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COMPARISON AND ANALYSIS OF INVESTIGATION RESULTS ON VOLUMETRIC ACTIVITY OF ^{137}Cs AT THE BALTIC SEA COAST IN 2003–2005

[1]**Dmitrijus Styro**, [2]**Rasa Morkūnienė**, [3]**Nijolė Astrauskienė**, [4]**Asta Daunaravičienė**
[1,3,4]*Dept of Physics,* [2]*Laboratory of Nuclear Hydrophysics, Vilnius Gediminas Technical University, Saulėtekio al. 11, LT-10223 Vilnius, Lithuania. E-mail: styra@fm.vtu.lt*
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Abstract. Measurements of ^{137}Cs volumetric activity (VA) in coastal waters of the Baltic Sea (near Juodkrantė) were carried out in 2005. The highest values occurred in springtime; later they decreased, and in the summer–autumn period approximate homogeneous values of ^{137}Cs varied in the range of 41–57 Bq/m^3, and the lowest values were observed in autumn. Comparison of these values with analogous data obtained in 2003 and 2004 was carried out. The highest average annual value of 63 Bq/m^3 is proved to be in 2004. Furthermore, that year the greatest ^{137}Cs volumetric activity variations in the range of 49–91 Bq/m^3 were observed. These variations are related to possible appearance of additional sources of radioactive contamination in the sea. Evidently, such sources did not exist in 2005, when a satisfactory coincidence of measured and calculated results was obtained.

Keywords: the Baltic Sea, radionuclide ^{137}Cs, volumetric activity, extreme values.

1. Introduction

For the first time radionuclides of an artificial origin were supplied into the Baltic Sea with the global fallout as a result of the tests of nuclear and thermonuclear weapons in the atmosphere [1]. Later this sea was also contaminated by various nuclear objects and by the permeation of radionuclide ^{137}Cs from the North Sea [1–3].

After the accident at the Chernobyl Power Plant (ChPP) in 1986, an average ^{137}Cs volumetric activity (VA) in the surface waters of the Baltic Sea grew by more than an order of magnitude [4–10] relative to the radioactive background (12 Bq/m^3) formed after nuclear tests in the atmosphere [1, 2]. In 1986 a growth in the VA of artificial radionuclides was also observed [11], but the surface waters of the Baltic Sea were practically purified from these radionuclides within rather short periods [2]. The publications [8–23], etc present the distribution of the ^{137}Cs radionuclide VA after the accident at the ChPP.

The fallout of radionuclide ^{137}Cs onto the surface of the Baltic Sea was nonhomogeneous. A maximum fallout of this radionuclide was deposited in the northern part of the sea and the Gulf of Bothnia [2]. During a period of time, a levelling of the ^{137}Cs VA in the surface waters took place; the end of this process may be ascribed to 1989, when its VA appeared to be about 150 Bq/m^3 [12]. Until now an increased ^{137}Cs VA (45–50 Bq/m^3) in the southeastern part of the Baltic Sea (SEPBS) considerably exceeded the radioactive background formed by the global fallout [20]. By 1996 the ^{137}Cs VA in the SEPBS appeared to be vertically homogeneous [22].

The description of the self-purification of the Baltic Sea from ^{137}Cs radionuclide was made by a number of authors using mathematical modelling [12, 13]. The authors of studies [13] considered the physical processes including the advection and mixing of the waters between the neighboring "boxes", sedimentation of the suspended matter over the entire depth, mixing and displacement of the sediments, and bioconcentrating. Using this model, one may forecast an average annual VA of a radionuclide in the seawater, sediments and biota.

The other model [12] is simpler in terms of calculations. This model includes the vertical turbulent diffusion in seawater, the natural radioactive decay and the global fallout occurring until now.

The first [13] and the second [12] models describe the variation of ^{137}Cs radionuclide VA in the surface waters of the Baltic Sea from 1950 to 2000 and from 1996 to 2022, respectively. One must note, that the VA values of this radionuclide obtained with the above models for 2000 are in a satisfactory correspondence to each other (50–60 Bq/m^3). These data, in some cases, are in agreement with the results of in situ measurements in the SEPBS and coastal waters.

However, in a number of cases pronounced divergences were observed between the theoretical and experimental results, when the measured VA of ^{137}Cs radionuclide exceeded its calculated values. In particular, the measurement results of ^{137}Cs VA in coastal waters in 2003 often exceeded the calculated data by 20–50 %.

