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ASSESSMENT OF THE INPUT OF PARTICULATE ^{239,240}Pu AND ^{137}Cs FROM THE NEMAN RIVER INTO THE CURONIAN LAGOON

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The Curonian Lagoon is a unique system that is a temporary reservoir of water, accumulating about 62% of the suspended solids in the river runoff. The average activity of plutonium in the suspended particulate matter in the water of the Neris River was 0.26±0.02 Bq/kg, and the activity of ^{137}Cs ranged from 23±8 to 49±12 Bq/kg. The average ^{137}Cs flux to the Curonian Lagoon in the particulate and dissolved species was estimated to be (78±61) × 10^9 Bq/year. Preliminary estimates indicate that the annual input of ^{239,240}Pu from the Neman River into the Curonian Lagoon is about 0.9 GBq, of which 0.56 GBq accumulates in the sediment of the lagoon.

Keywords: suspended sediment, Neman River, plutonium, caesium, Curonian Lagoon

1. Introduction

Plutonium is one of the most toxic radionuclides released into the environment due to its harmful effects on ecosystems and humans. The source of plutonium fallout in the Baltic Sea Region was mainly caused by testing of nuclear weapons (1945–1981) and the Chernobyl Accident (1986) [1]. The average ^{239,240}Pu deposition in the Baltic Region is estimated to be 40–50 Bq/m^2 [2].

Once plutonium reaches the soil surface, it binds strongly to fine particles and loses its mobility. The migration of plutonium depends mainly on the mineralogy of the soil and on redox reactions [3, 4]. The presence of organic material, iron minerals and clay particles slows down its migration in the soil considerably, and further movement of plutonium occurs mainly by particle transfer.

Leaching by rainfall flows and erosion processes initiate the transfer of ^{239,240}Pu with river waters from the entire catchment to the river mouth. The migration of soil particles ensures the transport of plutonium from watersheds to bottom sediments in coastal regions and estuaries [5]. For example, the contribution of plutonium from the global fallout to the Yellow River has been estimated to be 21±7×10^9 Bq/yr, with an average plutonium inventory in soil of 57±25 Bq/m^2 [6]. The Vistula and Neman rivers were found to contribute 0.19×10^9 Bq/year to the Gdansk Bay, and the total plutonium budget in sediments is 3.77×10^{12} Bq [7].

The Curonian Lagoon is located in the south-eastern part of the Baltic Sea and is a transitional zone where sediments originate mainly from the Neman River (Fig. 1). The Curonian Lagoon, a unique natural object, is currently under high anthropogenic pressure. Due to the shallow depth and low water exchange, the ecological situation of the Curonian Lagoon is very vulnerable to harmful factors [8]. The ecological situation of the lagoon is closely related to the water and sediment quality flowing into the rivers. The increasing technogenic pollution of the rivers leads to an increase in the input of pollutants into the Curonian Lagoon. The pollutants flowing in from the rivers can be plastics, toxic substances [9, 10], radionuclides [11] and other [12]. The high intensity of blue-green algae blooming (up to 100 g/m^3) and eutrophication

