

Activity concentration of some anthropogenic radionuclides in the surface marine sediments near the Saudi coast of the Arabian (Persian) Gulf

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Activity concentrations of some anthropogenic radionuclides (^{90}Sr , ^{137}Cs , ^{238}Pu , $^{239+240}\text{Pu}$ and ^{241}Am) have been measured in the surface of marine sediments along the Saudi coast of the Arabian (Persian) Gulf. The samples were collected at different locations and water depths. The spatial distribution of the concentrations of the measured radionuclides showed a heterogeneous pattern and is independent of location or water depth. The obtained results are discussed and some conclusions are drawn.

Introduction

The issue of actual and potential radioactive contamination in the marine environment has received substantial attention during the last two decades for radiation protection purposes. They include global fallout following atmospheric weapon tests, the Chernobyl accident, discharges of radionuclides from nuclear installations, dumping of nuclear wastes into the world's oceans and seas, nuclear submarine accidents, contributions from nuclear weapons test sites, loss of nuclear weapons and radioactive sources, and satellite burn-up.¹ Due to variations in the source inputs and subsequent dispersion, mixing and transport of these radionuclides in the world oceans and seas, the marine environment has been labeled with radionuclides differing from one region to another according to the predominant source in the region.² Due to the remoteness of the Arabian (Persian) Gulf from Europe and its absence of any nuclear installations, fallout from the atmospheric testing of nuclear weapons as well as from the Chernobyl accident are the expected sources of contamination. In this context, the Gulf is a poorly studied region. It appears as an interesting area to study radiological processes, with potential sensitivity to pollutants higher than that of the oceans due to the lower dilution capacity. The Gulf is used for transportation, recreation, as a source of sea food and other variety of purposes by people living around. In addition, a considerable percentage of potable water in Saudi Arabia is supplied from desalination plants built on the western shore.

This work presents the first study of the levels of the anthropogenic radionuclides in some surface marine sediments of the Gulf. Their residence within the water column is usually due to their affinity to associate with the downward falling particulate matter. The work is a part of a comprehensive national project, which aims to measure the marine radioactivity near the Saudi coasts

of the Gulf and the Red Sea, to establish a marine radioactivity database to serve as a reference value against which the impact of future anthropogenic inputs can be assessed. The future inputs could be as a result of unplanned accidents and/or nuclear installations that present or may be constructed in the near future on the eastern bank of the Gulf.

Experimental

Study area

The Gulf is set in an extremely arid region between 24° and 30° N latitudes (Fig. 1). The Gulf is some 965 km long and has a maximum width of 338 km, with an area of about 240,000 km². It is a shallow body of water, with a maximum depth of 110 m, and due to hydrological conditions, it does not develop high waves. Despite high temperatures and humidity, the Gulf rarely sees storms and gale-force winds, and therefore, it is an easily navigable body of water.³ The Gulf is geologically a young basin of saltwater. During the Pleistocene Ice Ages, the sea level was markedly lowered, and as little as 18,000–20,000 years ago the Tigris-Euphrates river system extended through what is now known as the Gulf.⁴

Sampling

A total of 26 samples of surface marine sediments were collected from nine sampling sites on May 2004. Each sample was collected from an area of about 10⁴ cm², carefully mixed, and about 1 kg was packed in a plastic bag and sent to the laboratory for processing. The sampling sites started from Al-Khafji region at the north boundary to Al-Edaid at the south boundary (Fig. 1). A number of samples (1–4 samples) were collected from each sampling site. The sites were selected to be distributed and rather representative to the

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region. Due to the limited depth near the coast, which is a characteristic feature of the Gulf at the western shore, the samples were collected at 2–10 kilometres from the Saudi shore, almost in the open sea and at water depth ranged from 0.5–25 m. In tidal areas with shallow water, sediment sampling was conducted with a semi-cylindrical metallic sampler, while deeper bottom sediments were collected using 7-liters grab sampler. The samples were collected in polyethylene bags and transported to the laboratory for analysis.

Radiotracers

^{242}Pu and ^{243}Am standard reference materials (purchased from the National Institute for Standards and Technology, NIST, USA, under the code No. 4334G and 4332D, respectively) were diluted and used as spiking tracers to control efficiency and chemical yield in this material balance technique. Each material was supplied as 5 g aqueous acidic solution of known mass activity concentration. These concentrations were $25.67 \pm 0.72\%$ and $36.27 \pm 0.78\%$ Bq/g, for ^{242}Pu and ^{243}Am , respectively.

