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Elution Effect of Distilled and seawater on the Contaminated Shore Sediment

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Abstract In marine environment radionuclides exist not only at the surface layers but also at the very deep water floor such as sea water, as human activities are intimately connected with coastal areas. Nuclides and their daughter products in the marine environment has increased greatly in recent years, studies centers largely around two important environmental questions: the aspects of cycling of water masses and particular materials in sea water and potential impact on man and the environmental of the disposal of radionuclides wastes. Removing radionuclides from water requires the knowledge of the processes by which radionuclides are absorbed and desorbed from the surface of sea water sediments. This work is concerned with the absorption and desorption of ^{134}Cs and ^{89}Sr from collected shore sediments samples. Physical properties along with chemical analysis of the north east Libya Mediterranean shore sediments (NELMSS) and sea water have been studied. The samples of this area were separated into its size fractions (natural sediments, sand, silt and clay). These sediment fractions were allowed to be in equilibrium with ^{89}Sr and ^{134}Cs solutions, and studies were carried out on the contaminated sediment samples. It was found that the capacity of the different size fractions of NELMSS to sorb ^{89}Sr and ^{134}Cs in the order clay >natural sediments > silt > sand.
Keywords: Adsorption, desorption, contaminated sediment, Cesium-134, Strontium-89.

استقصاء تأثير الماء المقطر وماء البحر على الرسوبات الشاطئية الملوثة بالإشعاع

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الملخص في البيئة البحرية تتواجد النويدات المشعة في الماء سواء السطحية أو في المياه العميقة وكذلك المواد الرسوبية البحرية نتيجة للنشاط البشري المرتبط بالمناطق الساحلية، النويدات وبناتها الناتجة عنها في البيئة البحرية قد زادت بصورة كبير خلال السنوات الماضية. لذلك فإن دراسة هذا الموضوع يتمحور حول إجابة سؤالين مهمين حول البيئة البحرية: شكل دوران (تقلب) الكتل المائية وما يتعلق بالمواد المتواجدة في مياه البحر، وتأثيرها المحتمل على الانسان والبيئة البحرية نتيجة التخلص من النفايات المشعة. إن إزالة مثل هذه النويدات المشعة من الماء تحتاج إلى معرفة الطرق التي يتم بها امتصاص هذه النويدات وتَحرُّرِها من المواد الرسوبية في قعر البحر. وقد تركز هذا البحث حول دراسة عنصري السيزيوم (^{134}Cs) والسترنشيوم (^{89}Sr) في 18 موقع على امتداد الشريط الساحلي من مدينة بنغازي إلى الحدود الليبية المصرية. وقد تم التحليل الفيزيائي والكيميائي للعينات بالطرق المعتادة وباستخدام عداد جايجر أو NaI (Tl)، حيث وُجِد أن الرمل يتخلص من النويدات المشعة التي تلتصق به بمقدار يفوق كثيرا تخلص الطمي والمواد الرسوبية نتيجة لكبر حجم حبيبات الرمل، والمنطقة قيد الدراسة مكونة بصورة أساسية من الرمل. لذلك تعتبر شبه خالية من هذه المواد المشعة.
الكلمات المفتاحية: امتصاص النويدات، تَحرُّر النويدات، المواد الرسوبية، السيزيوم (^{134}Cs) والسترنشيوم (^{89}Sr).

Introduction:

Concentration of radionuclides in the atoll biota varies over wide range; whereas; very little accumulation of actinides and long lived radionuclides occur in marine biota. Although marine ecosystem represents an ultimate reservoir for radioactive waste which may enter it through several ways [Black, P. et.al, 2014, IAEA, 2017]. The differences among the concentrations of radionuclides found provides a basis for understanding the process involved in the scavenging of radionuclides from sea water and the biogeochemical cycles that are operative. Particular matter is an important factor in the removal of radionuclides from the water column, [Yamato, A. 1991].

^{89}Sr is associated with carbonates precipitated in situ, while ^{134}Cs was incorporated in the lattice of clay minerals. Each size fractions shore or bottom sediments possess certain capacity to sorb any radiation contaminant, and the uptake of these radionuclides by the same type sediments took the following form of reaction, 3 steps in sea water media and 2 steps in distilled water media,

while the sand fraction showed 2 steps reaction in both distilled or sea water media.