The detailed measurements of ^{137}Cs VA in the coastal waters of the SEPBS were carried out in 2004. The obtained results showed that the measurement values exceeded calculated data two times or more [20]. However, a satisfactory coincidence of measured and calculated results in the summer and autumn of 2005 was observed.

The objective of the present investigation is analysis and generalization of experimental data and search of possible cause of the various course of ^{137}Cs VA in the coastal waters of the Baltic Sea in 2003–2005.

2. Measurement method

The method of radiochemical precipitation of radionuclide ^{137}Cs in seawater samples was used during this study as described in [17, 19]. The procedure of ferrocyanide-carbonate precipitation was carried out. Seawater samples were taken from 0,5 m depth in coastal waters to water tanks of 40–50 liters in volume [17, 19]. The yield of ^{137}Cs was determined gravimetrically in the form of $Cs_3Sb_2I_9$. The yield values of caesium varied within 60–80 %. The activity of ^{137}Cs was registered by a gamma spectrometer with a semiconductor detector. The determination error for ^{137}Cs VA amounted 10 %.

3. Measurement results and their analysis

The measurements of ^{137}Cs radionuclide VA in the coastal waters of the Baltic Sea near Juodkrantė (Fig 1) were carried out in 2003–2005. Some results were obtained in Nida too.

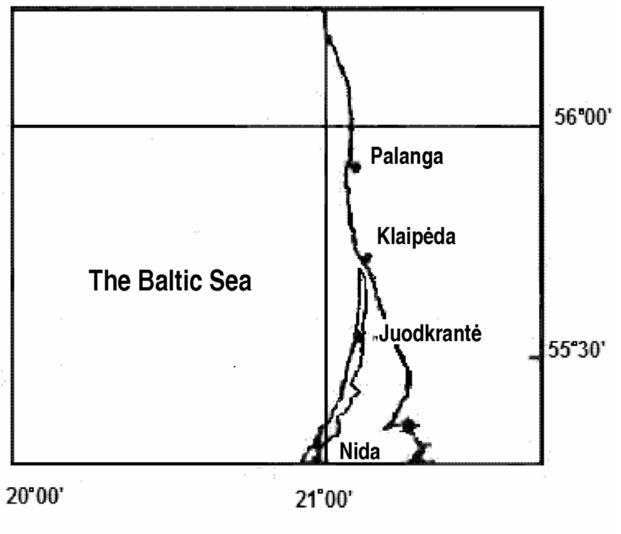

Fig 1. Lithuanian marine area of the Baltic Sea

As far as the global fallout of ^{137}Cs onto the surface of the Baltic Sea was nonhomogeneous [6], thus to the period of 1989–1990 the process of levelling of VA of the mentioned radionuclide was over. The ^{137}Cs VA field change in the surface waters of the Baltic Sea occurred under the effect of meteorological situations and the process of turbulent diffusion. The change in VA of this radionuclide in the coastal waters was observed too.

Since the beginning of ^{137}Cs VA field structure levelling in the surface waters of the Baltic Sea up to now, a gradual decrease in VA of this radionuclide was observed. However, temporal leaps of its VA were registered in 1999 and 2002 [23]. Similar leaps of ^{137}Cs VA are episodically observed in the coastal waters up to now.

The most reliable description of the course of VA of ^{137}Cs radionuclide in the coastal waters of the Baltic Sea refers to average annual values (Table 1).

Table 1. Average annual values of VA of radionuclide ^{137}Cs (Bq/m^3) in coastal waters of the Baltic Sea (near Juodkrantė)

Year	1996	1997	1998	1999	2000
VA	73	71	68	96	65
Year	2001	2002	2003	2004	2005
VA	61	78	58	63	54

However, the average annual data are obtained for a various number of measurements – from 4 to 32, that is why the temporal tendency of the course of ^{137}Cs VA should be considered as approximate with an exception for two last years, i e 2004 and 2005, when the number of measurements was 24 and 32, respectively.

During the period of observation in 1996–2005 (Table 1) the extreme values of individual measurements were observed (41 and 212 Bq/m^3) [17]. This fact confirms a possibility of temporary formation of anomalous high ^{137}Cs VA in the SEPBS. The stated above number differs by its magnitude from average annual values (Table 1).

The individual results of measurements in the coastal waters in 1996–2002 didn't exceed an annual number 11 [23], therefore, the greatest interest occurs in comparison of the data obtained from a greater number of measurements.

