ASSESSMENT OF THE EXTERNAL EXPOSURE DOSE TO HUMANS FROM ^{137}CS AND ^{90}SR IN THE COASTAL WATERS OF THE BALTIC SEA NEAR LITHUANIA

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Abstract. At present the Baltic Sea is considered to be the most contaminated with anthropogenic radionuclides in comparison to any other part of the World Ocean. Anthropogenic radionuclides (mainly ^{137}Cs and ^{90}Sr) found in the seawater are sources of the external exposure that contributes to the total radiation exposure to humans. The variations of activity concentrations of ^{137}Cs and ^{90}Sr in the water of the Baltic Sea near Lithuanian coast in 1985–2013 were analyzed. External exposure dose from these radionuclides to humans due to immersion in the seawater were calculated using the results of the measurements. An average external exposure dose from ^{137}Cs ranged between 6.28 $nSv \cdot h^{-1}$ to 1.5 $nSv \cdot h^{-1}$, from ^{90}Sr – between 1.73 $nSv \cdot h^{-1}$ to 0.53 $nSv \cdot h^{-1}$.
Keywords: *activity concentration,ionizing radiation, radionuclides, radioactivity, marine ecosystem*

Introduction

All living organisms, including humans, are being exposed to ionizing radiation at all times. Natural radiation comes from many sources including more than 60 naturally-occurring radioactive materials found in air, soil and water. Humans are also exposed to natural radiation from cosmic rays. According to the United Nations Scientific Committee on the Effects of Atomic Radiation report (UNSCEAR, 2010), the worldwide average radiation exposure from natural sources is approximately 2.4 mSv per year. However, the exposure to humans from ionizing radiation of natural sources is a continuing and inescapable feature of life on Earth.

Humans exposure to ionizing radiation also comes from anthropogenic sources. ^{137}Cs and ^{90}Sr are one of the most hazardous anthropogenic radionuclides due to their long physical and biological half-life (about 30 years) (IAEA, 2005). Anthropogenic radionuclides are penetrated in the marine environment mostly as a result of nuclear explosions, accidents at nuclear power plants and due to the operation of nuclear industry (HELCOM, 2009; Zalewska and Suplińska, 2013).

Review of Literature

For the first time ^{137}Cs and ^{90}Sr were penetrated into the Baltic Sea with the global fallout as a result of the tests of nuclear weapons in the atmosphere, particularly in the 1950s and 1960s (Livingston and Povinec, 2000; Ikäheimonen et al., 2009; Ilus, 2007). According to data published in Helsinki Commission proceedings (HELCOM, 2013), the total inputs of weapons test ^{137}Cs and ^{90}Sr into the Baltic Sea were 800 TBq and 500

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TBq, respectively. During a long period of time this sea was contaminated by the North Sea waters where radioactive waste was discharged from various nuclear reprocessing plants (HELCOM, 2009; Zalewska and Lipska, 2006; Saniewski, 2013). The increase of the global fallout proceeded up to 1963, when a treaty concerning nuclear and thermonuclear weapon test stopped in the atmosphere, hydrosphere and cosmos were signed. This moment was the beginning of the decrease in radioactive contamination of the marine environment. The process was observed up to 1986. The average activity concentrations of ^{137}Cs and ^{90}Sr in the seawater until 1986 were 12 Bq·m^{-3} and 24 Bq·m^{-3}, respectively (Styro et al., 1990).

