Journal of Environmental Radioactivity 101 (2010) 654–657

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Technical note

The concentration of ^{137}Cs in the surface of the Greek marine environment

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ARTICLE INFO

Article history:
Received 15 September 2009
Received in revised form
26 March 2010
Accepted 30 March 2010
Available online 11 May 2010

Keywords:
Caesium
Chernobyl
Surface seawaters
Food dosimetry
Greece

ABSTRACT

The radiological status of the Greek marine environment, prior to the Chernobyl accident, was characterized mainly by the fallout from nuclear weapon tests. However, the release of radioactivity into the environment from the accident in the Chernobyl Nuclear Power Plant and its deposition in the Greek marine environment resulted in an increase of the ^{137}Cs activity concentration by approximately one order of magnitude. In addition, the direct transport of radiocaesium into the North Aegean Sea has been further influenced by the late impact of the Chernobyl accident on the Greek marine environment, related to the transfer of ^{137}Cs, mainly through the Dnieper but also the Danube rivers, to the Black Sea and further to the North Aegean Sea through the Straits of Dardanelles. The aim of this work is to provide a present day picture of the geographic variation of the concentration of ^{137}Cs in the surface layer of the Greek marine environment and hence, to evaluate the annual committed effective dose delivered to humans through the ingestion pathway from marine sources.

1. Introduction

Over the last decades, anthropogenic radionuclides have been released to the marine environment as a result of fallout following nuclear weapon tests, the Chernobyl accident, discharges of radionuclides from nuclear installations, dumping of nuclear wastes into the world oceans and seas and via nuclear submarine accidents (Solomon, 1988; Paschoa and Steinhäusler, 2010). The variations in the source inputs and the subsequent dispersion and transport of these radionuclides in the marine environment, have resulted in their concentration to differ from one region to another.

Greece is a country surrounded by three main sea regions: the Aegean Sea in the east and south of mainland Greece; the Ionian Sea which lies to the west and, the Cretan Sea in the south. The three seas cover an area of about 270,000 km^2 (Zodiatis, 1993). The northeast region of the Aegean Sea is connected, through the straits of Dardanelles, with the Black Sea (Fig. 1).

The worldwide fallout from nuclear weapon tests and possible industrial/medical discharges to the sea had characterized the radiological regime in the Aegean Sea prior to the Chernobyl accident. The levels of ^{137}Cs measured in the Aegean Sea during the period 1984–1985 were approximately 2.6 $\pm$ 0.3 Bq m^{-3} (Florou and Kritidis, 1994). However, an increase in the radioactive contamination of the Aegean Sea, following the Chernobyl accident, would be expected due to an estimated average deposition of about 4 kBq m^{-2} and a total radiocaesium ($^{137}Cs + ^{134}Cs$) input of 530 TBq over the period 1986–1990 (Kritidis and Florou, 1990; Florou and Kritidis, 1994). Furthermore, the annual 4.1 $\times$ 10^{11} m^3 water flow from the Black Sea to the Aegean Sea results in an estimated annual flow of ^{137}Cs of about 48 TBq (Florou, 1992; Egorov et al., 1993). It should be noted that, since 1986, this caesium largely comes from Chernobyl via the Dnieper river.

The activity concentrations of ^{137}Cs measured in the surface layer of the Aegean, Ionian and Cretan Seas are presented in this paper for a reference date of the 2nd May 2009. This work presents concentrations of ^{137}Cs measured over the last two years in the surface layer of the Aegean, Ionian and Cretan Seas. Hence, the annual committed effective dose (CED) delivered to humans through the ingestion pathway and specifically the consumption of fish is evaluated. The study has focussed on ^{137}Cs only since ^{134}Cs, which accounted for about one third of the total caesium deposited in the Greek marine environment after the Chernobyl accident (Florou and Kritidis, 1994), has now totally decayed due to its half-life of 2.06 y.

2. Methodology

Surface samples, within the top 1 m, were collected from different locations in the three seas of interest (Fig. 1). The samples were the composite of sub-samples covering an area of 50 m $\times$ 50 m. The samples were analysed for ^{137}Cs using an ammonium molybdophosphate (AMP) radioanalytical pre-concentration method

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doi:10.1016/j.jenvrad.2010.03.016

Fig. 1. Location of the surface seawater samples collected for analysis.

(Folsom and Sreekumaran, 1970). The method is based on the ion-exchange of dissolved ^{137}Cs with ammonium molybdophosphate $[(NH_4)_3P(Mo_3O_{10})_4]$ which is an insoluble yellow reagent. Unfiltered seawater was analysed, since studies carried out indicated that almost all the injected Cs was in the soluble phase (Evangeliou et al., 2009). Specifically, 60–100 L of seawater (depending on the sensitivity of the gamma spectrometry system) are pumped to high volume vessels, previously rinsed with distilled water and 10% HNO_3. Acidification of the high volume sample is followed in situ using 65% HNO_3 to pH 2.0.

