

Sources and Distribution of Anthropogenic Radionuclides in Marmara Sea Environment

Marmara Denizi Çevresinde Antropojenik Radionüklilerin Menşei ve Dağılımları

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Abstract

The man-made or anthropogenic radionuclides entered to Marmara Sea marine environment from nuclear explosion tests, Nuclear Power Plants around Black Sea and Chernobyl accident. This paper provides comprehensive information on anthropogenic radionuclide levels in the Marmara Sea marine environment between 1970 and 2001 years. The results will be used as the national or international reference source for future studies in the regional scale.

Keywords: Marmara Sea, radionuclide, monitoring, biota, sediment

The anthropogenic radionuclides, entered marine environment from nuclear explosion tests. The estimated fission yields are 217 Mt since 1945. At the same time, the fission products of the atmospheric nuclear bomb tests are given to be 6 Mt from Chinese and French tests since 1973 (Holm, 1997). In addition, the estimated fission yields are 194 Mt (139 Mt as air bursts) among 1945 and 1963. The total of 194 megatons corresponds about 2.8×10^{28} fissioning atoms (National Academy of sciences, 1971). The fission products are released as radioactive fallout

and deposited worldwide. Some of these are ^{90}Sr , ^{95}Zr , ^{106}Ru , ^{131}I , ^{137}Cs , ^{144}Ce and ^{139}Pu (Eisenbud, 1973).

Environmental radioactivity from a nuclear power reactor can originate not only from low level emissions during normal operation but also from accident. It is well known that the amount of radioactive materials from nuclear power plants that actually reaches the environment is extremely small. During normal operation of the reactor new radionuclides are formed by fission of the nuclear fuel and neutron activation of corrosion products. Liquid wastes of the plants released to the environment include some radionuclides. The principal radionuclides present in coolant water of light-water reactors are ^3H , ^{51}Cr , ^{58}Co , ^{60}Co , ^{85}Kr , ^{90}Sr , ^{134}Cs , ^{137}Cs , ^{131}I , and ^{140}Ba , (Eisenbud, 1973).

The Chernobyl accident in early morning of April 26, 1986, had an impact on the marine and terrestrial environments. The released of long-lived radionuclides from the Chernobyl were observed as heavy local contamination and fallout pattern at many sites all over the world. The long-lived radionuclides are ^{134}Cs , ^{137}Cs , ^{90}Sr , ^{106}Ru , ^{144}Ce , $^{110\text{m}}\text{Ag}$, ^{125}Sb , $^{239+240}\text{Pu}$, ^{238}Pu , ^{241}Pu , ^{241}Am , ^{242}Pu and $^{243+244}\text{Cm}$ (Holm, 1997).

Biologically important of the radiouclides depended on concentration, chemical properties, physical half-life, solubility and oceanographic conditions, etc. It is well known that ^{137}Cs is one of the major dose contributing in the environment. At the same time, the ^{137}Cs radionuclide was the most important contaminant because of its high concentration in the Chernobyl fallout (Talmage and Mayers-Schöne, 1995). Moreover, the distribution of ^{90}Sr and ^{137}Cs radionuclides in the food chain organisms are similar to their stable chemical analogues. Both of the fission products ^{90}Sr and ^{137}Cs radionuclides will behave like calcium and will have properties like to potassium after entered marine environment.

Unfortunately we have not any data of ^{90}Sr , ^{137}Cs , plutonium radionuclides and other fission or activation products in biota and sediment samples of the Marmara Sea before Chernobyl

accident. On the other hand, the gamma spectrometric analyses were made in biota and sediment samples of the Küçükçekmece lake between the period of 1970 and 1973 (Ünlü et al., 1974). The results showed that the dominant contribution at the total radioactivities come from natural radionuclides in the 44 samples which have different origins.

