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Research article

Comparison of balance of tritium activity in waste water from nuclear power plants and at selected monitoring sites in the Vltava River, Elbe River and Jihlava (Dyje) River catchments in the Czech Republic

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A B S T R A C T

During the routine operation, nuclear power plants discharge waste water containing a certain amount of radioactivity, whose main component is the artificial radionuclide tritium. The amounts of tritium released into the environment are kept within the legal requirements, which minimize the noxious effects of radioactivity, but the activity concentration is well measurable in surface water of the recipient. This study compares amount of tritium activity in waste water from nuclear power plants and the tritium activity detected at selected relevant sites of surface water quality monitoring. The situation is assessed in the catchment of the Vltava and Elbe Rivers, affected by the Temelín Nuclear Power Plant as well as in the Jihlava River catchment (the Danube River catchment respectively), where the waste water of the Dukovany Nuclear Power Plant is discharged. The results show a good agreement of the amount of released tritium stated by the power plant operator and the tritium amount detected in the surface water and highlighted the importance of a robust independent monitoring of tritium discharged from a nuclear power plant which could be carried out by water management authorities. The outputs of independent monitoring allow validating the values reported by a polluter and expand opportunities of using tritium as e.g. tracer.

1. Introduction

Nuclear energy can play important role regarding sustainable energy future. Its potential to contribute to reduction of greenhouse gases could be significant (NEA, 2016). On the other hand, the radionuclides from a nuclear facility can be present through its surroundings and radiological contaminants can be released into the environment secondarily, e.g. while present in forest debris which is used as a fuel (Hejl et al., 2013).

The operation of nuclear power plants is accompanied with raised public awareness and with high requirements to keep radiation protection (Corner et al., 2011). This is understandable not only because past accidents at nuclear power facilities. These accidents and their consequences are well described in scientific literature; e.g. Fukushima accident in (Konoplev et al., 2016), Chernobyl accident in (Smith and Beresford, 2005) and accidents in the Southern Urals in (Chebotina and Nicolin, 2006) or in (Soyfer, 2002).

The everyday operation of a nuclear power plant is associated with production of different radionuclides that are partially emitted into the environment in very low concentrations. High attention is paid to tritium. It is emitted with waste waters from nuclear power plants globally. Tritium is a clear beta-emitter. The discussions concerning tritium emissions are evoked by the fact that activities of tritium released into surface water and other recipients from nuclear power plants are relatively high; they exceed releases of other radionuclides by several orders of magnitude. Elevated tritium values in natural waters were reported from several countries with technogenic emissions – e.g. Germany (Osman et al., 2016), France (Eyrolle-Boyer et al., 2015) and Russia (Chebotina et al., 2015). This leads to the conclusion that independent monitoring of tritium is essential especially in fresh waters.

Past atmospheric tests of nuclear weapons and tritium of natural origin are main sources of tritium in the environment. Tritium is permanently produced in upper layers of the atmosphere by

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nuclear reactions caused by cosmic rays. Production of tritium by natural processes is estimated 72 PBq/y and the tritium quantity of natural origin is constantly at a level of 1275 PBq. Tritium concentrations are still affected by the test of nuclear weapons in the atmosphere: It has been estimated that 186 EBq (i.e. 186 $\times$ 10^{18} Bq) was emitted into the environment (UNSCEAR, 2000). Projecting these data, it can be stated that the remaining tritium activity in 2016 was still 9.6 EBq. By 2051, the remaining tritium activity will be identical to that originating from natural processes (1.3 EBq).

Tritiated water is most common form of tritium occurrence in the environment. The human health risks related to tritium can enter human body by inhalation, ingestion, or absorption through skin. Tritiated water can be cleared from the human body relatively rapidly (biological half-life about 10 days), the retention time for organically bound tritium may be longer (Hunt et al., 2009). The human health risks related to tritium in water are very low (Dingwall et al., 2011). However some studies proposed that tritium can cause radiogenic effects in living organisms (Straume and Carsten, 1993).

Thanks to its occurrence in environment and its favourable properties, tritium can be used as a radiotracer. Tritium from a nuclear power plant was used as a radiotracer for pollutant transport modeling e.g. in Ebro River (Pujol and Sanchez-Cabeza, 2000). It is also widely used for estimation of groundwater mean residence and in modeling of pollutant transport in groundwater (Cox et al., 2013) or for determination of origin of waters (Gorr and Genc, 2012).

There are currently two nuclear power plants operating in the Czech Republic: the Nuclear Power Plant Temelín (NPP Temelín) and the Nuclear Power Plant Dukovany (NPP Dukovany). Impact of these NPPs on the environment has been assessed previously. It was described in several studies. The impact of NPP Temelín was described e.g. in (Hanslík et al., 2013) and in (Ivanovová and Hanslík, 2010). The occurrence of tritium in different matrices including organic tissues in vicinity of NPP Dukovany was studied by (Kovaříková et al., 2016).