After transportation to the laboratory, additional acidification to pH 1.5 is carried out and 0.5 Bq ^{134}Cs per litre of sample is added as carrier and yield tracer (chemical yield estimate). Moreover, 400 mg microcrystalline ammonium molybdophosphate (AMP) per litre of sample is added for co-sinking via ion exchange with Cs (Folsom and Sreekumaran, 1970; Florou and Kritidis, 1994). The sample is continuously stirred for 15–30 min and then is allowed to precipitate for 48 h. The supernatant is pumped away and the AMP slurry is transferred to a 2 L beaker using 0.05 N HNO_3 rinsing and it is again allowed to precipitate. After several transfers to decreased volume beakers sequentially, the slurry is placed in a "calibrated" measurement pot (radius 3.4 cm and height 2.0 cm) for gamma spectrometry and it is dried at 60 °C for 1 h. The yellow solid containing ^{137}Cs is finally analysed by gamma spectrometry. The estimated mean chemical yield of the radioanalytical method varied between 90 and 96%.

The required gamma spectrometry was carried out using a Canberra system comprising of a High Purity germanium (HPGe) Detector System with an efficiency of 90% (relative to a 3″ × 3″ NaI (Tl) crystal) and resolution of 2.1 keV at the 1.33 MeV photopeak of ^{60}Co. The HPGe detector is connected to an 8k multi-channel analyzer operated with the Genie 2000 software. The energy calibration has been performed using standard active sources of ^{22}Na, ^{54}Mn, ^{57}Co, ^{60}Co, ^{109}Cd, ^{133}Ba, ^{137}Cs, ^{241}Am covering an energy range up to 2000 keV, necessary for a range of different activities in the laboratory. The efficiency of the detector was calculated using a standard active source of ^{226}Ra (240 Bq) under the same geometry with the pot used for the measurement of the samples. The samples were measured for 70,000 s, whereas the relative statistical error (1σ) was kept always to less than 10%. In the present paper all the measured activity concentrations of ^{137}Cs were corrected for decay to the 2nd May 2009.

3. Results

The results of the ^{137}Cs activity concentrations (Bq m^{-3}) in the Greek marine environment are given in Table 1. Average values and the range of values encountered are given for the concentrations and the associated committed effective doses are given. It should be noted that the present level of ^{137}Cs, if the accident had not happened, would have been 1.5 Bq m^{-3}, constituting the contribution of ^{137}Cs to the radioactivity of the background in the area.

Table 1
Concentration of ^{137}Cs in the surface of the Greek marine environment and the annual CED delivered to the humans through the fish intake.

Location		^{137}Cs concentration (Bq m^{-3})			Annual CED (μSv)		
Map	Name	Average	Range		Average	Range	
	Ionian Sea						
1	North	4.10	2.40	5.30	0.06	0.04	0.08
2	Central	8.70	4.80	12.70	0.14	0.07	0.20
3	South	4.30	2.30	7.80	0.07	0.04	0.12
4	Patraikos Gulf	4.50	2.40	11.80	0.07	0.04	0.18
5	Korinthiakos Gulf	3.60	2.70	4.10	0.06	0.04	0.06
6	Saronikos Gulf	8.70	5.30	14.60	0.14	0.08	0.23
7	Evoikos Gulf	5.40	2.30	8.00	0.08	0.04	0.12
8	Pagasitikos Gulf	5.50	4.30	7.10	0.09	0.07	0.11
	Aegean Sea (north)						
9	Thermaikos Gulf	5.70	2.70	14.70	0.09	0.04	0.23
10	Kavala[a]	39.20			0.61		
11	Thasos[a]	4.80			0.07		
12	Alexandroupolis	17.00	9.70	24.30	0.27	0.15	0.38
13	Limnos	9.60	4.80	24.20	0.15	0.07	0.38
14	Dardanelle[a]	82.70			1.29		
	Aegean Sea (central)						
15	Lesvos	6.80	3.90	12.50	0.11	0.06	0.20
16	Chios[a]	2.00			0.03		
17	Milos[a]	2.90			0.05		
18	Ikaria	10.20	3.90	16.50	0.16	0.06	0.26
	Aegean Sea (south)						
19	Rodos	5.70	2.10	14.70	0.09	0.03	0.23
20	Kos	3.60	3.30	3.90	0.06	0.05	0.06
21	Karpathos[a]	4.70			0.07		
	Kriti						
22	Xania[a]	4.40			0.07		
23	Souda	9.50	4.30	13.60	0.15	0.07	0.21
24	Heraklion[a]	4.40			0.07		
25	Ag. Nikolaos[a]	4.30			0.07		
26	South coast	1.90	1.10	2.30	0.03	0.02	0.04

[a] Based on one sample analysed.

