PLUTONIUM ISOTOPES, ^{241}Am AND ^{137}Cs ACTIVITY CONCENTRATIONS IN MARINE SEDIMENTS OF GÖKOVA BAY AEGEAN TURKISH COAST

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ABSTRACT

Samples of marine surface sediments of different grain sizes collected in Gökova , a small bay on the Aegean Turkish Coast, have been examined to measure α- and γ- radioactivity. The purpose of this research is to define a baseline study of man-made radionuclides on sediments collected along Gökova Bay, using a combination of direct gamma spectrometry, radiochemical separation and α-spectrometry.

As the artificial radionuclides, ^{137}Cs for all sediment cores samples were under detection limit. The activity concentrations of the ^{239,240}Pu were observed to be in the range of 0.13±0.017-0.85±0.15Bq.kg^{-1}. ^{241}Am and ^{238}Pu were identified at a very low level.

INTRODUCTION

The major objective of marine radioecological investigations is to help to create a scientific basis to predict the impact of radioisotopes, which have been released into rivers, lakes and seas on the marine ecosystems. The accumulation of hazardous radionuclides in the marine environment and fallout isotopes, started after the first nuclear test and has continued ever since.

The behaviour of the natural and man-made radionuclides is not well understood and there is a lack of systematic data regarding the concentration levels and their physical and chemical speciation along the Aegean Sea Coast. Our efforts were directed to study the accumulation of these radionuclides in sediments. These initial data can later be used for quantitative estimation of distribution in the environment and for evaluation of the potential hazard for the biosphere and for humans. The most important man-made radionuclides that enter the biosphere due to nuclear tests and industry is gamma emitter ^{137}Cs and the alpha emitters ^{238}Pu, $^{239+240}$Pu and ^{241}Am.

Most of radioactive elements produced in atmospheric nuclear tests decay rapidly, only those of longer half-lives remaining at the present time. Fallout ^{239,240}Pu and ^{137}Cs are good tracers to study the behavior of these radionuclides in the environment, because of their great quantity of production(up to 1980 the amount injection into the atmosphere were 13PBq and 960PBq of ^{239,240}Pu and ^{137}Cs, respectively) (Lee et al., 1995). Plutonium belongs to the group of man-made radionuclides that have attracted considerable attention from the radioecological point of view

due to their high radiotoxicity, long physical half-life, high chemical reactivity and long residence in biological systems. The principal source of plutonium radionuclides in the Gökova Bay is atmospheric fallout from nuclear weapons test. Studies on the behavior of plutonium, its distribution and ultimate fate within a given ecosystem are important for assessing the radiation doses received by man via different environmental pathways.

The determination of fission product levels, in particular ^{137}Cs, in environmental samples such as soil and sediment may give information about atmospheric circulation. Although the great majority of this radionuclide was released and deposited in the Northern Hemisphere, it was also partially transported through stratospheric circulation to the Southern Hemisphere. With expection of the 12 atmospheric nuclear weapon tests conducted by the United Kingdom in Australia and the 26 tests performed by France in Mururoa, Tuamotu Archipelago (21°S, 137°W), the 383 other atmospheric nuclear weapon tests were carried out in the Northern Hemisphere (Godoy et al., 1998). As a result of the global fallout from weapon testings in the 1960s and of the localized fallout following the reactor accident at Chernobyl in 1986, many radionuclides were released into the environment and several long-lived radionuclides were deposited on the soil surface.

MATERIAL AND METHODS

Sample collection and pre-treatment

Marine sediment samples were collected from 6 sites in Gökova Bay, as shown in Fig 1. Sediment cores from Gökova Bay were recovered using a gravity corer with transparent PVC liners. Water content was determined by the difference between wet weight and freeze-dry weight. Salt correction was applied to all weights measured. The following radionuclides were analyzed: ^{137}Cs, $^{239,240}Pu$, ^{238}Pu and ^{241}Am

Gamma analysis

For gamma spectrometric analysis, we used a HPGe detector with efficiency of 25%, FWHM of 1.85keV and detection limit of 11 Bq/kg-dry sediment for 72 000 seconds of counting times. Its efficiency was calibrated by using mixed sources (Amersham, UK) in suitable substrates and geometries. ^{137}Cs is measured directly by its corresponding gamma peak at 662keV.

Radiochemical procedures

Plutonium separation

The sample material (2.5-10g) has been properly prepared and plutonium is separated from americium by anion exchange. The chemical recoveries were determined by the use of ^{242}Pu as plutonium tracer, a leaching procedure using HNO_3, HCl solution was performed. The separated and purified $^{239+240}Pu$ and ^{238}Pu from the particulates were electrodeposited onto stainless steel

discs and measured by alpha spectrometry using silicon surface barrier detectors. Fig. 2 shows schematically the used analytical procedure. The chemical yields were in the range of 40 to 97%. The detection limit was 0.005Bq/kg dry sediment for 760000seconds of counting times.