Following the Chernobyl accident, the radioactivity concentrations are determined in fish samples from the Marmara Sea (Topcuoğlu et al., 1988). The following fish species caught from the Marmara Sea during the period of 1986-1987: common horse mackerel, chub mackerel, bluefish, seabream, sardine, silverside, picarel, smooth dogfish and bluefin tuna. The total activity levels of ^{132}I , ^{134}Cs , ^{137}Cs , and ^{103}Ru were found among 52 - 65 Bq/kg wet weights during May 1986. The maximum total radioactivity level of Marmara Sea fish samples gradually decreased during first three months after that, Chernobyl radionuclides were not detected, except ^{137}Cs radionuclide. In 1988 samples, the ^{137}Cs radionuclide were found to be 19, 10, and 8 Bq/kg dry weight in common horse mackerel, bluefish and smooth dogfish, respectively (Table 1) (Topcuoğlu et al., 1990). The ^{134}Cs were detected to be below the lower limit detection (< 1 Bq/kg) except bluefish sample. The ^{137}Cs concentrations in common horse mackerel and mullet fish samples were found to be below the lower limit detection in 2000 samples. On the other hand, this radionuclide was found to be 6.4 ± 3.5 Bq/kg dry weight in picarel fish in 2000 (unpublished data).

The similar sized mussels were collected from the same point of the Bosphorus from May 1986 to February 1987. The high level activities found for ^{134}Cs and ^{137}Cs as 141.7 and 289.2 Bq/kg dry weights in soft tissues during May and June 1986, respectively (Table 1) (Topcuoğlu and Van Downen, 1997). The activity levels are also given both on the counting date and date of Chernobyl in Table 2. These results are also used for determination of depuration rate of ^{137}Cs radionuclides under contaminated field conditions. The depuration kinetic represented by a single component and biological half-life found

to be 63 days. The ^{137}Cs , ^{90}Sr and $^{110\text{m}}\text{Ag}$ radionuclides were also investigated in sea snail and oyster samples collected from the Bosphorus during 1986, 1987 and 1988 (Bulut et al., 1993). The ^{137}Cs activity detected to be 5 and 3 Bq/kg dry weights in the sea snail samples during December 1986 and June 1987, respectively. However, we have not detected the ^{137}Cs activity in the sea snail samples after August 1987. On the other hand, the ^{137}Cs radionuclide is found to be 22 Bq/kg dry weigh in the Bosphorus sea snail samples in 1998 (Topcuoğlu et al., 2001). We think that the increase of ^{137}Cs activity level in sea snail may depend on the mouth of Danube river the definite point source of radioactivity for the Bosphorus with a more or less to give notice quantity of ^{137}Cs discharge. The ^{90}Sr activity is found to be below 1×10^4 Bq/g for all samples. The $^{110\text{m}}\text{Ag}$ activity was detected in oyster and sea snail samples to be 45 and 3 Bq/kg dry weights during December 1986 and June 1987, respectively. The ^{137}Cs radionuclide were also determined to be <3 , 6.1 ± 3.6 and <3 Bq/kg dry weight in Menekşe, M.Ereğlisi and Şarköy mussels in 2000 samples, respectively. At the same time, this radionuclide was found to be <3 Bq/kg dry weight in the Bosphorus mussel during 1998 (unpublished data).

In a study, ^{106}Ru , ^{134}Cs and ^{137}Cs radionuclides were determined in different macroalgae species among the periods of 1983-1985 and 1987-1989 at the different stations of the Marmara Sea (Güven, et.al., 1990). These radionuclides were not detected in all algae collected during the period of 1983-1985. The high activity levels for ^{106}Ru , ^{134}Cs and ^{137}Cs were found for *Ulva lactuca* in Gelibolu station during 1987 to be 10, 5 and 11 Bq/kg dry weights, respectively (Table 1) . The ^{106}Ru activity was also found to be 10 Bq/kg dry weigh in *Coralina granifera* species collected from the Gelibolu but ^{134}Cs and ^{137}Cs not detected in same sample. The results of the study also showed that the Chernobil contamination of the Çanakkale region was higher than other parts of the Marmara Sea. We think that the radioactive pollutants not only entered from the Black Sea by Bosphorus surface current circulation to the Çanakkale and Dardanelles regions. The radioactive cloud is arrived to the region from Greece and Italy (Topcuoğlu and Pala, 1992).

