KEY RADIONUCLIDES IN THE AQUATIC ENVIRONMENT OF IGNALINA NPP

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INTRODUCTION

Under normal operation of the Ignalina NPP various radionuclides (137Cs, 134Cs, 54Mn, 58Co, 60Co, 59Fe, 51Cr, 95Zr, 95Nb, 131I, 90Sr, 3H) are discharged to aquatic system. Prevailing radionuclide in waterborne releases is 3H with annual activity of ∼1012 Bq, annually released activity of other radionuclides is usually in range of 10⁷ – 10⁸ Bq [1]. In water transfer processes very important are specific mobile radionuclides – tritium (H) and radiocarbon (14C) adding to the global artificial (from thermonuclear weapon testing) and natural (from cosmogenic production) inventories of those radionuclides in the Earth. 3H is a part of water molecule, and 14C is initially a part of atmospheric CO₂ and consequently is transformed into different forms of inorganic or organic carbon in the terrestrial and aquatic ecosystems [2, 3]. In the water systems prevailing chemical form of 14C is dissolved inorganic carbon (DIC), while small fraction from the total 14C activity can be in the form of dissolved organic carbon (DOC).

The main goal of this study is to give closer look on mobile radionuclides (H and 14C) activity dynamics in aquatic environment of INPP during the whole period of NPP operation and to compare 3H and 14C activity level with one of other radionuclides (137Cs, 90Sr, 60Co).

METHODS AND MATERIAL

The activity concentration of 14C in environmental samples during all observation period was...
measured by liquid scintillation spectrometry as described in [4, 5]. A conventional method for synthesis of benzene from DIC in water or from C\textsubscript{org} of biota was applied. The uncertainty of the results (statistical radiometric error) was reported at 1 sigma-level. The quality of 14C determinations was periodically tested through participating in various inter-comparisons. The results of Intercomparisons (FIRI, VIRI), organized by the University of Glasgow, indicated that most of the 14C results are of normal (3%) precision, with some of high (1.5%) precision. The 3H activity concentration in surface water and groundwater during all observation period is measured by the same liquid scintillation spectrometer. The normal precision of 3H measurements (direct counting and counting after enrichment) in the water was recognized during the IAEA Inter-comparisons of low-level tritium measurements in water (TRIC2004, TRIC2008). After water samples (20-40 l) evaporation gamma ray emitting radionuclides were detected in solid residual by high resolution gamma spectrometry and after radiochemical procedures on the same solid residual 90Sr was measured by liquid scintillation counting as described in [6, 7, 8].

The measurements of 3H and 14C activity concentration in water of the Lake Drūkšiai have been already started before Ignalina NPP (INPP) operation [9, 10, 11]. After the start of INPP operation the new monitoring points on surface water bodies related to INPP industrial site were established: cooling water inlet channel (IC), heated water outlet channel (OC), industrial discharge and rain water drainage (IRD) channels 1, 2, and 3 (Fig. 1).

![Fig. 1. Observation points on the main channels related to INPP industrial site](image1)

The water samples for 3H and 14C measurements were usually taken 1-2 times a year. Only during the period of 2003-2004 3H activity in water was measured almost every month. Because of the complicated methods of 14C determination, the samples for the 14C measurement in this period were taken only 1-2 times a year. Activity concentration of other radionuclides (90Sr, 60Co, 137Cs) in water from surface water bodies were measured rarely. The measurements of 3H and 14C activity concentration in groundwater started during INPP pre-operation period. The general view of groundwater monitoring network for recent years is shown in Fig. 2.

3H and 14C activity concentration was also measured in other environment objects such as birch sap, aquatic and terrestrial plants and bottom sediments.