Analysis

The sediment samples were dried in porcelain dishes at air temperature ranging from 30–41 °C for few days, then oven-dried at 105 °C, and then coarse grained sediments were screened by a coarse sieve (1 mm). The fine grained samples were homogenized and packed in 0.5-liter Marinelli-beakers for direct ^{137}Cs measurements by γ -spectrometry using the 662 keV energy peak.

An aliquot from each sample was analyzed for ^{238}Pu , $^{239+240}\text{Pu}$, ^{241}Am and ^{90}Sr using a combined radiochemical procedure.⁵ A portion of each dried fine grained sample was ashed in a programmable muffle furnace at 550 °C for 16 hours. Six equal portions of the same ashed sample of mass 0.5 g each (total 3 g) were loaded into 6 different PTFE-vials of the microwave digestion oven. Six ml of conc. HCl, 2 ml of conc. HNO_3 and 2 ml of conc. HF were added to each vial and the vials were tightly closed, then a digestion run of 30 minutes was carried out into the oven. In case if a sample is not completely digested, additional amounts of the acid mixture were added and the digestion run was repeated for complete dissolution. The six digested fractions were combined together in a clean 500-ml PTFE-beaker. Another 3 runs (3 g each) of the same sample were carried out and combined together with the contents of the previous run to get a total of 12 grams (completely digested) in the PTFE-beaker. This final sample solution was spiked with ^{242}Pu and ^{243}Am

radiotracers. Ten ml of Sr carrier (1 mg/ml, as nitrate) was added and the solution was evaporated to dryness. The residue was converted into nitrate form by adding 5 ml of conc. HNO_3 and then evaporated to dryness. The evaporation process was repeated two times to ensure complete conversion, then the residue was dissolved in 50 ml of 8M HNO_3 and filtered using a glass fibre filter, and submitted to the chemical separation procedure, where plutonium isotopes, ^{241}Am and ^{90}Sr were sequentially separated.

Before plutonium separation, the valence adjustment of plutonium isotopes in the sample was carried out to ensure complete mixing with ^{242}Pu radiotracer. Plutonium was separated by ion-exchange chromatography using the Cl^- -form of AG 1-X8 anion-exchanger, purified from possible thorium and stripped to get the pure plutonium fraction. Plutonium source⁶ was prepared for measurement by co-precipitation with Nd^{3+} as fluoride, filtered using 0.1 μm polypropylene membrane filter, mounted on stainless steel disk to get a thin layer appropriate for alpha-spectrometry and then measured using high resolution α -spectrometer.

The effluent was evaporated to dryness and the residue was taken up in 2M HNO_3 and loaded into 10-ml TRU spec. column to separate americium isotopes and possible lanthanides. The later effluent contains possible strontium isotopes together with the other components of the sample, so it was kept for Sr analysis. Americium isotopes and lanthanides were stripped with 4M HCl solution. Then americium was purified from rare earth elements using a column packed with the Cl^- -form of AG 1-X4 anion-exchanger. The pure fraction of americium was prepared for measurement, exactly as that of plutonium,⁶ and measured using high resolution α -spectrometer.

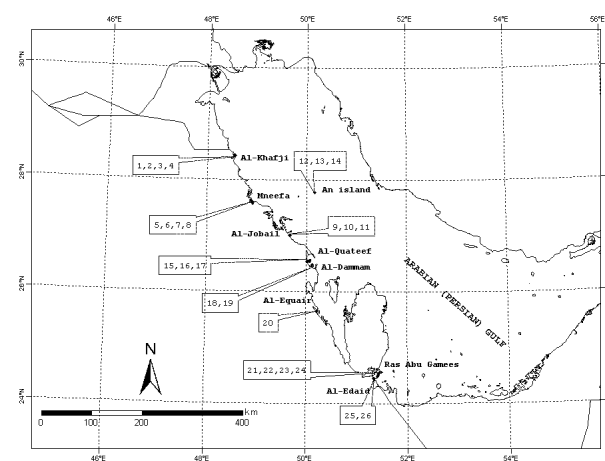


Fig. 1. Map showing the sampling sites on the Saudi coast of the Arabian (Persian) Gulf