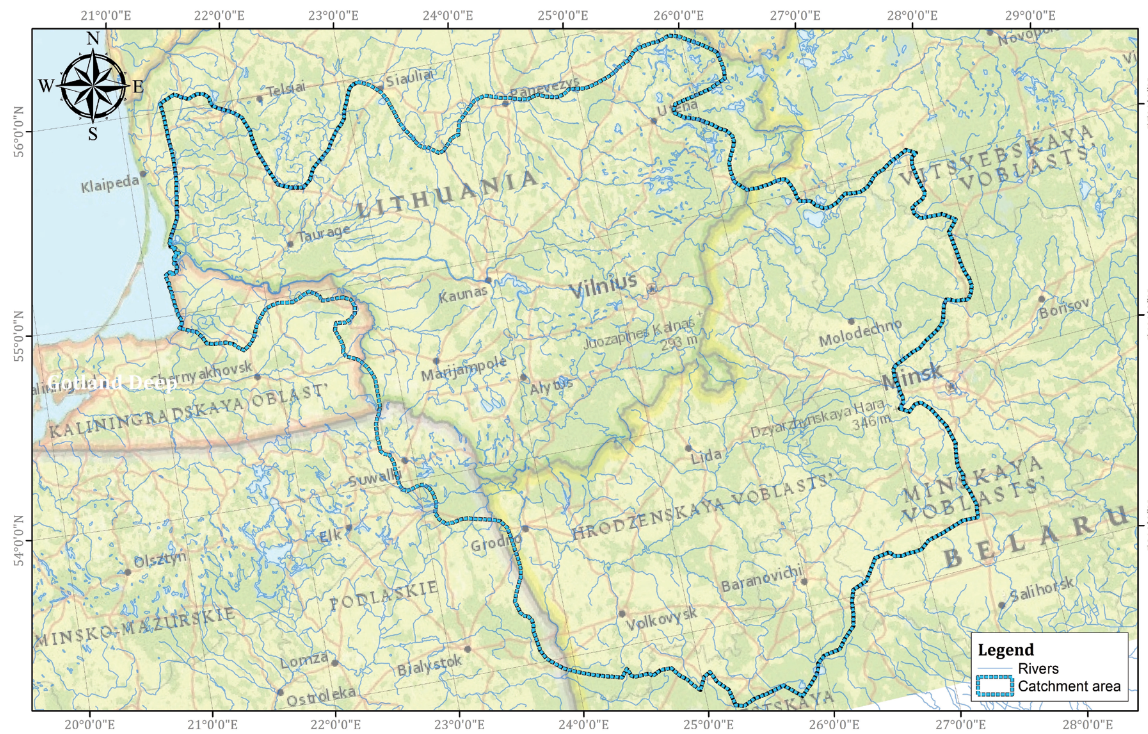

Fig. 1. Water catchment area of the Neman River (based on Ref. [15] data).

is one of the known problems of the Curonian Lagoon, which is accompanied by a decrease in oxygen content and fish mortality [13, 14].

The main input of suspended solids and associated pollution into the lagoon comes from the waters of the Neman River, which provides more than 90% of the total water inflow. The catchment area of the river is about 10^5 km^2 [16, 17]. The aim of this work is to make a preliminary assessment of the contribution of the river to the activities of ^{137}Cs and ^{239,240}Pu in the Curonian Lagoon based on measured and published data.

1.1. Caesium and plutonium sources in the study area

The main sources of plutonium in the watershed are the radioactive contamination from the global fallout from nuclear weapon testing and the Chernobyl Accident [18, 19, 20]. Published data indicate that the activity of ^{239,240}Pu reaches 15 Bq/m^2 in the lagoon sediments, 1.5 μBq/L in the waters and up to 1.3 Bq/kg in the suspended sediments [11, 21].

The transport of sediments in the waters of the lagoon is due to a unique water regime. In the southern and central parts, the frequency of water renewal exceeds 100 days [22], while in the northern part it does not exceed 80 days [23], which is due to the movement of water in the section between the delta of the Neman River and the section of the Strait of Klaipėda. The predominant direction of the water is the outflow into the Baltic Sea, but during the year a reverse movement of seawater into the lagoon can be observed, which can reach up to 30% of the annual outflow [24]. In the southern part of the lagoon there is a stagnant zone where water movement is mainly due to wind action [25]. According to various calculations, the amount of sediment accumulation in the lagoon is between 1.3×10^8 and 3.4×10^8 kg year^{-1} [26, 27].

2. Materials and methods

Particulate matter samples were taken at four stations in the Curonian Lagoon, Fig. 2(a), and at three stations in the Neris River, Fig. 2(b), in summer 2021. The samples in the Curonian Lagoon were taken in the coastal waters at the moorings. In the river, samples were taken at the deepest point of the water flow. The water was pumped through a set of three filter cartridges (*US Filter Plymouth Products*) with pore sizes of 0.2, 1 and 25 μm. The average volume of water pumped was 1 m^3. A conventional water meter was used to measure the volume of water pumped through the system.

