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Temporal variation of ^{137}Cs profiles in Lemnos deep basin, North Aegean Sea, Greece

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^{137}Cs activity concentration in seawater is an efficient radio-tracing technique to separate and identify the origin of different water masses along with depth. This technique has been applied for the study of deep basins at the North Aegean Sea [1], a marine region which is continuously enriched with ^{137}Cs originated from the Black Sea, through the Dardanelles Straits. In this work, a second sampling campaign (from 2008 to 2013) was carried out in Lemnos's deep basin and water quantities were collected from different depths. These quantities were chemically treated at HCMR to pre-concentrate ^{137}Cs by using the AMP precipitation method [2]. The produced samples were measured by means of γ -ray spectroscopy. The ^{137}Cs data profile in combination with other oceanographic data (salinity and temperature), provides significant information for the water masses origin with respect to depth. More specifically, at the surface layer, Black Sea water masses (characterized by enriched concentrations of ^{137}Cs) are observed, with ^{137}Cs concentrations about 4 Bq/m³. At the intermediate layers, Levantine water masses (characterized by low concentrations of ^{137}Cs) appear with ^{137}Cs activity reduced to 2 Bq/m³, while the deep layer dense water masses exhibit intermediate ^{137}Cs concentrations of 3 Bq/m³. The ^{137}Cs concentration results are compared with previous measurements performed in the same basin from 1999 to 2013, where the decline of ^{137}Cs concentrations in the surface layer is observed and similar concentrations about 3 Bq/m³ (within uncertainties) in the deep layer are obtained.

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INTRODUCTION

One of the most used radionuclides for radio-tracing techniques, is the artificial radionuclide of ^{137}Cs with half-life of approximately 30 years. The major sources of ^{137}Cs to the Mediterranean Sea are the fallouts following nuclear weapon testing in the early 1960's and the Chernobyl accident in 1986. The Chernobyl fallout produced an inhomogeneous input, where the maximum value appeared in the eastern Mediterranean and in the Adriatic Sea [3].

The marine environment is a dynamic system. Radionuclides are mainly introduced to surface waters by river discharges and rainfalls and they do not remain in steady-state conditions due to currents and other physical processes in the water column. Therefore, radionuclides are considered a useful radio-tracing tool to investigate oceanographic

phenomena [4]. ^{137}Cs radio-tracing technique can provide information of the water masses exchange that occurs between North Aegean and Black Sea (rich in ^{137}Cs) and the oceanographic phenomena that take place in the basins of Aegean. Especially North Aegean, is an appropriate area for applying this technique, due to its connection with Black Sea through Dardanelles Straits [1],[5]. In this work ^{137}Cs activity concentrations of nine seawater samples were utilized along with other physical properties (potential temperature, salinity) to study the movement of seawater masses and to understand the vertical transport processes in the deep basin region North of Lemnos Island.

MATERIALS AND METHODS

Study area and Sampling

The topography of the Aegean Sea, as well as the connection to the Black Sea through the Dardanelles Strait System and Levantine basins, through the Straits of the Cretan Arc, determine the hydrological structure of the Lemnos basin [6] Lemnos Basin is situated in the North Aegean Sea (Fig.1) and the study area of this work (station PNA-10) lies at latitude 40.25323 and longitude 25.28142. Additionally, Lemnos Basin is one of the distinct sub-basins of the North Aegean Sea with maximum depth of 1550 m, located nearest to the Dardanelles.[6].

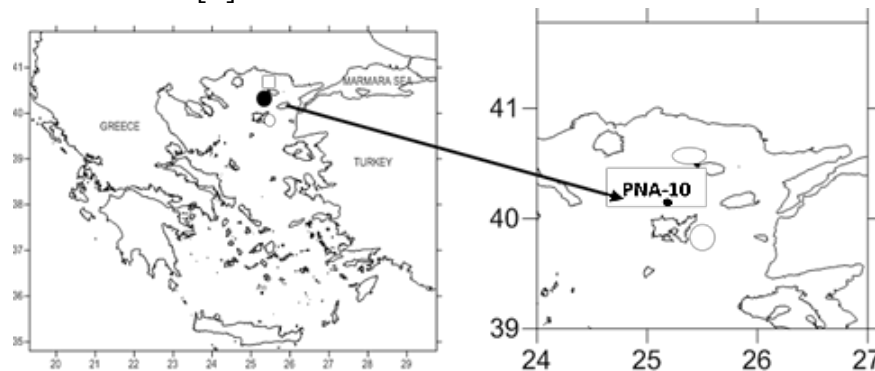


Fig. 1 The sampling location from the deep basin of Limnos in north Aegean Sea.