Americium separation

Am is precipitated with calcium oxalate and extracted into DDCP (Dibutyl-N, N-Diethyl Carbamyl Phosphonate). Am is separated from the rare earths by anion exchange using a mineral acid-methanol media. The separated and purified ^{241}Am from the particulates were electrodeposited onto stainless steel discs and measured by alpha spectrometry using silicon barrier detectors. Fig. 3 shows schematically the used analytical procedure. The chemical yields were in the range of 53 to 78%. The detection limit was 0.003Bq/kg dry sediment for 630000seconds of counting times.

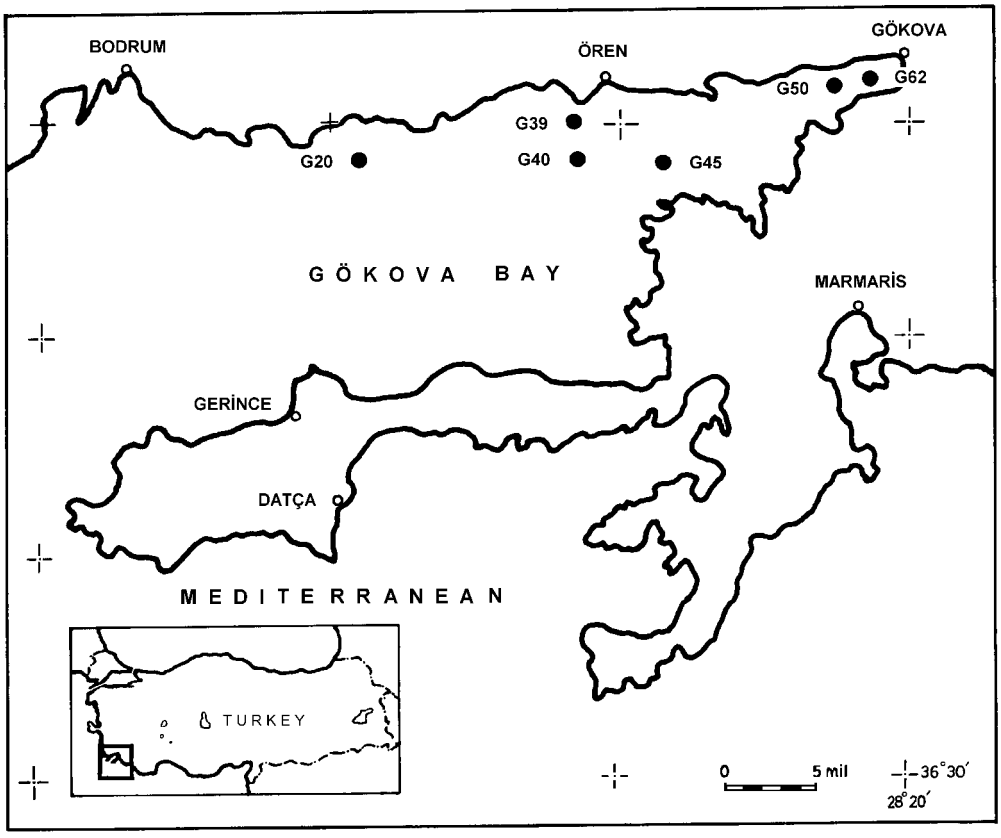

Fig.1. Locations of the sampling sites.