**RESULTS**

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3H and 14C activity concentration was also measured in other environment objects such as birch sap, aquatic and terrestrial plants and bottom sediments.
Visuomenės sveikata
Originalūs straipsniai
(Lake Drūkšiai till 1984, Lake Dysnos and Smalvelė river for later years) is decreasing with time. This is the evidence of the excess of \(^3\)H originated from thermonuclear weapon tests decline almost to the level of \(^3\)H activity in water bodies corresponding to cosmogenic production of \(^3\)H. Data being above a background line display an increase of \(^3\)H activity concentration originated from the radioactive effluents released by INPP in normal operation mode. For the period of 1980-2009 the highest \(^3\)H activity concentration in Lake Drūkšiai was observed in 2003 year and reached 24 Bq/l. \(^3\)H activity concentration in Lake Skripkos related to Visaginas wastewater treatment plant was the highest in 2000 year and reached 30 Bq/l. During this period \(^3\)H activity concentration in the background water bodies was 2-3 Bq/l, so approximately 20-25 Bq/l was originated from INPP releases (Fig. 3).

The \(^3\)H monitoring points of surface water bodies - cooling water inlet channel (IC), heated water outlet channel (OC), industrial rain drainage (IRD) channels 1,2, channel 3 has been systematically measured since 1992. The same \(^3\)H background line has been applied to these data. \(^3\)H activity concentrations in channels exceed the background level during all period of observation (Fig. 4).

### Table 1. \(^{14}\)C activity concentration in aquatic plants and bottom sediments of INPP area in 2007-2008

<table>
<thead>
<tr>
<th>Sampling point location</th>
<th>Species or material</th>
<th>Sampling date</th>
<th>(^{14})C activity relative to modern carbon, pMC(\pm 1\sigma^*)</th>
<th>(^{14})C activity concentration Bq/kg(\pm 1\sigma)</th>
</tr>
</thead>
<tbody>
<tr>
<td>INPP, outlet channel (OC)</td>
<td>Myriophyllum spicatum</td>
<td>28-08-2008</td>
<td>127.7(\pm 0.6)</td>
<td>129.9(\pm 0.6)</td>
</tr>
<tr>
<td>IRD-1,2</td>
<td>Ceratophyllum demersum</td>
<td>28-08-2008</td>
<td>172.9(\pm 7.7)</td>
<td>175.8(\pm 7.8)</td>
</tr>
<tr>
<td>Lake Drūkšiai, 1 station</td>
<td>Myriophyllum spicatum</td>
<td>27-06-2007</td>
<td>119.3(\pm 1.9)</td>
<td>121.3(\pm 1.9)</td>
</tr>
<tr>
<td>Lake Drūkšiai, 1 station</td>
<td>Ceratophyllum demersum</td>
<td>24-09-2008</td>
<td>118.8(\pm 1.0)</td>
<td>120.8(\pm 1.0)</td>
</tr>
<tr>
<td>Lake Drūkšiai, 4 station</td>
<td>Myriophyllum spicatum</td>
<td>27-08-2008</td>
<td>125.9(\pm 0.7)</td>
<td>128.0(\pm 0.7)</td>
</tr>
<tr>
<td>Lake Drūkšiai, 7 station</td>
<td>Ceratophyllum demersum</td>
<td>27-08-2008</td>
<td>123.4(\pm 0.9)</td>
<td>125.5(\pm 0.9)</td>
</tr>
<tr>
<td>Lake Drūkšiai, 1 station</td>
<td>Ceratophyllum demersum</td>
<td>27-06-2007</td>
<td>91.9(\pm 1.6)</td>
<td>28.3(\pm 0.5)</td>
</tr>
<tr>
<td>Lake Drūkšiai, 4 station</td>
<td>Sediments</td>
<td>27-06-2007</td>
<td>92.4(\pm 0.8)</td>
<td>12.5(\pm 0.1)</td>
</tr>
<tr>
<td>Lake Drūkšiai, 6 station</td>
<td>Ceratophyllum demersum</td>
<td>27-06-2007</td>
<td>113.6(\pm 1.3)</td>
<td>15.4(\pm 0.2)</td>
</tr>
<tr>
<td>Lake Drūkšiai, 7 station</td>
<td>Ceratophyllum demersum</td>
<td>27-08-2008</td>
<td>90.6(\pm 3.5)</td>
<td>30.4(\pm 1.2)</td>
</tr>
<tr>
<td>Lake Drūkšiai, 7 station</td>
<td>Sediments</td>
<td>27-06-2007</td>
<td>102.3(\pm 1.8)</td>
<td>1.3(\pm 0.0)</td>
</tr>
</tbody>
</table>