The radiological situation in the Baltic Sea changed after the Chernobyl Nuclear Power Plant (ChNPP) accident, which became the main source of ^{137}Cs after 1986 (Ikäheimonen et al., 2009; Juranová et al., 2015). After the accident, an average activity concentration of ^{137}Cs in the surface waters of the Baltic Sea grew by more than an order of magnitude relative to the radioactive background (12 Bq·m^{-3}) formed after nuclear weapons tests in the atmosphere (Styro et al., 1990). According to data calculations (HELCOM, 2013), direct input of ^{137}Cs into the Baltic Sea from ChNPP accident was estimated to be 4700 TBq. The Chernobyl fallout was scattered very unevenly over the Baltic Sea area (Weiss, 2011). The Bothnian Sea and the Gulf of Finland were both regarded as the most contaminated regions of the Baltic (Zalewska and Lipska, 2006). In 1986, the respective average concentrations of ^{137}Cs in these regions were 480 Bq·m^{-3} and 500 Bq·m^{-3}, whereas in the Baltic Proper the average activity concentration of this radionuclide was only 150 Bq·m^{-3} (Zalewska and Suplińska, 2013). Much less contaminated were the Bothnian Bay, the Belt Sea and the southern Baltic Proper where, in 1986, the average concentration was 84 Bq·m^{-3} (Zalewska and Suplińska, 2013). The activity concentration of ^{90}Sr following the accident of the ChNPP changed negligibly (Saniewski, 2013; Zaborska et al., 2014). The direct input of this radionuclide was estimated to be 80 TBq (Ikäheimonen et al., 2009). During a period of time, a leveling of the ^{137}Cs activity concentration in the surface waters took place; the end of this process may be ascribed to 1989, when its activity concentration appeared to be about 150 Bq·m^{-3} (Styra et al., 2008). This time was used as the beginning of self-purification process of the Baltic Sea from ^{137}Cs (Styra et al., 2008). During the period 1992–2010 concentrations of ^{137}Cs in the surface waters decreased in all parts of the Baltic Sea. The average activity concentration of ^{137}Cs in the Baltic Proper decreased to 40 Bq·m^{-3} in 2010. Concentrations in the Western Baltic and in the Gulf of Finland were lower at around 35 Bq·m^{-3} and 29 Bq·m^{-3}, respectively. The highest concentrations were reported in the Archipelago and Aland Sea (equal to 44 Bq·m^{-3}) (HELCOM, 2013). At present, concentrations of ^{137}Cs are relatively uniform in all regions of the Baltic and remain at approximately similar levels mainly because of the transport and mixing of water masses (Zalewska and Suplińska, 2013). ^{90}Sr concentration in the Baltic seawater varied in general from 5 Bq·m^{-3} to 15 Bq·m^{-3}. The ^{90}Sr concentration decreases slowly with time and its behavior in seawater is different from ^{137}Cs (HELCOM, 2013).

The decrease of activity concentrations of these radionuclides in the coastal waters near Lithuania was observed too (Styro et al., 2012). However, radionuclides in the coastal waters are sources of external exposure that contribute to the total radiation exposure to humans.

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The aim of this work is to analyze the average activity concentrations of ^{137}Cs and ^{90}Sr in the waters of the Baltic Sea near Lithuania and to assess the external exposure dose to humans due to these radionuclides.

Materials and Methods

The samples of the sea surface water for determining activity concentrations of ^{137}Cs and ^{90}Sr were taken in the Baltic Sea at the distance of 10 m from the seashore of Lithuania at 0.5 meter depth. Water samples were taken near Juodkrante (*Fig. 1*). The minimum volume of the analyzed water sample was 40–50 liters. The temperature, specific electric conductivity, water current direction, wind speed and direction were registered during the time of sampling.

Figure 1. *Sample locations of the seawater*

During the whole research period, the determination of activity concentrations of ^{137}Cs and ^{90}Sr in the coastal surface waters of the Baltic Sea were carried out using a single technique – radiochemical analysis and the same type of measuring equipment (Styro et al., 2011; Styro et al., 2012). This research technique consists of some main stages: the concentration of ^{137}Cs and ^{90}Sr together with a stable carriers, radiochemical cleaning and activity measuring.

The ^{137}Cs and ^{90}Sr were concentrated from the same seawater samples. After the radiochemical cleaning the yield of cesium was determined gravimetrically in the form of $Cs_3Sb_2I_9$. It varied within a range of 60–80 %. The activity of ^{137}Cs samples was registered by gamma spectrometer (CANBERRA) with HPGe detector (resolution 2 keV, efficiency 15 %). The determination error for ^{137}Cs amounted to 10 %.