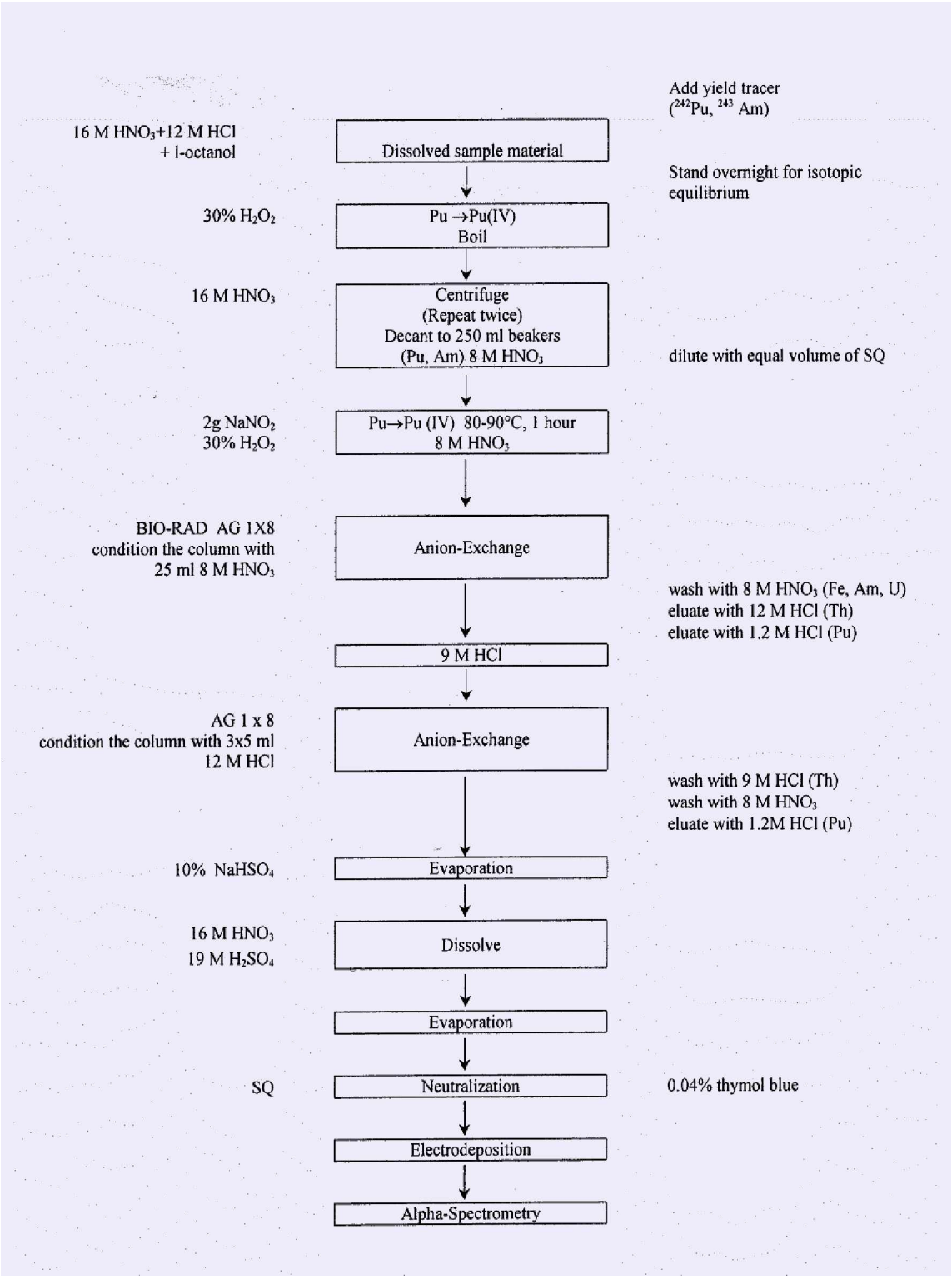

Fig. 2. Principle scheme of the analytical procedure used for plutonium determination.

