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Secondary radioactive contamination of the Black Sea after Chernobyl accident: recent levels, pathways and trends

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ABSTRACT

The recent radionuclide measurements have showed that concentrations of the Chernobyl-derived ^{137}Cs and ^{90}Sr in the surface Black Sea waters are still relatively high, reaching 56 and 32 Bq m^{-3}, respectively. This is comparable or even exceeds the pre-Chernobyl levels ($\sim$16 Bq ^{137}Cs and 22 Bq ^{90}Sr per m^3 as the basin-wide average values). The measurements have revealed that the Black Sea continues to receive Chernobyl radionuclides, particularly ^{90}Sr, by the runoff from the Dnieper River. An additional source of ^{90}Sr and ^{137}Cs was found in the area adjacent to the Kerch Strait that connects the Black Sea and the Sea of Azov. This may be caused by the inflow of the contaminated Dnieper waters, which come to this area through the North-Crimean Canal. The long-term monitoring of ^{137}Cs and ^{90}Sr concentration in the Black Sea surface waters and in the benthic brown seaweed *Cystoseira* sp., in comparison with the earlier published sediment records of the radionuclides, have showed signs of a secondary radioactive contamination, which has started to increase since the late 1990's. This may be the result of the combined effect of a higher input of radionuclides from the rivers in 1995–1999 due to an increased runoff; and a slow transport of the particulate bound radionuclides from the watersheds followed by their desorption in seawater from the riverine suspended matter and remobilization from the sediments adjacent to the river mouths.

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1. Introduction

The Black Sea has been one of the marine basins most contaminated with artificial radioactivity (Buesseler and Livingston, 1996, 1997; Egorov et al., 1999, 2008a, 2010; IAEA, 1999, 2005; Livingston et al., 1988; Polikarpov et al., 1991, 2008). During the pre-Chernobyl period, the main source of radioactive contamination of the Black Sea was the global fallout from the atmospheric nuclear weapon tests, which peaked in 1962 before the Test Ban Treaty was signed between the main nuclear states in 1963 (IAEA, 1999, 2005; Polikarpov et al., 1991). As maximum global fallout was observed within the 40–50° N latitude band, which runs exactly across the Black Sea, this semi-enclosed water body received high levels of the fallout radionuclides derived from the atmospheric weapons testing. The pre-Chernobyl inventory of man-made radionuclides was estimated in 1977 to be 510 $\pm$ 40 TBq ^{137}Cs and $\sim$700 TBq ^{90}Sr (e.g. Nikitin et al., 1988; Buesseler and Livingston, 1996, 1997; Polikarpov et al., 2008; Egorov et al., 2010). Being also a closest marine basin to the Chernobyl site, the Black Sea and its broad drainage areas have received in 1986 substantial amount of the long-lived artificial radionuclides, particularly ^{90}Sr, ^{137}Cs, and plutonium isotopes, released into the atmosphere from the damaged nuclear reactor and delivered with the air masses moving south- and westward from the accident area (Livingston et al., 1988; Nikitin et al., 1988; Polikarpov et al., 1991, 1992, 2008; Vakulovsky et al., 1994). As a result, concentration of man-made radionuclides in the Black Sea surface water increased sharply, exceeding the pre-Chernobyl levels by several times. In 1987, the total radionuclide inventory in the Black Sea water was of 2120 $\pm$ 420 TBq ^{137}Cs and 850 $\pm$ 190 TBq ^{90}Sr (Buesseler and Livingston, 1996, 1997; Egorov et al., 2010; Polikarpov et al., 2008; Vakulovsky et al., 1980, 1994). Besides the direct atmospheric deposition, the Black Sea received additional radioactive input by river runoff, particularly to its northwestern area from the Danube and Dnieper rivers (Polikarpov et al., 1992). Resulting from contribution of all these sources of radioactive contamination of the Black Sea, the ^{90}Sr concentration in its water ranks second after the Irish Sea, and third after the Irish and Baltic Seas with respect to ^{137}Cs (Egorov et al., 2010; Nielsen et al., 2010).

The extensive post-Chernobyl studies of radioactive pollution of the Black Sea has revealed that main environmental factors controlling the changes in concentration of the well-soluble

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radionuclides, such as ^{90}Sr and ^{137}Cs, in the upper Black Sea waters are the dilution with the less contaminated deep-sea waters, as well as outflow through the Bosporus Strait, while for the sorption-reactive plutonium isotopes the removal on sinking particles was a key process governed the decline of their concentration in seawater (Egorov et al., 2010; Gulin et al., 2012; Polikarpov et al., 2008). These factors led to a gradual decrease of radionuclide concentration in the Black Sea surface water with effective half-lives for ^{137}Cs of 5–7 years, 8–12 years for ^{90}Sr and 5–8 years for $^{239+240}$Pu (Gulin et al., 2012). The given half-lives are relatively short that is most likely caused by a faster dilution of the ^{137}Cs surface water concentration with riverine waters coming mainly from the Danube and Dnieper rivers, as well as by a loss of ^{137}Cs via the Bosporus Strait (Egorov et al., 2010; Gulin and Stokozov, 2005; Gulin et al., 2012).