Usually, the wastewater from a NPP is discharged into a large stream or ocean, where the dilution is substantial, e.g. as in Spain (Pujol and Sanchez-Cabeza, 2000), France, USA or Russia. However, the wastewater from the Czech NPPs is discharged in relatively small watercourses. Regarding the expected climate change impacts in the Czech Republic including the hydrological drought (e.g. (Potop et al., 2012), the tritium activity concentration has potential to increase in the rivers affected directly by NPPs. Additionally, the both NPPs influence surface water in tributaries of big European rivers: NPP Temelín through Vltava River influences Elbe River and NPP Dukovany influences Danube River through Jihlava River. Regarding the impact of the Czech NPPs on the environment, the elevated tritium activity levels in plant tissues as compared to background samples was reported for NPP Dukovany (Kovaříková et al., 2016). The increased tritium levels in Danube water after 1989 were interpreted to be connected with newly operating NPP Temelín (Maringer et al., 2004). Consequently, there is obvious need for robust independent monitoring of radioactive pollutants in affected recipients.

The objective of this study is to assess the potential of independent monitoring of tritium activity discharged from two nuclear power plants in the Czech Republic. The independent monitoring was based on tritium observations in surface water and it could be carried out by water management authorities. The outputs of independent monitoring allow validating the values reported by a polluter (NPPs) and expand opportunities of using tritium as e.g. tracer.

2. Data and methods

Balance of tritium activity was calculated using the results of monitoring of tritium concentration at sites both affected and unaffected by discharge of waste water from the NPP Temelín and the NPP Dukovany (Fig. 1). Annual water flow rates were provided by the Czech Hydrometeorological Institute (CHMI). Annual discharges of tritium activity were provided by the CEZ Group, the operator of both NPPs.

The samples for determination of tritium activity concentration were taken with the frequency of 12 times a year by river basin administrators: Povodí Vltava, State Enterprise, Povodí Labe, State Enterprise and Povodí Morava, State Enterprise. Tritium activity was measured by the Department of Radioecology of T. G. Masaryk Water Research Institute, p.r.i.

In relation to the NPP Temelín, the unaffected sites were on the Vltava River at Hluboká and on the Elbe River at Lysá; the affected sites were on the Vltava River at Solenice and at Prague-Podolí and on the Elbe River at Hřensko. The sites were monitored from 2002 to 2015. In relation to the NPP Dukovany, the unaffected site was on the Jihlava River at Vladislav and the affected sites were on the Jihlava River below the Mohelno reservoir and on the Dyje River at Pohansko. The sites were monitored from 2003 to 2015. Tritium occurrence at the unaffected sites represents the background of tritium in surface water.

Then, the balance of tritium activity (corrected by the background component obtained from the monitoring of surface water) was compared with the data provided by CEZ Group. The data included tritium discharge in the given year for both NPPs.

Determination of tritium concentration in the surface water samples was carried out according to the CSN EN ISO 9698 (Czech Office for Standards and Testing, 2011). We used ultralow level liquid scintillation spectrometers Quantulus 1220 by WALLAC and TriCarb 3170/TRSL by Canberra Packard. Since 2010, the samples from unaffected sites on the Vltava River at Hluboká and the Elbe River at Lysá have been electrolytically enriched. Measurement conditions were set so that the minimum detectable activity, c_{ND}, was 2.0 Bq/l for the affected samples, 1.0 Bq/l for the unaffected samples and 0.08 Bq/l for the electrolytically enriched samples.

Annual average values of volume activity were calculated. In the case of values lower than c_{ND}, a value equal to c_{ND} was used for the calculation of the average values instead.

For description of tritium concentration development at the unaffected sites, a first-order kinetics equation was used:

$$\ln c_{i,j} = -\lambda_{eff} \cdot t + q \qquad (1)$$

where.

$c_{i,j}$ - is the annual average activity of tritium in surface water at the unaffected site i in year j, representing the background tritium activity (contamination after nuclear tests, tritium creation by natural processes and releases from nuclear facilities abroad), based on the results of the monitoring (Bq/l)
λ_{eff} - effective (observed) decay constant for the decrease of tritium concentration from atmospheric tests of nuclear weapons (1/y)
t - years of monitoring (y).

Statistical significance of dependence (regression curve) was verified using the Pearson coefficient.

Effective half-life (T_{eff}) of tritium concentration decrease in surface water at the unaffected sites was calculated from the assessed effective decay constant (λ_{eff}) using the equation according (Smith and Beresford, 2005):