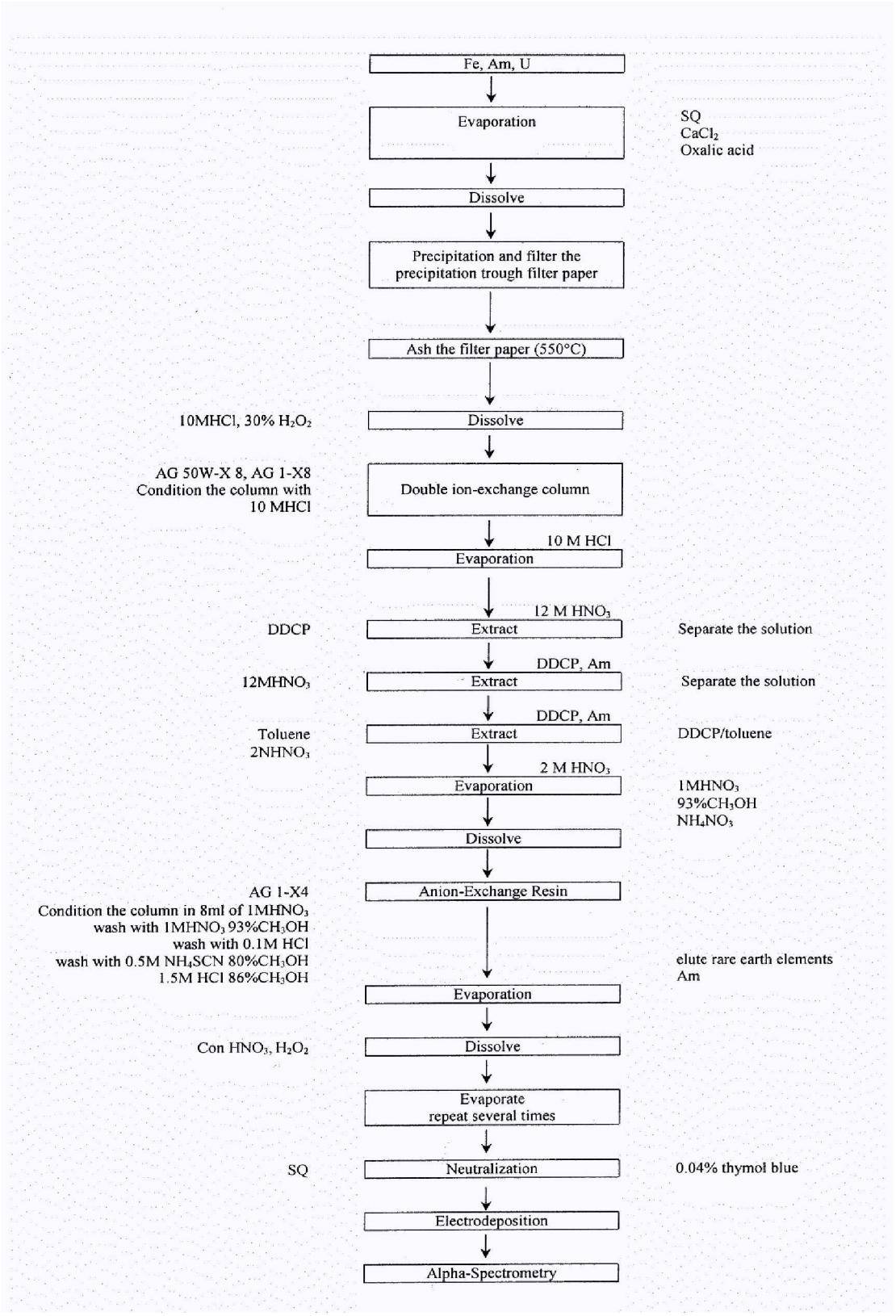

Fig. 2. Principle scheme of the analytical procedure used for americium determination.

RESULTS AND DISCUSSION

The results of artificial radionuclides ($^{239+240}$Pu, ^{238}Pu and ^{137}Cs) measured by direct γ-spectrometry, α-spectrometry in the studied sediment samples and the concentration ratios of ^{238}Pu/^{239,240}Pu and ^{241}Am/^{239,240}Pu are in given Table1. All the samples from the Bay showed less than the detectable limit for ^{137}Cs. This result confirm that there is no measurable fallout of ^{137}Cs and the deposition may influence by meteorological condition and topogrophical features of the earth's surface. Overall, the concentration levels for man-made radionuclide on surface sediments were very low, only ^{239,240}Pu, ^{238}Pu and ^{241}Am were measured at the head of the Gökova Bay (G62) and at the middle of the bay (G39). ^{241}Am were determined only in two samples collected from G-62 and G-39. The mean activity ratio ^{238}Pu/^{239,240}Pu in surface sediment was calculated to be 0.07. Similar values have been reported in global fallout and therefore they do not a sea-bed surface of contamination. Data were compared with the activity range from fallout to Algeria (Mediterranean Sea), published at literature (Noureddine et al., 1999). The Gökova Bay seems to contain comparable activity concentrations. It has been found that the ^{238}Pu/^{239,240}Pu and ^{241}Am/^{239,240}Pu activity ratios for surface sediments from Kara Sea range from 0.02 to 0.06 and 0.3 to 0.5, respectively (IAEA, 1994). Data here reported represent reference values to our country and are very important because experimental values of artificial radioactivity deposition are very sparce, specially to the Aegean Sea.

Table 1.Radionuclide concentrations (in Bq kg^{-1} dry matter) and activity ratios in surface sediments from the Gökova Bay.

	Radionuclide concentrations (Bq.kg^{-1}, dry weight)				Activity ratios	
Core	^{137}Cs	$^{239+240}$Pu	^{238}Pu	^{241}Am	^{238}Pu/$^{239+240}$Pu	^{241}Am/$^{239+240}$Pu
G-39	-	0.6±0.08	0.02±0.007	0.3±0.05	0.03±0.01	0.5±0.1
G-62	-	0.85±0.15	0.09±0.02	0.33±0.05	0.11±0.03	0.4±0.1
G-20	-	0.13±0.017	<LLD	<LLD	-	-
G-40	-	0.15±0.02	<LLD	<LLD	-	-
G-45	-	0.15±0.02	<LLD	<LLD	-	-
G-50	-	0.18±0.03	<LLD	<LLD	-	-
MA	-	0.34±0.05	0.06±0.01	0.32±0.05	-	-
LLD	11	0.005	0.002	0.003	-	-

MA: Mean Activity

LLD: Lower Limit of Detection

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