Literature also showed a similar result for the radioactive cloud progression over Europe.

The highest activity levels ^{134}Cs and ^{137}Cs activities in algae and sediment samples of the Bosphorus and Marmara Sea are given in Table 1. The highest accumulation of ^{137}Cs radionuclide found in *Enteromorpha linza* species. In general, the ^{134}Cs and ^{137}Cs values of sediment samples at the Bosphorus region are higher than Marmara Sea sediment. The highest levels of ^{134}Cs and ^{137}Cs detected at the Sariyer station of the Bosphorus (Topcuoğlu and Güngör, 1999). On the other hand, the ^{134}Cs and ^{137}Cs activities in Rumelikavağı, Anadolukavağı and Sarayburnu sediments of Bosphorus region are found to be significantly at low levels than the Sariyer samples. It is suggested that these differences depended on the sediment composition and underwater morphology of the Bosphorus. In addition, precipitation rate of the radionuclides was high in the Sariyer station, probably due to affinity of cesium radionuclides for particle suspended at Sariyer water column. The K_D (distribution coefficient) at Sariyer sediment was found to be significantly at high level than the other sediment samples collected from the Rumelikavağı and Sarayburnu stations (unpublished data). The ^{137}Cs activities in *Ulva lactuca* and *Cystoseira barbata* macroalgae species were found to be below the lower limit of detection (<2 Bq/kg) collected from Şarköy and M.Ereğli stations of the Marmara Sea in 2000. This radionuclide was also determined in sediment samples to be 17.1 ± 2.9 and 10.5 ± 1.6 Bq/kg dry weight in M.Ereğli and Şarköy in 2000.

In one study, the ^{137}Cs concentration was investigated in shallow-water sediments in Marmara Sea environment by Shimkus et al., (1993). The values reported within the range 130-1670 Bq/kg. These results are not in agreement with the results given above for sediment samples collected from Bosphorus and Marmara Sea. In previous studies, the ^{137}Cs concentrations reported within the ranges 8.5 – 142.7 and 11 – 94 Bq/kg in sediment samples collected from eastern and western Black Sea resgions, respectively (Varinlioğlu et al.,

Table 1. Radioactivity concentrations (Bq/kg in dry weights) in biota and sediment samples from the Bosphorus and Marmara sea

Sample	Location and date	^{134}Cs	^{137}Cs
MACROALGAE			
<i>Enteromorpha linza</i>	Bosphorus (06.1989)	<1	20±14
<i>Cystoseira barbata</i>	Bosphorus (06.1989)	<1	17±10
<i>Ulva lactuca</i>	Bosphorus (06.1989)	<1	2±1
<i>Ulva lactuca</i>	Marmara Sea (06.1987)	5±3	11±3
<i>Enteromorpha linza</i>	Marmara Sea (06.1989)	<1	6±1
<i>Cystoseira barbata</i>	Marmara Sea (06.1989)	<1	3±1
<i>Enteromorpha linza</i>	Marmara Sea (06.1990)	<1	4±2
<i>Codium fragile</i>	Marmara Sea (03.1990)	<1	<1
MUSSEL*			
<i>Mytilus galloprovincialis</i>	Bosphorus (05.1986)	114±40	229±84
<i>Mytilus galloprovincialis</i>	Bosphorus (02.1987)	6±4	13±5
FISH			
Bluefish, <i>Pomotomus saltator</i>	Marmara Sea (10.1988)	1.5±1	10±7
Hors mackerel, <i>Trachurus trachurus</i>	Marmara Sea (10.1988)	<1	19±11
Smooth dogfish, <i>Mustellus mustellus</i>	Marmara Sea (10.1988)	<1	8±7
SEDIMENT			
Sample-1	Bosphorus (06.1989)	9±2	80±3
Sample-2	Bosphorus (03.1990)	6±4	47±5
Sample-3	Marmara Sea (03.1990)	2±1	11±2

*The values of the activity were corrected for decay to the date of Chernobyl accident.