To separate ^{90}Sr from the other components of the sample, 3 ml of 65% HNO_3 were added to the strontium-containing effluent to get the sample in 3M acid solution. The sample solution was loaded into a column containing 3 g of Sr-spec. supported crown ether to separate strontium and to strip it by distilled water. Strontium was precipitated as oxalate from alkaline medium by adding 3 ml of conc. ammonia solution. The precipitate was filtered, washed, dried and weighed for chemical recovery determination as strontium oxalate monohydrate by gravity. For strontium source preparation, the sample was dissolved in 1 ml of 1M HNO_3 , mixed with 15 ml of OptiPhase "HiSafe" 3 liquid scintillation cocktail and counted, using a liquid scintillation counter.

For quality control, the techniques were previously tested against some IAEA-reference materials⁷ for determination of ^{241}Am and the Pu α -emitters. The procedures were tested, in this work, against the IAEA-375 reference material for the determination of the analyzed radionuclides (Table 1).

Apparatus

A high resolution HPGe γ -spectrometer, Canberra type Model No. GX 4014, with about 40% relative efficiency, and 1.90 keV energy resolutions, was used for direct measurement of ^{137}Cs . Energy calibration of the detector was done using a set of γ -ray emission standard sources including ^{137}Cs (disk type). For the quantification of ^{137}Cs content in the samples, IAEA reference material (IAEA-Soil-6) in 0.5-liter Marinelli-beakers was used as comparator, where the Marinelli geometry of the beaker was utilized for measurement.

A Canberra Model 7404 "Quad Alpha" α -spectrometry system, consisting of four 450 mm² silicon surface barrier detectors was used for measurement of ^{241}Am and plutonium isotopes. Mixed standard source (purchased from Canberra) containing ^{238}U , ^{234}U , ^{238}Pu and ^{241}Am (100 dpm each), electroplated on stainless steel disk was used for energy calibration of the system.

Ultra low-level liquid scintillation spectrometer, Quantulus 1220 from Wallac, was used for ^{90}Sr measurement. Standard and blank samples were treated as the unknown samples to be used as control samples.

The lower limit of detection (LLD) for the α -emitters was 0.02 Bq/kg, based upon 12 g sample size and 48 hours count time. This limit was 0.08 Bq/kg for ^{90}Sr based upon 500-minute count time. For ^{137}Cs , the lower limit of detection was 0.06 Bq/kg, based upon 500 g sample size and 24 hours count time.

Results and discussion

The selected surface marine sediment samples were analyzed for ^{90}Sr , ^{137}Cs , ^{238}Pu , $^{239+240}\text{Pu}$ and ^{241}Am and the results are given in Table 2. For ^{90}Sr , the results showed different activity concentrations which ranged from 0.65–5.33 Bq/kg, with a mean value of 1.89 Bq/kg and a median value of 1.23 Bq/kg. These values are rather lower than (or comparable to) the values of 1.4–7.4 Bq/kg and 0.98–6.42 Bq/kg reported for ^{90}Sr levels in marine sediments collected from Ghazaouet Bay and from Barcelona and Tarragona areas in the Mediterranean, respectively.^{8,9}

The measured activity concentrations of ^{137}Cs varied also with location in a wide range. It ranged from 0.17–9.79 Bq/kg with a mean value of 3.89 Bq/kg and a median value of 3.74 Bq/kg. The obtained mean ^{137}Cs concentrations are in agreement with values reported in sea sediments in the Mediterranean (6.9–8.5 Bq/kg ash and 0.71–6.42 Bq/kg)^{8,9} and the Red Sea (0.16–10.1 Bq/kg dry weight, with a mean value of 4.1).¹⁰ The present values of ^{90}Sr and ^{137}Cs are comparable to the reported values from the pre-Chernobyl period cited in the literature for marine sediments from different regions.^{11,12}

The activity concentration of ^{238}Pu ranged from <0.02 up to 0.106 Bq/kg for the different locations along the coast, with a mean value of 0.045 Bq/kg and a median value of 0.054 Bq/kg, which falls in the range (<detection limit up to 0.06 Bq/kg) reported for sediments collected from the Red Sea,¹³ the Mediterranean^{8,9} and the Kara Sea,¹⁴ which are not suffering from nuclear waste discharge activities.