^{90}Sr was measured by ^{90}Y emission using a low level background beta radiometer. A stable strontium yield was determined by the atomic absorption spectrometer and Y – gravimetrically in Y_2O_3 form. The yield values varied within a range of 60–80 %. The determination error for ^{90}Sr activity concentration amounted to 15 %.

The external exposure dose to humans from ^{137}Cs and ^{90}Sr was calculated with the following equation (Eckerman and Ryman, 1993; IAEA, 2001):

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$$E_{im} = f\, DF_{im}\, C_V, \qquad \text{(Eq. 1)}$$

where E_{im} – external exposure dose to humans from radionuclide ($Sv \cdot h^{-1}$); f – exposure time to external radiation ($h \cdot y^{-1}$); DF_{im} – dose conversion coefficient for water immersion ($Sv \cdot m^3 \cdot Bq^{-1} \cdot y^{-1}$), C_V – activity concentration of the radionuclide in the seawater ($Bq \cdot m^{-3}$).

Results and Discussion

The average values of ^{137}Cs and ^{90}Sr activity concentrations in the coastal waters of the Baltic Sea near Lithuania (Juodkrante) in 1985–2013 are illustrated in *Fig. 2*. An average activity concentration of ^{137}Cs was 18 $Bq \cdot m^{-3}$ and ^{90}Sr – 28 $Bq \cdot m^{-3}$ before the ChNPP accident.

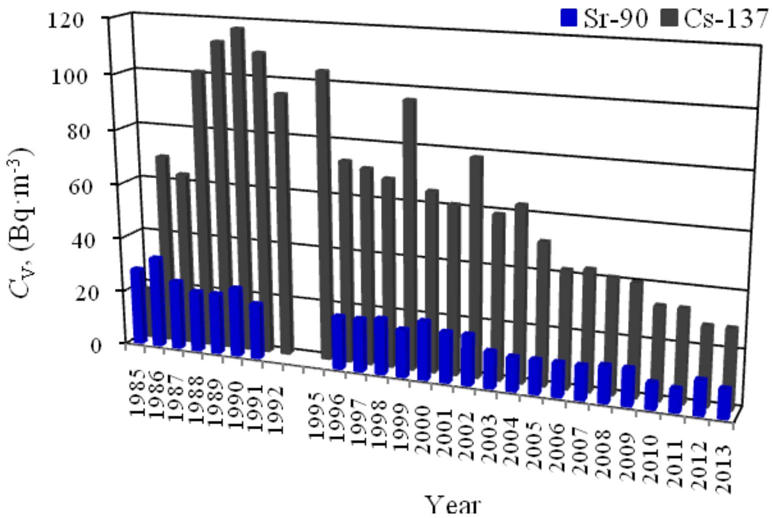

***Figure 2.** Average values of ^{137}Cs and ^{90}Sr activity concentrations in the coastal waters of the Baltic Sea near Lithuania (Juodkrante) obtained in 1985–2013 (data from 1985–2003 after Nuclear Hydrophysics laboratory of Vilnius Gediminas Technical University)*

After this accident the average activity concentration of ^{137}Cs considerably increased. The highest average values of ^{137}Cs in the coastal waters varied in the range from 95 $Bq \cdot m^{-3}$ to 117 $Bq \cdot m^{-3}$ were found from 1988 to 1995. This increase was attributed to the transport of more polluted water from the northern Baltic as well as to considerable riverine discharges (Styra et al., 2008). Since 1995 the situation stabilized and the activity concentration of ^{137}Cs started to decrease as a consequence of many processes, mainly radioactive decay, bioaccumulation and sedimentation (Styro et al., 2012). However, an unexpected increases of the average values of ^{137}Cs were also observed in 1999 and 2002. During 2003–2013, an average activity concentration in the coastal waters varied in the range from 63 $Bq \cdot m^{-3}$ to 30 $Bq \cdot m^{-3}$. However, in 2013 this average value exceeded almost two times the value found in 1985.