* 100 pMC - \(^{14}\)C level in the environment before thermonuclear weapon testing (before 1950); at that time \(^{14}\)C specific activity relative to stable carbon was 226 Bq/kg, or 100 pMC=226 Bq/kg C.
** For modern air-dried organics carbon makes 45% from dry weight, for older organics of bottom sediments carbon makes 60% from dry weight. These data are based on measured fraction of organics and measured fraction of C\textsubscript{org} in the bottom sediments.

### Table 2. \(^{90}\)Sr, \(^{137}\)Cs and \(^{60}\)Co activity concentration in groundwater of INPP area in 2007-2008

<table>
<thead>
<tr>
<th>Sampling point location</th>
<th>Sampling date</th>
<th>(^{90})Sr, Bq/m(^3)(\pm 1\sigma)</th>
<th>(^{137})Cs, Bq/m(^3)(\pm 1\sigma)</th>
<th>(^{60})Co, Bq/m(^3)(\pm 1\sigma)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Well 1429</td>
<td>28-06-2007</td>
<td>33(\pm 4)</td>
<td>1.5(\pm &lt;1)</td>
<td>1.5(\pm &lt;1)</td>
</tr>
<tr>
<td>Grinkiniškės, piezometer (G.P.)</td>
<td>29-06-2007</td>
<td>1.0(\pm 0.3)</td>
<td>1.4(\pm &lt;1)</td>
<td>1.5(\pm &lt;1)</td>
</tr>
<tr>
<td>Well 35955</td>
<td>29-06-2007</td>
<td>30(\pm 4)</td>
<td>1.1(\pm &lt;1)</td>
<td>1.1(\pm &lt;1)</td>
</tr>
<tr>
<td>Well 40035</td>
<td>28-06-2007</td>
<td>1(\pm &lt;1)</td>
<td>1.0(\pm &lt;1)</td>
<td>1.0(\pm &lt;1)</td>
</tr>
<tr>
<td>Well 40036</td>
<td>09-25-2008</td>
<td>&lt;0.2(\pm &lt;0.2)</td>
<td>&lt;2(\pm &lt;2)</td>
<td>&lt;2(\pm &lt;2)</td>
</tr>
<tr>
<td>Well 35221</td>
<td>29-09-2008</td>
<td>1.4(\pm 0.3)</td>
<td>0.2(\pm &lt;0.2)</td>
<td>&lt;0.2(\pm &lt;0.2)</td>
</tr>
<tr>
<td>Well 35221</td>
<td>29-06-2007</td>
<td>1(\pm &lt;1)</td>
<td>2.1(\pm 1.7)</td>
<td>11.5(\pm 1.7)</td>
</tr>
<tr>
<td>Well 35221</td>
<td>26-09-2008</td>
<td>0.5(\pm 0.1)</td>
<td>3(\pm &lt;3)</td>
<td>11.0(\pm 2.0)</td>
</tr>
</tbody>
</table>
The flow rate in the industrial rain drainage (IRD) channels 1,2, and especially in channel 3 is low compared to flow rate in the heated water outlet channel. Therefore radioactive effluents in IRD channels are less diluted and more variable in terms of $^3$H activity concentration compared to whole Lake Drūkšiai water body. The $^3$H variations were considered with the higher temporal resolution in 2001-2004 when the samples were taken more frequently – about 1 time per month. At that time $^3$H activity concentration varied from 10 to 50 Bq/l in the water from channel 1,2.

$^{14}$C activity measurements in DIC of surface water bodies with different frequency have been performing since 1975. $^{14}$C activity concentration in background water bodies well fits with the international data for Northern Hemisphere. The excess of $^{14}$C originated from thermonuclear weapon tests declines almost to the $^{14}$C level of cosmogenic origin for all studied surface water bodies. From the period of 1992-1993 $^{14}$C of cosmogenic origin predominates in the atmosphere and in the surface water all over the world. Almost for all period of $^{14}$C monitoring in surface water the influence of INPP has been hardly estimated (Fig. 5). Since 2002 only the $^{14}$C excess in water caused by INPP was observed. The traceable $^{14}$C activity increase was observed in 2005. It was possible to notice by precise methods of $^{14}$C measurement.