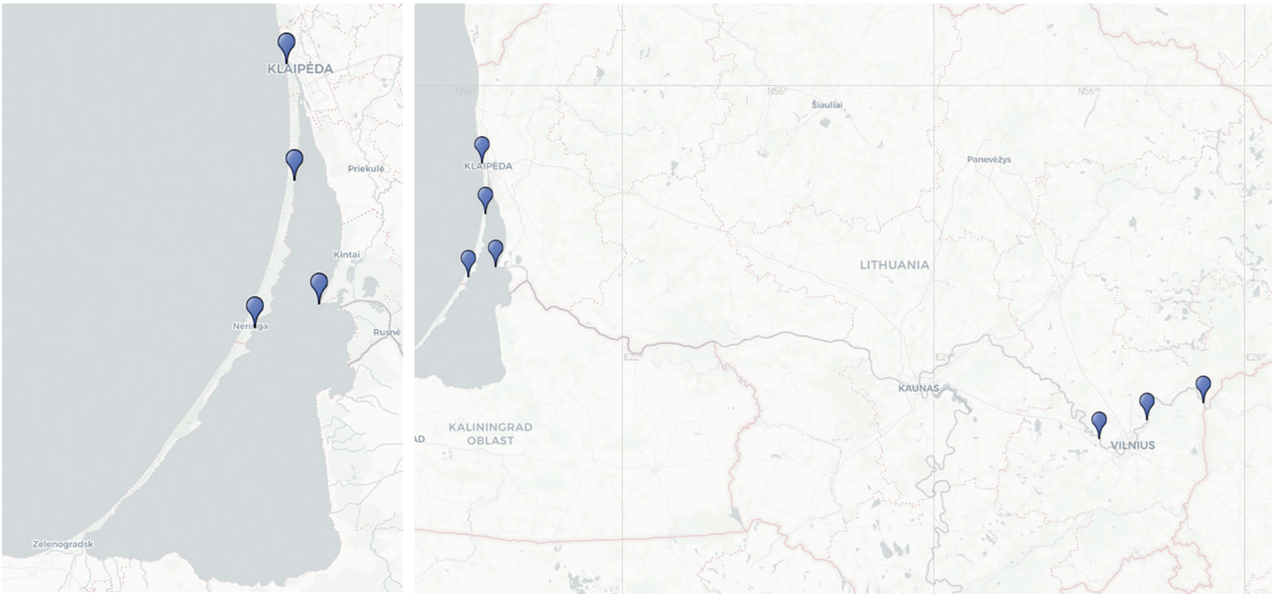

Fig. 2. Sampling design: in the Curonian Lagoon (a) and in the Neris River (b).

The concentration of suspended solids was determined by membrane filtration. A membrane of 0.2 μm was dried at 60°C for 24 h and weighed to the nearest 0.01 g. About 20 L of water was filtered through the membrane. The membrane was then dried again at 60°C and weighed. The concentration of suspended particles C was calculated using a simple equation

$$C = (m_1 - m_2) / V_{\text{water}}, \tag{1}$$

where m_1 and m_2 are the mass of the membrane before and after sampling, respectively, and V_{water} is the water volume pumped through the membrane.

The filters with suspended solids were dried at 25°C for one week. Then filters were ashed at 450°C overnight.

The surface sediments were sampled in the Neris River (Point N2, second from the right in Fig. 2(b)). The sample was taken about 1 m from the bank. The precipitate was dried at 60°C until the weight stabilised and then sieved through a 63 μm sieve before gamma-ray spectroscopy.

Measurements of gamma-ray emitting radionuclides were performed using an *ORTEC* gamma-ray spectrometer with an HPGe GWL-120-15-LB-AWT detector (resolution 2.25 keV at 1.33 MeV).