At the same time, extrapolative assessments predict that input of the Chernobyl-derived radionuclides to the Black Sea from rivers, even of highly mobile ^{90}Sr, will reach the pre-accident levels by 2020–2025 only (Egorov et al., 1999, 2010). Also, the geochronological reconstruction of contamination history in sediment cores, which were taken in front of mouths of the main north-western and eastern Black Sea rivers Danube, Dnieper and Coruh, has shown significant delay of input to the Black Sea of particulate fraction of the Chernobyl radionuclides due to their retention in the watersheds (Gulin et al., 1997, 2002, 2003; Mirzoyeva et al., 2005). This implies that substantial amount of the Chernobyl-derived radionuclides may influence upon the Black Sea radioactivity in a longer-run.

The purpose of this study was to analyze the recent data on radioactive pollution of the Black Sea with a special emphasis on the changes in concentration of the Chernobyl radionuclides caused by their input from the drainage areas, as well as by contribution of other possible sources of the secondary radioactive contamination. This seems to be important for more accurate and reliable prediction of the long-term after-effects of the Chernobyl accident on this unique marine basin and its radioecological response.

2. Material and methods

The study was based on results of radioecological monitoring of the Black Sea, which was carried out during the entire post-Chernobyl period by the IBSS Department of Radiation and Chemical Biology in the framework of national and international projects (Egorov et al., 1999, 2010; Gulin and Stokozov, 2005; Gulin et al., 2012; Polikarpov et al., 1991, 1992, 2008). The most recent data on concentration of the man-made radionuclides were obtained during the 70th cruise of our R/V "Professor Vodyanitskiy" in August 2011, and in the land expedition in July 2012 in area of the North-Crimean Canal that brings the Dnieper water to the Crimea peninsula (Fig. 1).

The sampling was conducted primarily in the NW Black Sea area (Fig. 1), which has been subjected to radioactive pollution at the most extent due to proximity to the Chernobyl accident site and is influenced by the inflow from the Danube and Dnieper rivers accounting for 75% of the total river runoff to the Black Sea (Polikarpov et al., 1992). Also, the seawater samples were taken at two our permanent reference stations – one in the western part of the abyssal Black Sea, which is the area of a cyclonic mid-gyre (Fig. 1, Table 1 – station 14), and another located at the outer roadstead of Sevastopol (Fig. 1, Table 1). Finally, two samples were collected along the southern coast of the Crimea peninsula in August 2011 (Fig. 1, Table 1 – stations 3 and 11). The radionuclide concentration was also monitored in the Black Sea brown seaweed *Cystoseira* sp. collected in the near-shore zone of the outer roadstead of Sevastopol at the above-mentioned near-shore reference station.

The ^{137}Cs concentration in seawater was measured in the large-volume (>600 L) samples taken with an outboard silicone hose and an on-deck peristaltic pump (MasterFlex I/P 7591-55) coupled to an

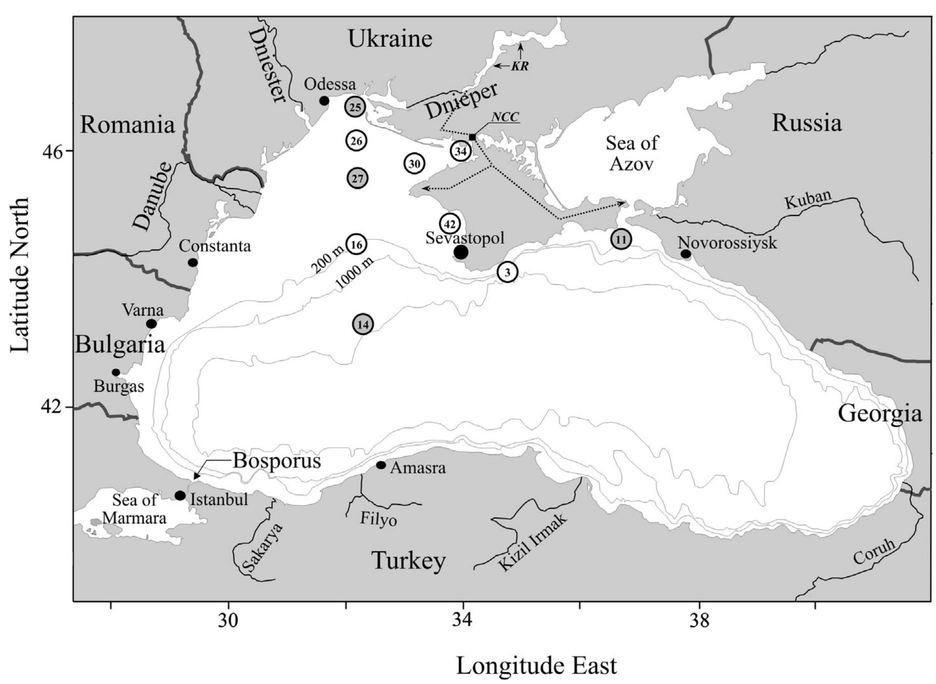

Fig. 1. Location of sample sites during the 70th cruise of RV "Professor Vodyanitskiy" in August 2011 and in the overland expedition in July 2012 in area of the North-Crimean Canal. The filled (gray) circles show the stations where both ^{137}Cs and ^{90}Sr concentration in surface water were measured. The open circles represent stations where only ^{90}Sr concentration was determined. The numbers inside the circles correspond to the ordinal numbers of stations of this cruise according to Table 1. Dotted arrows in the Crimea map indicates schematically the directions of the North-Crimean Canal (marked as NCC, showing also the place of our sampling therein). Location of the Kakhovskoye Reservoir is marked as KR.

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