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Doses with α-particles of plutonium anthropogenic radioisotopes to the Black Sea hydrobionts

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Abstract. The data on assessment of the irradiation doses of the Chernobyl $^{239,\,240}$Pu to the Black Sea hydrobionts of different trophic levels are presented. The rates of adsorbed doses of internal irradiation from the alpha-radiation of incorporated $^{239,\,240}$Pu to the Black Sea hydrobionts varied from 0.01 to 0.354 μGy · years^{-1} Comparative analysis of dose levels formed with $^{239,\,240}$Pu to studied hydrobionts of the Black Sea in this work and doses formed with natural alpha-emitting radionuclide of ^{210}Po showed that $^{239,\,240}$Pu irradiation doses are 10^3–10^4 times lower, than irradiation doses with ^{210}Po. Hence, doses created with $^{239,\,240}$Pu to the Black Sea hydrobionts were in n · 10^6–10^7 times less than the dose level offered by International Atomic Energy Agency (IAEA), at which there are no negative consequences in populations of aquatic organisms.

1. INTRODUCTION

About 950 tons were produced and about 10 tons of anthropogenic plutonium (Pu) were nebulized all over the Earth, that thousand times exceeds natural Pu induced with cosmic rays in upper layers of the Earth's crust [1].

In radioecology and ecotoxicology Pu occupies a special place due to its chemical toxicity, as a heavy metal, and high radiotoxicity, as one of the group of radioactive nuclides, mainly alpha-emmiting ones (among 15 known Pu isotopes 12 of them are alpha-active isotopes). Especially ^{239}Pu stands out of other mentioned radioisotopes, because it has great practical value, and $^{239+240}$Pu together with ^{90}Sr and ^{137}Cs potentially highlyt contribute to radiation doses to men via sea products as a food [2, 3].

A special interest to plutonium contamination of marine environment appeared by necessity of evaluation and prediction of consequences of the Chernobyl nuclear power plant (ChNPP) accident in 1986 because $^{239,\,240}$Pu of the Chernobyl origin are potentially an additional source of anthropogenic radioactivity in the marine environment.

The objective of the investigation is to assess modern irradiation doses of the $^{239,\,240}$Pu to the Black Sea hydrobionts of different trophic levels.

2. MATERIALS AND METHODS

2.1 Research objects

The Black Sea sediments and water samples were collected between 1998 and 2005 from several location in open area of the sea during the research expeditions of the research vessel "Professor Vodyanitskiy" and in coastal areas during expeditions of research boat "Antares".

The biota consisted of multicellular algae (*Cystoseira crinita (Desf.), Ulva rigida Ag.*), mussels (*Mytilus galloprovincialis* Lam.) and fishes (*Sprattus sprattus phalericus* Risso, *Merlangus merlangus euxinus* Nordman and *Trachurus mediterranium ponticus* Aleev) were sampled in 1998–2004 in the coastal water of the Black Sea area near to Sevastopol.

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2.2 Plutonium determination method

Plutonium was determined with well-known radio-chemical methods [4]. Procedure was based on thermic and chemical pretreatments of the environmental and biological samples with further adsorption and desorption of Pu on ion-exchange resin (Dowex 1-X2 or AG 1-X2 chloride form 50–100 mesh for Pu refining). After refining and separation, Pu isotopes were co-precipitated with fluoride of lanthanum (LaF_3) [5] and obtained samples were measured with alpha-spectrometer "EG & G ORTEC OCTETE PC" provided by IAEA to the Dept of Radiation and Chemical Biology, IBSS. ^{242}Pu was added to samples as a radio-tracer standard for determination of chemical yield. Total error of the Pu concentration determination was not more than 13% for hydrobionts samples and 20% for water samples. Measured Pu concentrations in hydrobionts were based on their wet weight (ww).

2.3 Doses calculation method

Absorbed dose rates formed with alpha-emitting radioisotopes were calculated by using their concentrations in hydrobionts and alpha-particles energy of the proper Pu radioisotopes in accordance to widely accepted procedures [6, 7]:

$$D = 5.04 \cdot 10^{-6} \cdot C_{hb} \cdot E, \quad (1)$$

where:
D – absorbed dose, formed during a eyar, Gy;
C_{hb} – radioisotope concentration in hydrobiont, Bq/kg ww;
E – energy of alpha-particles of radioisotope irradiation, M∋V.

Equivalent dose rates were calculated by multiplication of the absorbed dose rates and radiation weighted factor equaled to 20 for alpha-particles:

$$D_{\ni KB} = Q \cdot D_{\pi}, \quad (2)$$

where:
D_{EQU} – annual equivalent dose, formed during a eyar, Sv;
D – absorbed dose, formed during a eyar, Gy;
Q – radiation weighted factor of alpha-particle irradiation.