The sediments and suspended solid samples for Pu isotope measurements were ashed at 550°C and then dissolved in strong acids. The TOPO/cyclohexane extraction and radiochemical purification with TEVA resins (100–150 μm) were used to separate the Pu isotopes. ^{242}Pu was used as yield tracers (*AEA Technology UK*, *Isotrak*, *QSA Amersham International*, PRP10020 and ATP10020) in the separation procedures. The overall recoveries of ^{242}Pu was about 80%. Pu was electroplated onto stainless steel disks after purification and measured using an alpha-spectrometry system with passivated implanted planar silicon (PIPS) detectors of the 450 mm^2 active area (*AMETEK*, Oak Ridge, Tenn., USA) [21].

3. Results and discussion

3.1. Caesium activities

The activity of ^{137}Cs in the suspended solids in the water of the Neris River ranged from 23±8 to 49±12 Bq/kg with an average value of 32 Bq/kg (0.26 Bq/m^3). The level of ^{137}Cs activity in the fraction (<63 μm) was six times lower than in the suspended matter samples and was 5±3 Bq/kg.

The data obtained can be compared with previously published studies for this flow. Lujanas et al. (2002) [28] reported the ^{137}Cs activities in suspended sediments observed seasonally during 3 years (12 measurements) at the Buivydžiai Station, ranging from 20 to 180 Bq/kg (Fig. 3). The range, corrected for ^{137}Cs decay, is between 12 and 108 Bq/kg. The measured values of caesium activities in the suspended sediments are thus within the expected range published in previous studies.

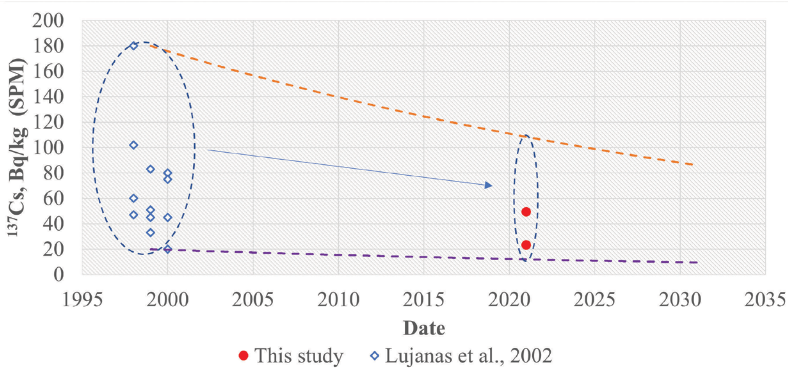

Fig. 3. Caesium activities in suspended solids.

The massic activity of ^{137}Cs in the suspended particles in the water of the Curonian Lagoon ranged from 2.2±0.7 to 12±2 Bq/kg with an average value of 9.4 Bq/kg. The average value is about three times lower than that found in the Neris River. The values observed in this study are close to those of previous studies [28], in which the average ^{137}Cs activities in the particulate matter for the stations near the mouths of the Neman and Neris rivers differed by about three times, with a maximum value of up to 12 Bq/kg. It can therefore be concluded that the values observed in this study correspond to the reality.

According to the previous observations [28], the average massic caesium activity in the suspended matter in the Neman River (Southern Lithuania) approximately corresponds to the activity in the Neris River (Western Lithuania). Taking into account the average particle concentration of 39.7 mg/L and the contribution to the Curonian Lagoon of $(4.8\pm3.8)\times10^8$ kg/year [29] with an average massic activity of caesium in the suspended matter of 32 Bq/kg, the average annual flux of caesium in the suspended matter to the Curonian Lagoon can be assumed to be $(16\pm12)\times10^9$ Bq/year. Assuming that about 80% of caesium occurs in a dissolved form in the Neman River [30], the total flux is estimated to be $(78\pm61)\times10^9$ Bq/year, which is close to the previous estimates [28] of $(41\pm34)\times10^9$ Bq/year (decay corrected to 2021).