Nine water samples were collected from different depths, during October 2013 with HCMR's research vessel "Aegaeo", in the frame of the FP7 "PERSEUS" project. Simultaneously with the water sampling, in-situ measurements with a Conductivity-Temperature-Depth (CTD) instrument were performed. Each sample was about 20 L and was acidified immediately at pH=1.5 to minimize the adsorption of ^{137}Cs onto the bottles' surfaces. The samples were transported to the facilities of environmental radioactivity laboratory of Hellenic Centre for Marine Research for further treatment and measurements.

Table 1. In-situ measurements of temperature, salinity and dissolved oxygen along the water column. The volume of water samples are presented in the last column.

Depth (m)	Temperature (°C)	Salinity (‰)	DO ₂	Volume (L)
2	19.184	35.735	5.024	22.8
10	20.529	36.790	5.091	21.0
20	19.830	36.784	5.047	20.0
50	17.053	38.766	5.677	19.5
100	15.919	38.981	5.253	20.7
200	14.935	38.887	5.119	20.0
500	13.774	39.007	4.744	19.3
1000	13.676	39.029	4.840	40.0
1595	13.665	39.032	4.745	24.0

Methodology

The samples were prepared by a standard procedure described in details elsewhere [5]. Briefly, a quantity of 15gr of ammonium molybdate-phosphate (AMP) is diluted to the acidified water sample aiming to adsorb and pre-concentrate caesium ¹³⁷Cs. During this procedure the experimental determination of chemical efficiency of pre-concentration was obtained by additionally diluting 1ml of ¹³⁴Cs (with well-known activity concentration) in each sample. The AMP quantity containing the total adsorbed caesium (¹³⁷Cs and ¹³⁴Cs), is then collected from the water, dried and finally is placed in standard containers for gamma-ray spectrometry. The activity concentration measurements were performed by High Purity Germanium (HPGe) detector (with 50% relative efficiency) setting acquisition time for all samples 24h. The activity concentrations (A in Bq/m³) of both ¹³⁷Cs and ¹³⁴Cs were derived by analysis of the gamma-ray spectra using the SPECTRW software package [7] according to the formula (2.1).

$$A = \frac{\text{counts}}{\text{eff} \cdot \text{rec} \cdot t \cdot V \cdot I_\gamma} \quad (2.1)$$

where counts is the number of detected gamma-rays measured under the photopeaks of 661 keV (emitted by ¹³⁷Cs) and 604 keV, 795 keV (emitted by ¹³⁴Cs), eff is the absolute efficiency of the detector, rec the chemical efficiency of the pre-concentration obtained by ¹³⁴Cs measurements, t the acquisition time, V the samples' volume and I_γ the gamma-ray intensity for the aforementioned photopeaks. The energy and efficiency calibration procedures of the detector were performed using a reference ¹⁵²Eu source prepared at NCSR "Demokritos" described in details elsewhere [8].

RESULTS AND INTERPRETATION

The in-situ measurements in the water column of potential temperature (θ), salinity (S) and dissolved oxygen (DO₂) are presented in Fig. 2a, along with the activity concentrations of ¹³⁷Cs (Bq/m³) measured by the aforementioned method.

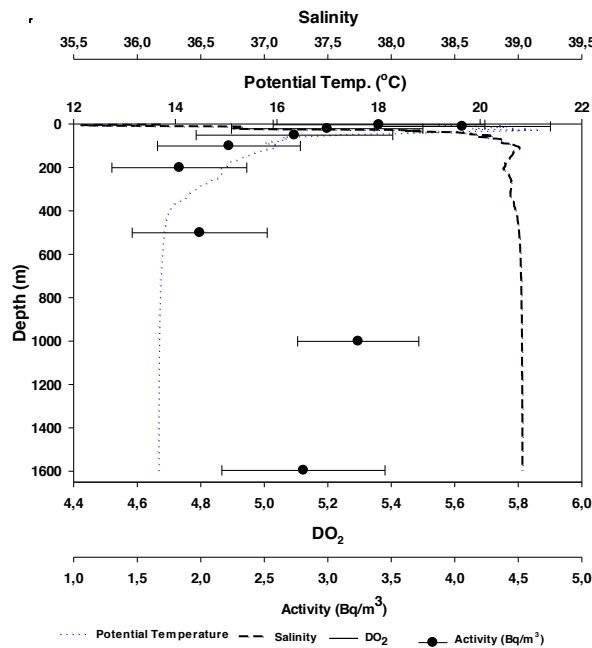


Fig. 2a Vertical profile of salinity, potential temperature, dissolved oxygen and the activity concentration ^{137}Cs (Bq/m^3) along the depth.