$^3$H activity concentrations most systematically were measured in the groundwater from monitoring wells 71z, 1429, 1430, and 1431, which are close to INPP. These monitoring wells form profile according to groundwater flow direction from the INPP to the Lake Drūkšiai. The period of $^3$H observation in groundwater was from 1987 to 2009 (Fig. 6).

At the same time samples were taken from the background observation well Būdiniai, 17. This well is located on the northern shore of the Lake Drūkšiai and is not related to INPP industrial site. $^3$H activity concentration was confidently higher than background level only in observation piezometer 71z, which is closest to INPP. The highest $^3$H activity (18.3 Bq/l) was measured...
in 2001. \(^{3}H\) background level at that time was about 2 Bq/l. In down-flow direction from INPP the level of \(^{3}H\) activity in groundwater is decreasing and mostly is very close to the background level. The \(^{3}H\) concentration in the observation well 1431 installed very close to the lake was somewhat higher than background level when there was inflow to the well from the Lake Drūkšiai.

There are less data on \(^{14}C\) measurements in groundwater compared to \(^{3}H\) data. The sampling (large water volume is required) and analytical procedures (benzene synthesis) of \(^{14}C\) determinations are more complex compared to that for \(^{3}H\). For 1987-2008 period \(^{14}C\) activity concentration measurements in groundwater are presented in Fig. 7.

As the main chemical form of \(^{14}C\) in groundwater is the bicarbonate ion, therefore \(^{14}C\) activity concentration is approximately calculated from two experimentally measured values such as \(\text{HCO}_3^-\) concentration and \(^{14}C\) specific activity relative to carbon. The \(^{14}C\) activity in groundwater has never been higher than the global level. The influence of INPP was impossible to notice except one quarry observation well.

**DISCUSSION**

\(^{3}H\) activity concentration in the background water body (Smalvelė river) was 1.6±0.3 Bq/l in 2007-2009, meantime in the Lake Drūkšiai it was 4.2 – 13.2Bq/l. Water in lake is well mixed because of natural and anthropogenic processes (pumping and thermal discharges). Therefore \(^{3}H\) distribution was even in the whole lake water volume with some what elevated activities close the mouth of IRD 1,2. As before \(^{3}H\) activity concentrations in water from IRD channels were elevated and reached 13.6 – 36.9 Bq/l (see Fig. 4).

\(^{14}C\) activity concentration in water from the Lake Drūkšiai and from cooling water inlet channel has increased in 2001-2006, while decreased again in 2007. The highest activity of \(^{14}C\) was in 2005 – 13.6±0.2 Bq/m³ while background level was about 10.0±0.2 Bq/m³. The increase of \(^{14}C\) activity was about 3.6 Bq/m³. The \(^{14}C\) activity reduced in later years and reached 9.5 Bq/m³.

\(^{3}H\) activity concentrations in groundwater ranged between 0.5 and 7.7 Bq/l in 2007-2009. From all of observation wells \(^{3}H\) activity concentration raised only in observation well 71z, which is closest to INPP, and well 35220, which is in the center of the quarry (Fig. 2). The increase of \(^{3}H\) activity is related to \(^{3}H\) transfer from the INPP industrial site. In the other observation wells \(^{3}H\) activity is mostly caused by the global sources and local features of water mixing in unsaturated zone and in shallow groundwater.

In 2007-2008 in the quarry observation well 35221 \(^{14}C\) activity level was somewhat higher than the activity caused by global sources. \(^{14}C\) activity here was 40.4±0.4 Bq/m³. In previous period before re-cultivation in that quarry the sludge from the Visaginas wastewater treatment plant was disposed off.