Table 2. The radioactivity levels in mussels after the Chernobyl accident

Collection date	Counting date	Activity levels on counting date		Activity levels on the date of Chernobyl (Bq/kg dry)		$^{134}\text{Cs}/^{137}\text{Cs}$	
		^{134}Cs (Bq/kg dry)	^{137}Cs (Bq/kg dry)	$^{134}\text{Cs}/^{137}\text{Cs}$	^{134}Cs ^{137}Cs		
17.05.1986	11.06.1990	27.8± 9.8	207.6±75.8	0.13	114.4±40.4	228.7±83.5	0.50
04.06.1986	09.06.1990	35.5±14.4	263.1±64.2	0.13	141.7±57.5	289.2±70.6	0.49
16.06.1986	11.06.1990	30.5±17.2	222.1±72.2	0.14	122.1±68.9	244.1±79.4	0.50
25.07.1986	12.07.1990	22.7± 9.5	173.0±59.6	0.13	93.3±39.1	190.5±65.6	0.49
11.08.1986	21.05.1990	21.9±11.8	159.8±26.3	0.14	85.9±46.4	175.4±28.9	0.49
21.08.1986	16.06.1990	16.1± 7.2	122.4±33.3	0.13	64.6±29.0	134.6±36.6	0.48
07.09.1986	28.05.1990	14.2± 4.2	104.2±28.7	0.14	56.1±16.6	114.5±31.5	0.49
28.09.1986	19.07.1990	8.6± 4.5	69.1±18.5	0.12	35.8±18.7	76.1±20.4	0.47
20.10.1986	01.06.1990	8.8± 3.9	67.9±14.6	0.13	35.1±15.5	74.6±16.0	0.47
05.12.1986	19.06.1990	8.3± 4.8	64.5±12.8	0.13	33.3±19.4	70.9±14.1	0.47
12.12.1986	14.06.1990	6.3± 3.9	49.9±14.7	0.13	25.3±15.7	54.9±16.2	0.46
18.12.1986	06.06.1990	3.8± 2.9	30.2±11.3	0.13	15.3±11.6	33.2±12.4	0.46
28.01.1987	19.07.1990	1.4± 0.8	12.0±5.3	0.12	5.9± 3.3	13.2± 5.8	0.45
28.02.1987	11.06.1990	1.5± 0.9	11.7±4.7	0.12	5.8± 3.6	12.9± 5.2	0.45

1995; Topcuoğlu et al., 2001). It is well known that the Marmara Sea environment was less polluted than the Black Sea environment by the Chernobyl impact.

The total β radioactivities of Marmara Sea waters in Yenikapı station measured as periodically by Çekmece Nuclear Research and Training Center. The total β activity of Yenikapı water detected fairly constant to be 5.5 ± 0.2 Bq/l (Health Physics Bulletin, 1992-1999). Güven et al., (1993) also investigated total β activity levels in different species of the Marmara macroalgae and found that to be ranged 154 – 1392 Bq/kg in dry weights. It is well known that the dominant contribution of total β activity in the environmental samples come from natural radionuclides.

In the Marmara Sea environment, no data on the transuranic radionuclides have been published in scientific literature.

Özet

Yapay ya da antropojenik radyonüklidler Marmara denizel ortamına, nükleer bomba denemelerinden, Karadeniz'in çevresinde bulunan nükleer güç santrallerinden ve Çernobil kazasından girmiştir. Bu makalede, 1970-2001 yılları arasında Marmara'nın denizel çevresinde saptanan yapay radyonüklid konsantrasyonları hakkında geniş bilgi verilmiştir. Bulgular, bölgede yapılacak olan yeni çalışmalara hem ulusal ve hem de uluslararası düzeyde referans kaynak olacaktır.

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