The resulting caesium flux can come from the Chernobyl Accident as well as from the global fallout. The reason for this is the variability of the proportion of Chernobyl fallout in the suspended matter due to the different deposition in the watersheds and the uneven distribution of Chernobyl fallout. The proportion of Chernobyl caesium predominates in the southern part of the Neman River, while in the western part the relative proportion of caesium originating from Chernobyl is 2–4 times lower. However, the average activity of Chernobyl caesium in the bottom sediments and floodplains is 47% of the total river [31]. We can therefore assume that the resulting contribution of these sources from the catchment area into the Curonian Lagoon may be the same.

3.2. Plutonium activities

The massic activity of ^{239,240}Pu that was measured in the suspended sediments was 0.26 Bq/kg (2.1 mBq/m^3). This value can be compared with a theoretical estimate of the plutonium input to the river from soil erosion in the catchment. River water brings the sediment and associated plutonium from a catchment area of 10^5 km^2 [31].

Theoretically, the annual plutonium supply from the catchment can be estimated by Eq. [32]

$$Q = D \times I \times \ln(2) / t, \tag{2}$$

where D is the catchment area of feeding river, I is the reference tracer inventory on the catchment area, and t is the plutonium residence time in the catchment area.

3.2.1. Inventory

The inventory of ^{239,240}Pu fallout in the 50–60 latitude band [33] was estimated to be 1.3±0.5 mCi/km^2(48.1±18.5 Bq/m^2). UNSCEAR (1982) [34] reported 39 Bq/m^2 of plutonium deposition in the Northern Hemisphere. The fallout from

Chernobyl is unevenly distributed in this area and its contribution to the total plutonium in the soil can be as high as 96% [35]. Mietelski (2001) [36] reported that the percentage of plutonium detected in the soil layer in Poland is about 5% (centre), 15% (east) and in some cases 100% (north-east). The inventory of leachable ^{239,240}Pu of the global fallout ranged from 40 to 59.8 Bq/m^2 and the Chernobyl derived plutonium was up to 1.16±0.28 Bq/m^2. Lukšienė et al. (2015) [37] studied undisturbed Lithuanian meadow soils and found that the Chernobyl derived plutonium varied in the range between 6.5 and 59.1% of total plutonium. The average ^{239,240}Pu inventory in the southwestern transect was 26.0±4.0 Bq/m^2, while in the northeastern transect it was 19.0±3.0 Bq/m^2. In general, the available inventory values of ^{239,240}Pu do not exceed those reported in Ref. [34]. For a conservative estimate, the value of 39 Bq/m^2 was chosen because the entire catchment lies between 50- and 60-degrees north latitude.

3.2.2. Removal time by erosion

Since plutonium is firmly bound to soil particles, it is removed from the soil surface mainly by erosion processes. The removal of plutonium from the catchment can be influenced by land use and changes in soil erosion rates [39]. However, as the first approximation, the average annual rate can be considered constant [40]. The annual amount of eroded plutonium in a catchment depends on the residence time of plutonium (=ln(2)/[eroded fraction]).

There are several values for the residence time of plutonium in the catchment that have been used by the authors (Fig. 4).

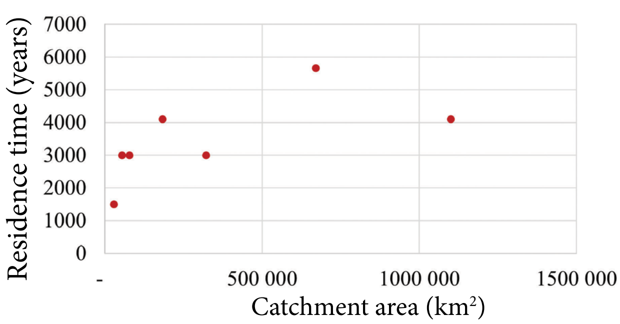

Fig. 4. Catchment area and plutonium removal time by soil erosion.