For the $^{239+240}\text{Pu}$ radionuclides, the activity concentrations varied from 0.03–0.76 Bq/kg, with a mean value of 0.34 Bq/kg and a median value of 0.35 Bq/kg. These values also lie within the range of (or rather lower than) the reported values (0.053–1.25 Bq/kg) for $^{239+240}\text{Pu}$ levels in marine sediments collected from the Red Sea,¹³ the Mediterranean Sea^{8,9} and the Kara Sea.^{14,15} Other previous studies^{16,17} reported higher concentration of $^{239+240}\text{Pu}$ in sea sediments of some areas receiving nuclear effluents directly discharged into the sea.

Table 1. The results of the determination of the radionuclides of interest in IAEA-375 reference material (in Bq/kg)

| Radionuclide | Replicates | Mean measured value | IAEA confidence interval |
|-----------------------|------------|---------------------|--------------------------|
| $^{239+240}\text{Pu}$ | 3 | 0.33 ± 0.05 | 0.26–0.34 |
| ^{238}Pu | 3 | 0.066 ± 0.019 | 0.056–0.085 |
| ^{241}Am | 3 | 0.14 ± 0.03 | 0.11–0.15 |
| ^{90}Sr | 3 | 116 ± 8 | 101–114 |
| ^{137}Cs | 1 | 5307 ± 323 | 5200–5360 |

Table 2. Location, depth and activity concentration of the radionuclides of interest of the surface sediments of the Arabian Gulf