Such a period of observation appeared to be in 2003–2005. The extreme values of ^{137}Cs VA registered in this period are presented in Table 2.

Table 2. Extreme values of radionuclide ^{137}Cs VA (Bq/m^3) in 2003–2005 in coastal waters of the Baltic Sea (near Juodkrantė)

2003		2004		2005	
min	max	min	max	min	max
48	76	49	91	41	77

The greatest difference between maximum and minimum values of radionuclide ^{137}Cs VA (42 Bq/m^3) falls to 2004. Since July to October of 2004 the cyclic variations of VA of this radionuclide were observed, i e the series of its increase and decrease [18]. However, in 2005 the absolute magnitude and time trend of ^{137}Cs VA in coastal waters appeared to be different in comparison with those in 2004.

At the end of April of 2005 the average value of ^{137}Cs VA was 70 Bq/m^3, however, after the 1st of May this value considerably decreased, and during summer the values of individual observations were in the range of

45–57 Bq/m^3. In autumn these values were decreasing further and varied in the limits of 41–53 Bq/m^3 with an average value of 45 Bq/m^3 (Table 3).

The obtained results of ^{137}Cs VA (Table 3) differ essentially from the results of individual measurements in previous years [23] in absolute values and time trend. The comparison of average monthly data obtained in 2003–2005 is illustrated in Fig 2.

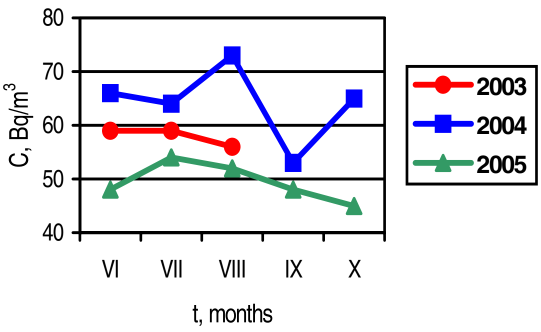

Fig 2. Average monthly values of radionuclide ^{137}Cs VA in the coastal waters of the Baltic Sea

The highest values are registered in 2004, when the greatest instability of absolute values of ^{137}Cs VA was observed [20]. The curves presented in Fig 2 define a possible tendency of changes in ^{137}Cs VA in coastal waters of the Baltic Sea.

4. Discussion of experimental data

A wide information on changes in ^{137}Cs VA in the coastal waters of the Baltic Sea at the observation station of Juodkrantė was obtained in 2003–2005. But the number of measurements per month is different, and it hinders to make a reliable comparison of the absolute values of ^{137}Cs VA and the tendency of its change in time as well. Thus the averaged values are to be assumed as approximate ones.

However, the results of individual observations appeared to be different. In certain cases they did not outgo beyond the limits of experiment errors but fairly often ^{137}Cs VA changed significantly, sometimes its absolute value differed two times, i e the "spots" of ^{137}Cs VA formed, which dissipated within a few days. This phenomenon was the most characteristic for 2004 year.

Table 3. Volumetric activity of radionuclide ^{137}Cs (Bq/m^3) and some hydrometeorological parameters of coastal waters of the Baltic Sea (Juodkrantė) in 2005

No	Date	^{137}Cs VA, Bq/m^3	Temperature of water, °C	Conductivity, mS/m	Average speed of wind, m/s	Averaged wind direction
1	2	3	4	5	6	7
1.	04,22	62±6	4,5	970	3	northwest
2.	04,23	69±7	5,0	965	3	north
3.	04,25	74±7	6,0	990	3	east
4.	04,27	66±7	7,0	990	4	east
5.	04,29	75±8	7,0	1050	3	north
6.	05,01	75±8	6,0	1050	3	east
7.	05,03	48±5	7,0	1025	5	southeast
8.	05,05	50±5	8,0	1050	4	north
9.	05,07	56±6	8,0	1025	4	west
10.	05,08	46±5	8,0	1025	3	southwest
11.	06,28	45±5	17,0	–	6	northwest
12.	06,29	50±5	17,5	995	6	north
13.	07,01	54±5	15,0	900	2	northwest
14.	07,03	55±6	16,0	900	2	northwest
15.	07,05	54±5	16,0	900	3	east
16.	07,07	54±5	17,0	960	2	east
17.	07,09	52±5	17,0	960	3	east
18.	08,23	49±5	18,0	990	3	east
19.	08,25	50±5	18,0	930	4	southeast
20.	08,27	54±5	17,0	960	6	west
21.	08,29	51±5	19,0	955	5	west
22.	08,31	57±6	19,0	965	2	northwest
23.	09,02	48±5	19,5	980	2	east
24.	09,04	47±5	18,5	–	3	north
25.	10,14	46±5	14,0	880	3	west
26.	10,15	41±4	13,0	920	6	north
27.	10,16	46±5	11,0	940	4	north
28.	10,18	53±5	9,0	960	2	northeast
29.	10,20	41±4	9,0	920	5	southwest
30.	10,22	45±5	10,5	960	6	south
31.	10,23	44±4	10,0	950	4	west
32.	10,25	48±5	9,0	960	6	southeast