After ChNPP accident activity concentration of ^{90}Sr slightly increased. In 1986, the average concentration was 33 $Bq \cdot m^{-3}$. However, in 1987 it reached the value found before the accident (*Fig. 2*). During 1987–2002, an average activity concentration in the coastal

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waters varied in the range from 25 Bq·m^{-3} to 18 Bq·m^{-3}. In 2003, an average value of ^{90}Sr decreased to 13 Bq·m^{-3}. Since then, the average activity concentration of ^{90}Sr in the seawater varied within a relatively narrow range from 13 Bq·m^{-3} to 10 Bq·m^{-3}. The average activity concentration of ^{90}Sr in 2013 exceeded almost three times the value found in 1985.

According to data published in (Zalewska and Suplińska, 2013; Druteikienė et al., 2011; Lujanienė et al., 2010) and variations of average activity concentrations of ^{137}Cs and ^{90}Sr in the coastal waters near Lithuania, the Baltic is still the most contaminated seas in the world.

Certainly, variations of activity concentration of radionuclides in seawater have an impact on external exposure to humans. The summer season in the coastal Baltic Sea near Lithuania is from June to September. According to (Kahru et al., 2015), the number of days with the seawater warmer than 17 °C have almost doubled (from 29 to 56 days) over the past 30 years. Approximately, humans are being exposed when swimming in the seawater from few to few dozen hours per year.

External exposure dose to humans calculated using the average activity concentrations of ^{137}Cs and ^{90}Sr in the seawater and equation (*1*) are presented in *Figs. 3* and *4*.

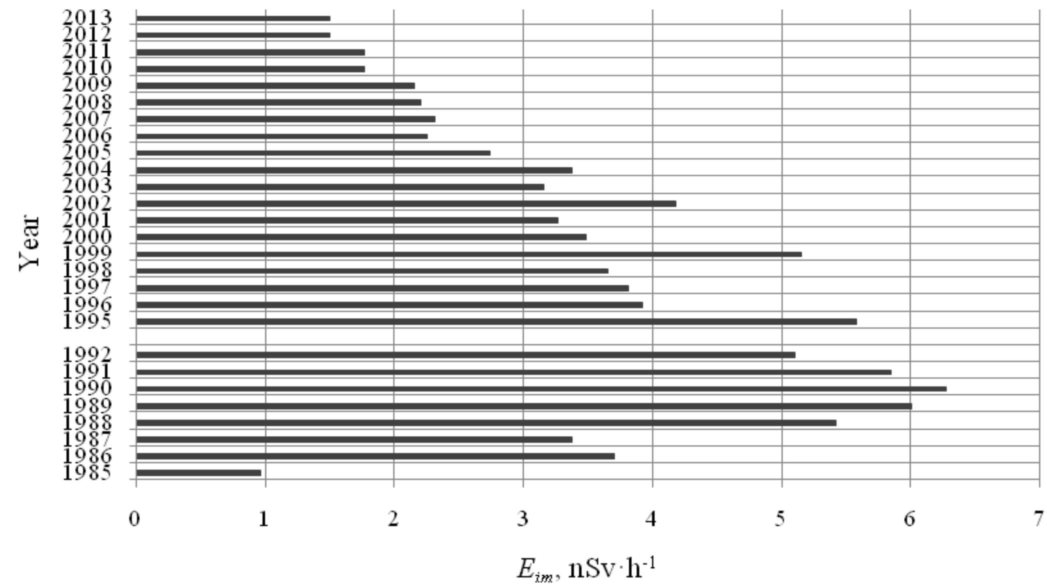

Figure 3. *External exposure dose to humans from ^{137}Cs in the coastal waters of the Baltic Sea (Juodkrante) in 1985–2013*

According to data obtained by the calculations, an average external exposure dose to humans from ^{137}Cs in the coastal waters in 1985–2013 ranged between 6.28 nSv·h^{-1} and 1.5 nSv·h^{-1} (*Fig. 3*), from ^{90}Sr – between 1.73 nSv·h^{-1} and 0.53 nSv·h^{-1} (*Fig. 4*). The highest exposure dose from ^{137}Cs was calculated in 1990, from ^{90}Sr – in 1986. After thirty years after ChNPP accident the exposure dose from ^{137}Cs decreased four times. The external exposure dose from ^{90}Sr due to immersion in the water decreased slower, almost three times.