| Sample No. | Sampling site | Location | Depth, m | Activity concentration, Bq/kg | | | | |
|------------|----------------|------------------------|----------|-------------------------------|-------------------|-------------------|-----------------------|-------------------|
| | | | | ⁹⁰ Sr | ¹³⁷ Cs | ²³⁸ Pu | ²³⁹⁺²⁴⁰ Pu | ²⁴¹ Am |
| 1 | Al-Khafji | 28 25 625N, 48 32 688E | 8 | 2.11 ± 0.28 | 3.72 ± 0.38 | 0.071 ± 0.012 | 0.26 ± 0.04 | 0.23 ± 0.03 |
| 2 | | 28 25 798N, 48 32 977E | 12 | 0.76 ± 0.13 | 0.17 ± 0.03 | 0.063 ± 0.012 | 0.43 ± 0.04 | 0.36 ± 0.05 |
| 3 | | 28 26 035N, 48 33 286E | 18 | 1.12 ± 0.21 | 1.55 ± 0.18 | <0.02 | 0.05 ± 0.01 | 0.09 ± 0.01 |
| 4 | | 28 25 754N, 48 33 872E | 25 | 2.14 ± 0.25 | 5.26 ± 0.43 | 0.046 ± 0.010 | 0.51 ± 0.06 | 0.38 ± 0.04 |
| 5 | Mincefa | 27 35 364N, 48 54 519E | 5 | 1.26 ± 0.16 | 3.17 ± 0.36 | 0.092 ± 0.014 | 0.47 ± 0.06 | 0.31 ± 0.04 |
| 6 | | 27 35 850N, 48 54 638E | 10 | 4.13 ± 0.50 | 6.12 ± 0.59 | 0.094 ± 0.014 | 0.42 ± 0.06 | 0.25 ± 0.03 |
| 7 | | 27 35 939N, 48 54 800E | 14 | 3.31 ± 0.38 | 9.79 ± 0.76 | 0.046 ± 0.011 | 0.45 ± 0.06 | 0.33 ± 0.04 |
| 8 | | 27 36 297N, 48 54 996E | 15 | 2.65 ± 0.29 | 8.80 ± 0.68 | <0.02 | 0.24 ± 0.03 | 0.15 ± 0.03 |
| 9 | Al-Jobail | 27 00 665N, 49 40 637E | 7 | 1.33 ± 0.16 | 4.25 ± 0.45 | <0.02 | 0.27 ± 0.03 | 0.19 ± 0.03 |
| 10 | | 27 00 572N, 49 41 283E | 11 | 1.16 ± 0.17 | 4.22 ± 0.41 | <0.02 | 0.26 ± 0.04 | 0.25 ± 0.03 |
| 11 | | 27 00 152N, 49 41 497E | 8 | 0.88 ± 0.12 | 3.32 ± 0.38 | 0.076 ± 0.012 | 0.48 ± 0.05 | 0.34 ± 0.04 |
| 12 | An island | 27 46 694N, 50 10 695E | 0.5 | 2.33 ± 0.26 | 4.27 ± 0.38 | 0.073 ± 0.012 | 0.26 ± 0.04 | 0.18 ± 0.03 |
| 13 | | 27 46 709N, 50 10 665E | 0.5 | 1.24 ± 0.15 | 3.36 ± 0.36 | 0.075 ± 0.013 | 0.35 ± 0.04 | 0.30 ± 0.04 |
| 14 | | 27 46 782N, 50 10 510E | 1 | 0.65 ± 0.10 | 0.95 ± 0.12 | <0.02 | 0.36 ± 0.05 | 0.27 ± 0.03 |
| 15 | | 26 32 537N, 50 01 880E | 9 | 1.03 ± 0.17 | 4.54 ± 0.45 | <0.02 | 0.03 ± 0.01 | 0.03 ± 0.01 |
| 16 | Al-Quateef | 26 32 868N, 50 02 894E | 3.5 | 1.29 ± 0.18 | 3.64 ± 0.42 | 0.049 ± 0.010 | 0.35 ± 0.05 | 0.27 ± 0.03 |
| 17 | | 26 32 289N, 50 04 419E | 13 | 3.32 ± 0.36 | 5.65 ± 0.46 | 0.046 ± 0.011 | 0.29 ± 0.04 | 0.25 ± 0.04 |
| 18 | Al-Dammam | 26 29 129N, 50 08 081E | 5 | 5.33 ± 0.51 | 4.58 ± 0.54 | 0.050 ± 0.012 | 0.19 ± 0.03 | 0.19 ± 0.03 |
| 19 | | 26 29 408N, 50 08 146E | 3 | 2.61 ± 0.33 | 3.36 ± 0.43 | 0.061 ± 0.012 | 0.24 ± 0.03 | 0.18 ± 0.03 |
| 20 | Al-Equair | 25 39 144N, 50 13 121E | 0.5 | 0.69 ± 0.12 | 1.28 ± 0.14 | <0.02 | 0.09 ± 0.01 | 0.11 ± 0.02 |
| 21 | Ras Abu Gamees | 24 32 743N, 51 27 947E | 17.5 | 1.35 ± 0.16 | 3.76 ± 0.40 | 0.061 ± 0.011 | 0.34 ± 0.04 | 0.26 ± 0.03 |
| 22 | | 24 31 861N, 51 26 718E | 5 | 2.54 ± 0.28 | 4.98 ± 0.43 | 0.046 ± 0.011 | 0.66 ± 0.08 | 0.38 ± 0.05 |
| 23 | | 24 30 347N, 51 26 724E | 9.5 | 0.92 ± 0.11 | 2.52 ± 0.31 | <0.02 | 0.18 ± 0.03 | 0.19 ± 0.03 |
| 24 | | 24 28 614N, 51 25 394E | 14.5 | 1.31 ± 0.16 | 1.38 ± 0.18 | 0.058 ± 0.012 | 0.38 ± 0.04 | 0.27 ± 0.04 |
| 25 | Al-Edaid | 24 26 704N, 51 21 740E | 14 | 1.26 ± 0.17 | 2.34 ± 0.27 | 0.072 ± 0.013 | 0.52 ± 0.06 | 0.34 ± 0.03 |
| 26 | | 24 26 770N, 51 21 046E | 7.5 | 2.54 ± 0.31 | 3.81 ± 0.32 | 0.106 ± 0.027 | 0.76 ± 0.08 | 0.42 ± 0.05 |

The obtained results of ^{241}Am vary from 0.03–0.42 Bq/kg, with a mean value of 0.25 Bq/kg and a median value of 0.26 Bq/kg, along the coast line. The obtained values are also in agreement with the reported values (0.211 ± 0.013 Bq/kg and $<0.03\text{--}0.45$) in Mediterranean sea sediments.^{8,9} In the case of ^{241}Am , prediction of the concentration of this radionuclide is more complicated by the fact that in addition to the other common factors affecting the availability of any radionuclide, ^{241}Pu decays to ^{241}Am making its availability dependent on the relative releases of both parent and daughter, and their respective mean availability times. There is no reason to expect an isotopic equilibrium with the parent ^{241}Pu ($T_{1/2} = 14.4$ y) due to the longer half live of the daughter ^{241}Am ($T_{1/2} = 432.7$ y).