The formation of enlarged ^{137}Cs VA observed in 2004 is impossible only due to natural hydrophysical processes. The lifting of ^{137}Cs from deep to surface waters is rejected too because ^{137}Cs VA in deep and surface waters differs insignificantly [21, 22]. The lifting of the radionuclide to surface waters from bottom sediments is not real as well, since the radionuclide practically is not washed out owing to its strong bonds with sediments [24]. Of course, a certain influence of freshwater from the Curonian Bay is possible at the observation station near Juodkrantė and at the whole coast of the Curonian Spit (Fig 1). However, it can be attributed to the local phenomena due to which ^{137}Cs VA can only decrease. It should be noted that such facts are easily verified according to change in salinity (conductivity) and temperature of sea water.

The impact of intensity of precipitations practically did not afflect the magnitude of ^{137}Cs VA if to take into consideration the fact that in August of 2005 the amount of precipitations in the sea coast area tripled the normal level. Therefore, the most probable is the transfer of the radionuclide from the regions of a greater activity to coastal waters. Unfortunately, the information about such regions is unavailable for the authors. However, it is found that radionuclide ^{137}Cs is not a fresh decay product, since in the analysed seawater samples radionuclide ^{134}Cs, which is easily found, practically was absent, i e its activity did not exceed the limits of measurement errors.

Average annual ^{137}Cs VA values appeared to be inconstant in the last three years (Table 1), and the extreme values appeared to be different as well (Table 2). Discrepancy between average annual experimental [18] and calculated [12, 13] values in 2004 and a satisfactory coincidence of the same values in the summer–autumn period of 2005 were observed.

The data obtained in 2004 may be interpreted as formation of local radioactive sources of contamination in surface waters as a result of the dumping of radioactive waste in the marine zone of Lithuania or its transfer from the other sea regions. The leaps of ^{137}Cs VA did not correlate with hydrometeorological parameters, therefore, these phenomena appeared to be independent of the variations of hydrometeorological conditions.

In the summer–autumn period of 2005 the local sources of radioactive contamination apparently were absent or they were diluted by intensive precipitations and increased flow of terrestrial waters. In this period a satisfactory coincidence of experimental and calculated data was observed; they were found to be within the limits of experiment errors.

5. Conclusions

1. Significant variations of ^{137}Cs VA (41–91 Bq/m^3) in coastal waters of the Baltic Sea (near Juodkrantė) in 2004 and comparatively homogeneous values of ^{137}Cs VA (41–57 Bq/m^3) in the summer–autumn period of 2005 were obtained.

2. Maximum average monthly values were obtained in 2004.

3. A satisfactory coincidence of measured and calculated data of ^{137}Cs VA was in the summer–autumn period of 2005, what indicates the absence of additional sources of contamination.

4. Correlation between ^{137}Cs radionuclide VA and hydrometeorological parameters or the intensity of precipitations was not observed; it confirms independence of formation of the leaps of ^{137}Cs radionuclide VA of hydrometeorological parameters.