3. RESULTS AND DISCUSSION

3.1 Pu content in abiogenic and biogenic component of the Black Sea ecosystem

After ChNPP accident a lot of radionuclides, including such radioecologicaly important transuranium element as Pu, entered to the Black Sea. The most radionuclide discharge took place in April–May 1986 by atmospheric way. Later the entry of Pu to the Black Sea took place in different ways and Pu migration has formed the summary (global fallout after atmospheric nuclear weapon tests and entry after Chernobyl NPP accident) Pu concentrations in the Black Sea water. More than 15 years after ChNPP accident in the north-western and western parts of shelf and in the open area of the Black Sea the levels of Pu average summary concentrations were 3.1–4.0 μBq/l of ^{239,240}Pu. At the Crimea coast ^{239,240}Pu concentration was 3–4 μBq/l, in the open area in the central and southern parts of the Black Sea – 4.1–6.4 μBq/l, in the north-easten part in nearshore areas – about 8.2 μBq/l [8–10]. Average ^{239,240}Pu concentration for the whole Black Sea was estimated for the surface water, and on 01.01.2000 it was equal to 5.3 μBq/l [11]. In addition the lowest ^{239,240}Pu concentration in the Black Sea surface water was about 2 μBq/l and the highest one – 20 μBq/l.

After Chernobyl-derived Pu entry to the Black Sea water the Pu migration to abiogenic and biogenic components of the sea ecosystem began. Bottom sediment takes very important place in these processes.

Bottom sediment in marine ecosystems as well as in freshwater ecosystems is main depository of Pu. In the Black Sea a role of bottom sediment as Pu depository is special and it intensified with anoxic conditions in deep part of the sea [8, 12]. The modern Pu average summary concentrations in bottom sediments were n · 10^2 mBq/kg ($^{239,\,240}$Pu) and n · 10^1 mBq/kg (^{238}Pu).

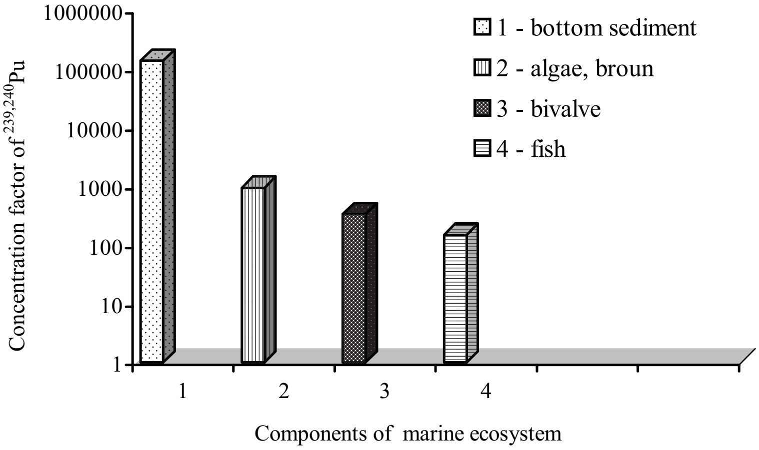

Figure 1. $^{239,\,240}$Pu concentration factors for abiogenic and biogenic components of the Black Sea ecosystem.

Marine hydrobionts are characterized by significant accumulative ability in respect to plutonium, they accumulated Pu radionuclides in concentrations, considerably higher than Pu concentration in the sea water. So marine biota could increase the Pu flow in food chains. The modern $^{239,\,240}$Pu average summary concentrations in the Black Sea studed biota were: in algae – 1–14 mBq/kg, in bivalve – 0.8–2,4 mBq/kg and in fish – 0.3–1.8 mBq/kg [10, 13]. The concentration factor (F_c) of Pu for the Black Sea hydrobionts becomes lower at the range: algae–bivalves–fish. The $^{239,\,240}$Pu F_c for abiogenic and biogenic components of the Black Sea ecosystem are presented on figure 1 [14].

3.2 Modern absorbed and equivalent doses of the Black Sea hydrobionts

Basing on data regarding to Pu contamination levels as well as to dose levels of irradiation to abiogenic and biogenic components of marine ecosystems with proper by long-lived radioisotopes of anthropogenic origin we have estimated of the modern radiation-ecological situation in the Black Sea during about 20 years after the Chernobyl NPP accident.

The rates of adsorbed doses of internal irradiation from the alpha-radiation of incorporated $^{239,\,240}$Pu to the Black Sea hydrobionts varied from 0.01 to 0.354 μGy · years^{-1} [14]. The range of changes of the adsorbed doses rates in specieses of the Black Sea hydrobionts inhabited in geographically different off-shore marine areas are represented in the table 1 (calculated by us on our and other authors published data) [9, 10, 14]. Concentrations of ^{238}Pu in studied the Black Sea marine organisms were below the limit of detection [10].

