropogenic releases of ^{3}H from the nuclear industry including reprocessing plants and other uses of tritiated materials take continuously place.

Processes of ^{3}H production in nuclear reactors are reported in detail by the IAEA (1981). ^{3}H is produced in the fuel, core components and coolant and is distributed wherever gas or fluid streams take place in the NPP. ^{3}H is produced in all nuclear reactors as a by-product of ternary fission, which is the largest source of its production in power reactors. ^{3}H is additionally produced in reactors by neutron activation of ^{2}H, ^{3}He, ^{6}Li and ^{10}B, and is thus formed in many reactor components (McKay, 1979).

Leakage of NPP systems allows for an eventual release to the environment. The normalized ^{3}H releases from BWRs between 1975 and 1989 are averaged to 1.85×10^{12} Bq·GW(e)$^{-1}$·a^{-1} as a gaseous effluent and 3.70×10^{12} Bq·GW(e)$^{-1}$·a^{-1} as a liquid effluent (IAEA, 2004).

Under normal operation of the INPP, the prevailing radionuclide in liquid releases by activity was ^{3}H, with annual release rate of $\sim 10^{12}$ Bq/year. After the closure of the INPP for decommissioning, the ^{3}H liquid release rate was reduced by two orders of magnitude, though remained observable and variable due to periodic discharges of bypass water (Fig. 2). Annually released activity of other radionuclides to aquatic environment from INPP was usually in range of 10^{7}–10^{8} Bq (http://europa.eu/radd/index.dox). The estimated values of release rates of radionuclides to environment are usually reported with large uncertainties due to several reasons. Firstly, estimates derived from environmental data depend on very complex environmental processes which are oversimplified in model approaches. Secondly, data based on direct routine monitoring of release rates take into account only main release points and pathways and are sensitive to time resolution.