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^{137}Cs RADIONUKLIDO TŪRINIO AKTYVUMO TYRIMŲ BALTIJOS JŪROS PRIEKRANTĖJE 2003–2005 m. REZULTATŲ PALYGINIMAS IR ANALIZĖ

D. Styra, R. Morkūnienė, N. Astrauskienė, A. Daunaravičienė

S a n t r a u k a

2005 m. atlikti išsamūs ^{137}Cs radionuklido tūrinio aktyvumo (TA) tyrimai Baltijos jūros priekrantės vandenyse (ties Juodkrante). Didžiausios reikšmės nustatytos pavasario laikotarpiu; vėliau jos gerokai sumažėjo, o dar vėliau, vasaros ir rudens laikotarpiu, kito nežymiai – nuo 41 iki 57 Bq/m^3. Mažiausios TA reikšmės gautos rudenį. Nustatytos TA reikšmės palygintos su 2003 m. ir 2004 m. atliktų tyrimų duomenimis. Didžiausia vidutinė metinė reikšmė – 63 Bq/m^3 ir didžiausi ^{137}Cs TA reikšmių pokyčiai – nuo 49 iki 91 Bq/m^3 buvo 2004 m. Šie svyravimai gali būti siejami su papildomų radioaktyviosios taršos šaltinių atsiradimu jūroje. Teorinių skaičiavimų ir eksperimentiniu būdu gautų rezultatų sutapimas 2005 m. buvo patenkinamas, nes taršos šaltinių tiriamuoju laikotarpiu nebuvo.

Reikšminiai žodžiai: Baltijos jūra, ^{137}Cs radionuklidas, tūrinis aktyvumas, ekstremaliosios reikšmės.

СРАВНЕНИЕ И АНАЛИЗ РЕЗУЛЬТАТОВ ИЗМЕРЕНИЙ ОБЪЕМНОЙ АКТИВНОСТИ РАДИОНУКЛИДА ^{137}Cs У БЕРЕГА БАЛТИЙСКОГО МОРЯ В 2003–2005 гг.

Д. Стыро, Р. Моркунене, Н. Астраускене, А. Даунаравичене

Р е з ю м е

Проведены подробные исследования объемной активности (ОА) радионуклида ^{137}Cs в прибрежных водах Балтийского моря (близ Юодкранте) в 2005 г. Наиболее высокие значения объемной активности радионуклида засвидетельствованы в весеннее время; далее они заметно уменьшились, после чего наблюдались сравнительно однородные величины ОА ^{137}Cs, которые летом–осенью колебались в пределах 41–57 Бк/м^3, а наименьшие значения пришлись на осень. Проведено сравнение этих величин с аналогичными данными, полученными в 2003 и 2004 гг. Наиболее высокой среднегодовой величиной оказалась 63 Бк/м^3 в 2004 г. Кроме того, в течение летне-осеннего периода этого года наблюдались наибольшие колебания ОА ^{137}Cs в пределах 49–91 Бк/м^3. Эти колебания связываются с возможным появлением в море дополнительных источников радиоактивного загрязнения. Однако такие источники, по-видимому, отсутствовали в 2005 г., когда наблюдалось удовлетворительное совпадение измеренных и расчетных результатов.

Ключевые слова: Балтийское море, радионуклид ^{137}Cs, объемная активность, экстремальные величины.

Dmitrijus STYRO. Dr Habil, Prof, Dept of Physics, Vilnius Gediminas Technical University (VGTU).

Doctor Habil of Natural Sciences (physics and mathematics), Sevastopol Marine Hydrophysical Institute (1984). Doctor of Natural Sciences (physics and mathematics), Vilnius University (1968).Publications: author of over 200 scientific publications. Conferences: participant of over 100 international and national conferences. Research interests: nuclear hydrophysics, radioecology and predictions of anomalous effects on the Earth.

Rasa MORKŪNIENĖ. Dr, senior research worker, Laboratory of Nuclear Hydrophysics, Vilnius Gediminas Technical University (VGTU).

Doctor of Technological Sciences (2000), Master of Physics (1995), VGTU. First degree of Chemistry, Vilnius University, 1992. Publications: author of over 30 scientific publications. Conferences: participant of over 20 international and national conferences. Research interests: radiochemistry, radioecology and environmental protection.

Nijolė ASTRAUSKIENĖ. Dr, Assoc Prof, Dept of Physics, Vilnius Gediminas Technical University (VGTU).

Doctor of Natural sciences (geophysics) (1972), Vilnius University. First degree in Physics (1963), Vilnius University. Publications: author of over 60 scientific publications. Conferences: participant of over 55 international and national conferences. Research interests: atmosphere physics, ecology.

Asta DAUNARAVIČIENĖ. Doctoral student, Dept of Physics, Vilnius Gediminas Technical University (VGTU).

Doctoral student (environmental engineering), VGTU, 2003. Master of Ecology, VGTU, 2003. Publications: co-author of 4 research papers. Conferences: participant of 4 conferences. Research interests: radioecology and environmental protection.