The highest average concentrations are observed in the northern Aegean Sea and in particular at a location where seawater exiting from the mouth of the Dardanelles was sampled and analysed. A ^{137}Cs activity concentration of 82.7 Bq m^{-3} was measured there. This is in accordance with studies which have estimated an annual outflow of ^{137}Cs from the Black Sea to the Aegean Sea of about 48 TBq (Florou, 1992; Egorov et al., 1993; Florou and Kritidis, 1994). The north Aegean Sea is the area where the Aegean Sea interacts with the Black Sea with the consequence that its upper layer is greatly influenced by the Black Sea water flux and any ^{137}Cs activity that it carries. Hence, the high ^{137}Cs activity which is observed particularly close to the Dardanelle mouth.

However, elevated activities are observed along the northern part of the Aegean Sea, not only close to the Dardanelle straits but also further away, especially for Kavala, Alexandroupolis and Limnos (39.2, 17 and 9.6 Bq m^{-3}, respectively).

In the rest of the Greek marine environment considered in this study, the average ^{137}Cs activity, from all the samples measured, is 5.2 Bq m^{-3}, with the values ranging between 1.1 and 16.5 Bq m^{-3} (Table 1) Although these values are lower than in the northern Aegean, the majority of them are higher than what would be considered as background radioactivity if Chernobyl had not happened.

The radiocaesium distributed in the marine environment would eventually reach humans through the consumption of fish. It is assumed that the external exposure to the population from seawater is not significant. The ^{137}Cs activity concentrations in marine fish after the Chernobyl accident were one order of magnitude higher than those observed before April 1986 (Florou et al., 1987).

In order to convert the ^{137}Cs activity in the seawaters to a dose for the public, a Transfer Factor (TF) for ^{137}Cs to fish of 100 (kBq/kg)/(kBq/L) is considered (IAEA, 2004). An annual fish consumption rate of 12 kg is assumed (Kritidis et al., 1990). A Dose Conversion Factor (DCF) for adults of 1.3×10^{-8} Sv/Bq (ICRP, 1996) is used to convert the annual intake of ^{137}Cs activity, through the fish consumption, to the committed effective dose (CED) delivered to the humans.

The estimations of the CED are shown in Table 1 for the different regions of the Greek marine environment considered in the study. A range of CED values are encountered ($0.02–1.29 \times 10^{-3}$ mSv), depending on the ^{137}Cs activity concentration in the sea and hence the fish which would be consumed. Nevertheless, they represent a small fraction of the total annual allowable dose of 1 mSv due to all possible sources and pathways (ICRP, 1991). Therefore, it could be concluded that from the ^{137}Cs calculations presented here, the consumption of fish from the seawaters studied are of minor radiological importance to the public.

The Derived Intervention Level (DIL), in the case of fish contaminated with ^{137}Cs has been evaluated. The DIL represents the maximum amount of the radionuclide activity that man can consume without exceeding the reference level of annual dose at 1 mSv. The DIL (Bq/kg) is expressed by means of the following relationship (Randell, 1988):

$$\mathrm{DIL} = (\mathrm{RLD}/m \cdot d)$$

where RLD is the reference level dose (Sv/y), m is the mass of food consumed annually (kg/y) and d is the dose per unit intake (Sv/Bq). In the present study, these parameters take the following values: RLD = 1 mSv, $m = 12$ kg and $d = 1.3 \times 10^{-8}$ Sv/Bq (ICRP, 1996). This yields a value for DIL of 6410 Bq/kg which represents the maximum allowable intake of fish by man without exceeding the annual dose limit of 1 mSv. This DIL value is 770 times higher than the specific activity of ^{137}Cs in fish at the exit of Dardanelles. In the rest of the seawaters, with the DIL would be up to three orders of magnitude higher.

4. Conclusions

The Chernobyl accident resulted in elevated ^{137}Cs activities in the Greek marine environment compared to the background radioactivity prior to the accident. The ^{137}Cs activity was particularly high in the northern part of the Aegean due to its higher deposition immediately following the Chernobyl accident (Kritidis and Florou, 1990). Furthermore, some of the inventory of ^{137}Cs within the Black Sea, as a result of caesium discharges from Chernobyl via the River Dnieper, would eventually reach the northern Aegean, via the Dardanelles straits, to produce elevated values in this region. Average activity concentrations of ^{137}Cs of about 11 Bq m^{-3} and 5.2 Bq m^{-3} were observed in the northern Aegean and the rest of the Greek marine environment considered in this study, respectively. These values are higher than the 1.5 Bq m^{-3}, which would have been the background radioactivity at the time of the measurements if the accident had not happened.

The presence of ^{137}Cs in the Greek marine environment would result in a dose to the public, through the ingestion pathway and their intake of marine fish. Nevertheless, the resulting annual committed effective dose would be well below the annual recommended dose of 1 mSv. Hence, the presence of ^{137}Cs in fish caught and consumed in the northern Aegean Sea region would be of minor radiological importance to the public. The DIL, which represents the maximum allowable intake by man without exceeding the annual dose limit of 1 mSv, was found to CED be at least 770 times higher than could be attributed to the activity concentrations of ^{137}Cs in Greek marine waters.

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