The results of estimation of maximum annual doses as equivalent doses to separate taxonomic groups of the Black Sea hydrobionts (multicellular algae, bivalves, fishes) in areas at the Crimean coast near Sevastopol and at areas at the Caucasian coast near Anapa, Gelendzhic and Sochy are presented in fig. 2. The obtained data testify that absolute values of the annual doses were differed in the studied districts of the Black Sea to the same groups of hydrobionts, but there was the general tendency of change of dose levels between different taxonomic groups. The obtained results showed that doses with $^{239,\,240}$Pu internal irradiation to different taxonomic groups of the Black Sea hydrobionts were diminished more, than one order of magnitude in a series: multicellular brown algae > bivalves > fishes.

Comparative analysis of dose levels formed with $^{239,\,240}Pu$ to studied hydrobionts of the Black Sea in this work and doses formed with natural alpha-emitting radionuclide of ^{210}Po [15] showed that $^{239,\,240}Pu$ irradiation doses are 10^3–10^4 times lower, than irradiation doses with ^{210}Po. Hence, doses created with $^{239,\,240}Pu$ to the Black Sea hydrobionts were in $n \cdot 10^6$–10^7 times less than the dose level offered by IAEA [16], at which there are no negative consequences in populations of aquatic organisms.

Table 1. Minimum and maximum of the absorbed dose rates, formed in the Black Sea hydrobionts with alpha-radiation of the incorporated $^{239,\,240}Pu$ radioisotopes.

Area of sampling	The Black Sea species of hydrobionts	Rate of absorbed dose, μGy/year	
		min	max
Area of the Black Sea at the Crimean coasts	*Cystoseira crinita* (Desf.) Bory	0.026	0.129
	Ulva rigida Ag.	0.031	0.071
	Mutylus galloprovincialis Lam.	0.020	0.049
	Trachurus mediterramium ponticus Aleev	0.016	0.016
	Merlangus merlangus euxinus Nordman	0.010	0.016
	Sprattus sprattus phalericus (Risso)	0.016	0.029
Area of the Black Sea at the Caucasian coasts	*Cystoseira barbata* (Good. et Wood)	0.168	0.354
	Mutylus galloprovincialis Lam.	0.039	0.062
	Trachurus mediterramium ponticus Aleev	0.010	0.047
	Sprattus sprattus phalericus (Risso)	0.010	0.013

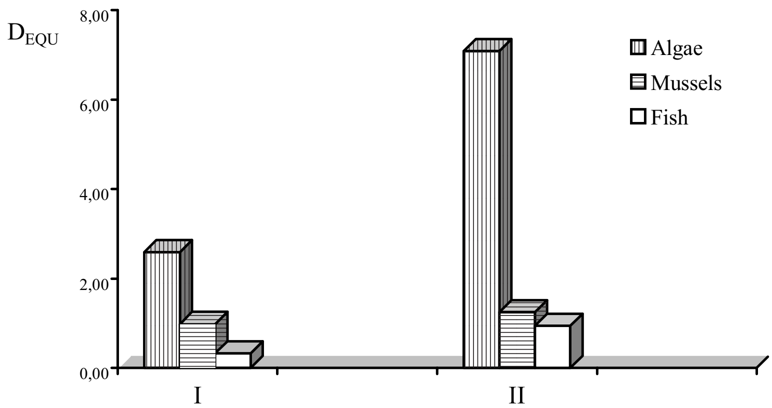

Figure 2. Maximum annual equivalent doses (D_{EQU}, μSv) created with $^{239,\,240}Pu$ to different taxonomic groups of the Black Sea hydrobionts in off-shore areas: I – at the Crimean coast and II – at shores of the Caucasus.

As the half-life of $^{239,\,240}Pu$ exceeds thousand years, their internal irradiation to hydrobionts is practically a permanent irradiation. In obedience to the radiation-ecological conceptual model of ecological influence zones of chronic ionizing irradiation [17], doses with alpha-radiation of $^{239,\,240}Pu$ to the Black Sea hydrobionts are characteristic for the "well-being zone" and for the "uncertainty zone". Therefore, remaining for many hundred years as registered radiation-ecological factor, nevertheless the modern concentration levels of the Chernobyl origin plutonium in the Black Sea do not render noticeable influence upon the biological functions and structures in components of the Black Sea ecosystems. Also the obtained regularities may be of immediate practical radioecological value in a case of unexpected (but not excluded in the nuclear epoch) an appearance of much greater amounts of Pu in marine ecosystems.

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