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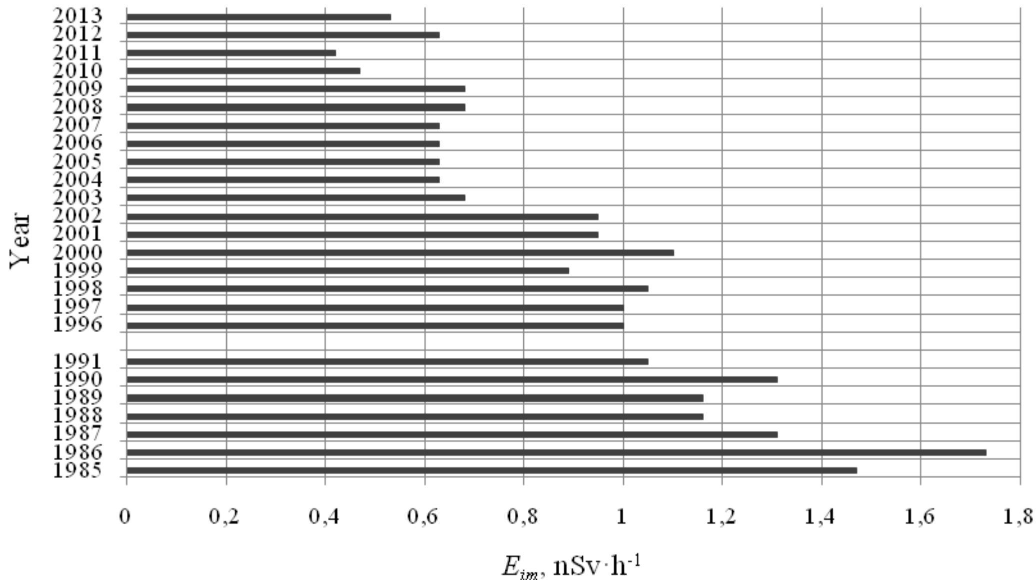

Figure 4. *External exposure dose to humans from ^{90}Sr in the coastal waters of the Baltic Sea (Juodkrante) in 1985–2013*

The external exposure from ^{137}Cs and ^{90}Sr in the seawater to humans when swimming is unavoidable. However, the average external exposure dose was very small in comparison to natural sources and did not make any risk for humans health.

Conclusions

Activity concentrations of ^{137}Cs and ^{90}Sr in the Baltic Sea waters decrease in time due to their natural radioactive decay and the decrease of the environmental radioactive pollution. According to average values, the activity concentration of ^{137}Cs in the coastal waters near Lithuania has been decreasing since 1990. The average activity concentration of ^{137}Cs decreased from 117 Bq·m^{-3} found in 1990 to 30 Bq·m^{-3} in 2013. However, the average activity concentration of ^{137}Cs in 2013 exceeded almost two times the value found in 1985. During the same period the average activity concentration of ^{90}Sr decreased almost three times, i.e. to 10 Bq·m^{-3}.

During the period from 1985 to 2013, an average external exposure dose to humans from ^{137}Cs in the coastal waters varied in the range from 6.28 nSv·h^{-1} to 1.5 nSv·h^{-1}, from ^{90}Sr – from 1.73 nSv·h^{-1} to 0.53 nSv·h^{-1}. The level of the external exposure was considerably lower than its limit (1 mSv·y^{-1}) and did not make any risk for humans health.

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