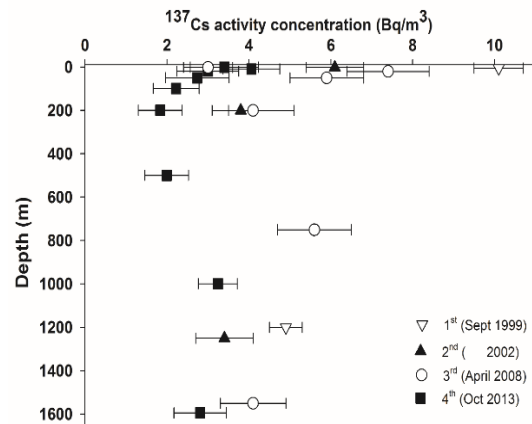


Fig. 2b Vertical profiles of ^{137}Cs activity concentration for the sampling campaigns from 1999 to 2013.

At depths of 0-40 m high values of temperature and dissolved oxygen are observed, and low values for salinity. Low salinity and high activity concentration of ^{137}Cs (^{137}Cs is rich in Black Sea waters) indicate contribution of water masses originating from the Black Sea [5]. At depths of 40-100 m salinity is increased while temperature and ^{137}Cs concentration are decreased. Levantine's water masses are expected to have temperature and salinity near 15.5°C and 39.01, respectively [3]. Similar values ($\theta=15.93$ °C, $S=39.01$) are observed in the depth range of 40-100 m indicating mixing of Black Sea with Levantine Water masses. At depths of 100-200 m potential temperature, DO₂ and ^{137}Cs concentrations decrease and salinity remains almost constant. This behavior indicates mixing of Levantine water masses with Aegean's intermediate water masses [9]. At depths of 400-1600 m, temperature and salinity do not considerably vary ($\theta=13.8$ °C and $S=39$). These almost constant values characterize a layer of homogeneous dense North Aegean's deep water. Finally, at depths of 600-1500 m concentrations of DO₂ (4.8 ml/l) and ^{137}Cs (3.2 Bq/m^3 at 1000 m) were found higher than those of deeper layers (4.7 ml/l and 2.8 Bq/m^3). This behavior is possibly attributed to Deep Water Formation events occurred during the previous years.

In Fig 2b are presented the vertical profiles of activity concentration of ^{137}Cs for the four sampling campaigns from 1999 to 2013. The highest activity concentration of ^{137}Cs is observed in the surface layers. At depth of 5 m its the temporal variation of it is observed. The aforementioned concentration decreases during the 14 years of the sampling campaigns in a higher rhythm (0.5 concentration/year) than the decay rhythm of the ^{137}Cs (0.1 concentration/year), indicating an extra mechanism (except from the decay of ^{137}Cs) for the removal of the isotope from the water column. At depth of 20 m, the activity concentration of ^{137}Cs in the 3rd campaign (7 Bq/m^3) is higher than the value of the 4th

campaign ($\sim 3 \text{ Bq/m}^3$), indicating a possible contribution of the Black Sea Water masses. Additionally, at depth of 750 m the activity concentration of ^{137}Cs in the 3rd campaign is higher than the values obtained in all the other sampling campaigns, indicating that a deep formation event occurred in the previous years. This event was verified from hydrographic data [10]. In the deep layers (below 1000 m) similar concentrations about 3 Bq/m^3 (within uncertainties) are obtained, indicating a steady state condition in the basin.

DISCUSSION-CONCLUSION

In this work ^{137}Cs concentration was used as an additional tracer to identify water masses as well as to study the mixing process of North Aegean waters with Black Sea and Levantine water masses. In the surface layer (0-40 m) the intrusion of Black Sea waters (rich in ^{137}Cs) was identified, at depths of 100-200 m mixing with Levantine waters (low ^{137}Cs) was observed and at depth of 400-1600 m homogeneous dense North Aegean waters were found. Additionally, at depths of 40-100 m the mixing of Black Sea and Levantine waters were also observed. Moreover, deep formation events resulted in the renewal of deep waters in the range of 600-1500 m. In all cases, ^{137}Cs activity concentration proved a complementary parameter. Therefore, utilization of ^{137}Cs as a tracer for water masses identification, transport and mixing proved crucial. Systematic monitoring of ^{137}Cs activity concentration may progress the understanding of water masses movement and mixing, may contribute to the evolvement of a vertical column profile model and also is important to justify new influence of radionuclides due to more recent than Chernobyl's nuclear accidents (e.g. Fukushima). The measured data were in the same level within uncertainties with previous data [1] [5] measured before Fukushima accident.

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