The activities of \(^{90}Sr\) (<MDA to 33 Bq/m³) and \(^{137}Cs\) (<MDA to 1.4 Bq/m³) in surface and ground water were often very low. Only in the water from quarry observation well (No 35221) activity concentration of \(^{60}Co\) was determined around 11 Bq/m³.
**CONCLUSIONS**

By continuous observations using modern experimental techniques it is possible to observe $^3$H and $^{14}$C fraction originated from INPP and dispersed in the environment which already is traced by globally distributed $^3$H and $^{14}$C.

$^3$H released from INPP can be observed in IRD channels in low diluted conditions and even in the Lake Drūkšiai. $^3$H released from INPP can be observed in groundwater in fence zone near the radioactive waste storage area only.

Very insignificant fraction of $^{14}$C originated from INPP can be observed in the channels and in the Lake Drūkšiai. Only in 2005 $^{14}$C activity in water from outlet channel compared to background level has increased about 30%. It is impossible to evaluate $^{14}$C activity originated from INPP in the groundwater except one quarry observation well. In this quarry well $^{14}$C activity was about 30% higher than background level.

The activities of $^{90}$Sr and gamma ray emitting radio-nuclides in surface and ground water were very low and mostly less than minimal detectable activity (<MDA). Very insignificant activity concentration of $^{60}$Co was observed in the water from quarry observation well.

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Būdingieji radionuklidai Ignalinos AE aplinkos vandens terpėse

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Santrauka

Tyrimų tikslas – atskleisti mobiliųjų radionuklidų (3H ir 14C) aktyvumų dinamiką su Ignalinos AE susijusiuose vandens telkiniuose per visą AE eksploatacijos laikotarpį, palyginti mobiliųjų radionuklidų aktyvumo lygijus su kitų dirbtinės kilmės radionuklidų (137Cs, 90Sr, 60Co) aktyvumų lygiais.

Metodai. Pastaraisiais metais po tam tikros pertraukos, įvairiems su branduoline energetika susijusiems projektams vykdyti prireikus informacijos, vėl atkreiptas dėmesys į Drūkšių ežero ir apylinkių radioekologinės būklės vertinimus. Šiame darbe vertintas kai kurių radionuklidų (3H, 14C, 90Sr, 137Cs ir 60Co) paplitimas Drūkšių ežero, su Ignalinos AE susijusių kanalų ir gruntiniame vandenyje. Specifinis radionuklidas 14C dar papildomai tirtas tarsių saušmos ir vandens biotoje.

Rezultatai. Ignalinos AE kilmės 3H pastebima kanalų aplinkoje, dar palyginti mažai praskiesto vandens sistemėje, ir mažesniu mastu – Drūkšių ežere, dažniausiai šalia lietaus kanalizacijos kanalo (PLK-1,2) žiočių. Ignalinos AE kilmės 3H pastebima tik prie AE perimetro, šalia radioaktyviųjų atliekų saugyklių. 2005 m. 14C tūrinis aktyvumas PLK-1,2 buvo 70 proc. didesnis už fono lygį. Gruntiniame vandenye Ignalinos AE kilmės 14C neįmanoma nustatyti, išskyrus specifinį karjero, į kurį buvo šalinamas dumblas su dirbtiniais radionuklidais, gręžinį. Čia 14C tūrinis aktyvumas gruntiniame vandenye šiek tiek didesnis nei fono lygis (prieaugus iki 30 proc.). Paviršiniame ir gruntiniame vandenye labai maži 137Cs (nuo < MDA iki 1,4 Bq/m³) ir 90Sr (nuo < MDA iki 33 Bq/m³) tūriniai aktyvumai. Tik karjero centre gruntiniame vandenye 60Co buvo apie 11 Bq/m³.

Apibendrinimas. Jeigu minėtų objektų vanduo būtų vartojamas kaip geriamasis, apšvitos dozės būtų gerokai mažesnės už nebekontroliuojamajį lygį – 10 µSv metinę efektinę dozę.

Raktažodžiai: Ignalinos AE, radionuklidai, paviršinis vanduo, gruntinis vanduo.

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