Generally, the obtained data showed wide range of concentrations for each of the different radionuclides in the different locations indicating heterogenous pattern independent on location or water depth. Relatively higher concentrations of ^{90}Sr and ^{137}Cs were found in the sediments of Mineefa, whereas the relatively higher concentrations of the actinide α -emitters (Pu isotopes and ^{241}Am) were found in Al-Edaid. This may be attributed to the different absorption affinities of the different radionuclides that depend closely upon the chemical properties of the nuclide of interest, the local characteristics of sea water and the physicochemical properties of the sediments in the region. Based upon the average values of the activity concentration of the measured radionuclides, their abundance sequence was: $^{137}\text{Cs} > ^{90}\text{Sr} > ^{239+240}\text{Pu} > ^{241}\text{Am} > ^{238}\text{Pu}$. In addition, the obtained data for the measured radionuclides indicated that these rather low values may be related to the fact that neither nuclear power plants nor other direct sources lie nearby the semi-closed area of the Gulf. In addition, the smaller rainfall index and/or the geochemical composition of the sediments may also contribute in lowering these values. However, the fallout accumulation may be the main contamination source.

The activity concentration ratios as $^{238}\text{Pu}/^{239+240}\text{Pu}$, $^{239+240}\text{Pu}/^{137}\text{Cs}$ and $^{137}\text{Cs}/^{90}\text{Sr}$ ratios can be used to identify the source. It is difficult to use either $^{239+240}\text{Pu}/^{137}\text{Cs}$ or $^{137}\text{Cs}/^{90}\text{Sr}$ ratios, under the current conditions, to predict some conclusions about the source due to the difference in sorption affinity of the different radionuclides towards sediments and soils causing environmental fractionation. Selective adsorption of one of the radionuclides seriously affects these ratios with time due to the different environmental pathways of the different elements. However, the investigation of the isotopic $^{238}\text{Pu}/^{239+240}\text{Pu}$ activity ratio is much more reliable to identify the source due to the negligible impact of the environmental conditions on the fractionation of different isotopes of the same element.

Table 3. Activity concentration range, mean and median values of the radionuclides of interest (in Bq/kg)

| Radionuclide | Activity concentration range | Mean | Median |
|-----------------------|------------------------------|-------|--------|
| ^{90}Sr | 0.65–5.33 | 1.89 | 1.32 |
| ^{137}Cs | 0.17–9.79 | 3.89 | 3.74 |
| ^{238}Pu | $<0.02\text{--}0.106$ | 0.045 | 0.049 |
| $^{239+240}\text{Pu}$ | 0.03–0.76 | 0.34 | 0.35 |
| ^{241}Am | 0.03–0.42 | 0.25 | 0.26 |

Based upon the obtained mean values for plutonium isotopes (Table 3), the $^{238}\text{Pu}/^{239+240}\text{Pu}$ activity concentration ratio was in the range of 0.13 ± 0.06 . The mean isotopic ratio in the sea sediments due to fallout following global atmospheric detonation is about 0.06.¹² On the other hand, the $^{238}\text{Pu}/^{239+240}\text{Pu}$ ratio in soils contaminated by the Chernobyl accident are known to fall in the range 0.3–0.7.^{18,19} Accordingly, it may be concluded that Chernobyl accident has some contribution in the study area together with the global fallout contamination.

Conclusions

The surface marine sediments along the Saudi coast of the Arabian (Persian) Gulf have measurable concentrations of ^{90}Sr , ^{137}Cs , ^{238}Pu , $^{239+240}\text{Pu}$, and ^{241}Am . Based upon the mean values of the obtained results, their activity concentration sequence was: $^{137}\text{Cs} > ^{90}\text{Sr} > ^{239+240}\text{Pu} > ^{241}\text{Am} > ^{238}\text{Pu}$. The radioactivity levels of the different radionuclides and mean $^{238}\text{Pu}/^{239+240}\text{Pu}$ activity concentration ratio indicated a global fallout contamination due to nuclear weapon tests with some contribution from the Chernobyl accident. These data could be considered as a baseline data on marine surface sediments along the Saudi coast of the Gulf, to evaluate future releases of these radioactive pollutants into the Gulf.

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