Smith et al. (1987) [38] used a time-dependent two-component model to simulate the transport of fallout radionuclides through the Saugenay catchment (78,000 km^2). According to the model conditions, the residence time of ^{239,240}Pu for 3,000 years agreed well with the other soil erosion model. On the basis of the removal rate of ^{239,240}Pu in the Columbia catchment (670,000 km^2), Beasley et al. (1984) [40] concluded that the removal of transuranic radionuclides by erosive processes is about 6,000 years. Zhuang et al. (2019) [41] assigned a residence time of 3,000 years to the Haihe River catchment (321,000 km^2). Wang et al. (2021) [42] studied the plutonium inventory in the Bohai and Yellow seas and assigned residence times of 800, 1,500 and 4,100 years to the areas of the catchments <5,200, 5,200–78,000 and >78,000 km^2. For the selection of the residence time, only approaches based on modelling or experimental data [38, 40] were considered. Thus, the residence time of 3,000 years was considered more appropriate for the 10^5 km^2 study catchment, as this is close to the catchment size of 0.78×10^5 km^2 used in the calculations of Smith et al. (1987) [38].

According to Eq. (2), the total input of ^{239,240}Pu from the river was estimated to be 905 MBq/year. Assuming that about 62% of the incoming sediment is trapped by the lagoon, 0.56 GBq/year of plutonium remains in the lagoon and 0.34 GBq/year enters the Baltic Sea.

Using the annual sediment input of (4.8±3.8)×10^8 kg/year, the estimated annual average concentration in the suspended sediments is to be 5±4 Bq/kg. This estimation is close to the data reported by Ref. [11] that ^{239,240}Pu massic activities were observed within a range of 0.13 to 1.3 Bq/kg at the Klaipėda Strait. The value observed in this study (0.26 Bq/kg) is close to the previously published values. Using the current data, we can assume that plutonium activities in the suspended sediments of the Neris River can vary by one order of magnitude. More measurements are needed to estimate the range of plutonium fluctuations in the suspended sediments, and additional data are necessary to specify a calculation model for the Neman River catchment.

4. Conclusions

The massic activities of ^{137}Cs in the suspended sediments in the water of the Neris River ranged from 23±8 to 49±12 Bq/kg with an average value of 32 Bq/kg (0.26 Bq/m^3). The comparison with the previously published data shows that the values are similar to those observed earlier and are decreasing due to the radioactive decay.

The average ^{137}Cs flux into the Curonian Lagoon was estimated to be (78±61)×10^9 Bq/year, taking into account particulate and dissolved fractions.

The theoretically estimated plutonium input from the Neman catchment (10^5 km^2) is 0.9 GBq/year. It was assumed that the average plutonium inventory in the catchment is 39 Bq/m^2. The residence time of plutonium in the catchment was set to 3,000 years for the calculations, which corresponds to the annual erosion rate of 0.023%.

The observed ^{239,240}Pu activity in the suspended sediments was similar to the previously published data. The average annual concentration of ^{239,240}Pu in the suspended sediments was 5±4 Bq/kg according to the theoretical estimates.

The calculations show that the amount of plutonium accumulating in the lagoon each year is about 0.56 GBq.

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^{239,240}Pu ir ^{137}Cs PATEKIMO SU KIETOSIOMIS DALELĖMIS NEMUNU Į KURŠIŲ MARIAS ĮVERTINIMAS

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Santrauka

Kuršių marios yra unikali sistema, veikianti kaip laikinas vandens rezervuaras, sukaupiantis apie 62 % kietųjų dalelių, įtekančių su upių vandenimis. Vidutinis suspenduotų kietųjų dalelių plutonio aktyvumas Neries upės vandenyje buvo 0,26±0,02 Bq/kg, o ^{137}Cs aktyvumas svyravo nuo 23±8 Bq/kg iki 49±12 Bq/kg. Vidutinis ^{137}Cs srautas į Kuršių marias, įskaitant kietąsias daleles ir ištirpusias formas, yra (78±61) × 10^{9} Bq per metus. Preliminariais vertinimais, per metus ^{239,240}Pu įtekėjimas į Kuršių marias su Nemuno upės vandenimis siekdavo apie 0,9 GBq, o Kuršių marių dugno nuosėdose susikaupdavo 0,56 GBq.