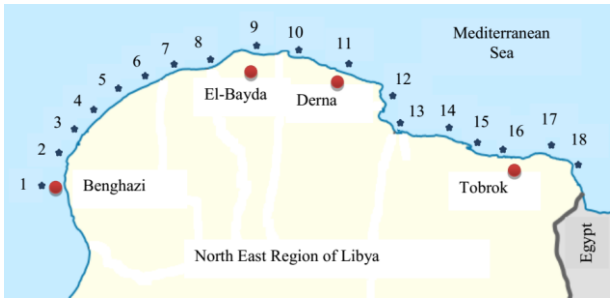


figure (1): shows locations of collected sediments from the north east region of Libya.

Sorption and desorption method:
Step one: 500 mg from different NELMSS size fractions namely natural sediment, sand, silt and clay were weighed in plastic bottles, 80 ml of 10^{-3} M/l $SrCl_2$ and $CsCl$ were added. The used isotopes were ^{89}Sr and ^{134}Cs such that the count rate was adjusted to give about 10^4 cpm/mol. The contents of each bottle were shaken for 3 days. [Kumar, B., et. al., 2007]
Step two: The sediments were separated from the supernatant by centrifugation, then washed with the least amount $(2-3\,ml)$ of absolute alcohol (96%), washing was repeated 2-3 times.
Step three: An allocated amount of 150 mg of the sediment were counted, and then transferred to plastic bottles to be shaken with 25 ml of filtered seawater or doubly distilled water.
Step four: At predetermined time intervals of contact time, aliquots of 1 ml of the supernatant were withdrawn for radioassay by using GM tube or Nal (Tl) crystal according to the tracer used. All samples were returned to the corresponding bottle after being counted.
Calculation of the percentage released for desorption at different contact time intervals was carried as follows:

$$Desorption = \frac{cpm/ml \times sol.vol.}{initial\ count\ of\ sediment} \times 100\%$$

Results and Discussion:
Different size fractions were separated and carried out on four samples each of 250 g, the result of this separation gave mean values of fractions from these samples as shown in table (1).

Table 1: fractions of the sediment size

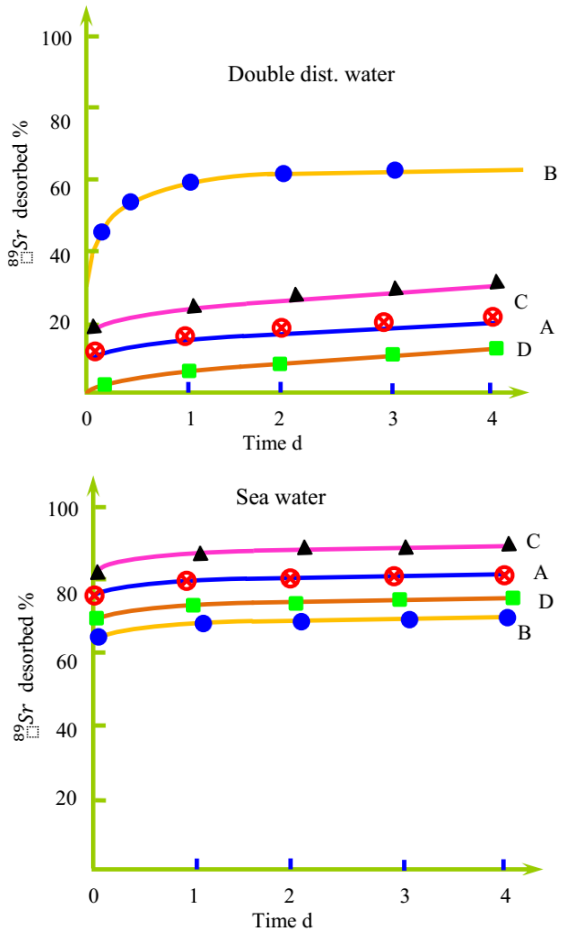
Sand	100 - 20 µm	80.85%
Silt	30 - 10 µm	14.66%
Clay	10 - 2 µm	4.39%
Total		99.90%

Thermogravimetric analysis showed that, the sediments are in good agreement with the above ratios and clearly shows that the major constitution of the sample is sand fraction.
The chemical analysis of sediments and water did not show much difference in concentrations of the elements.

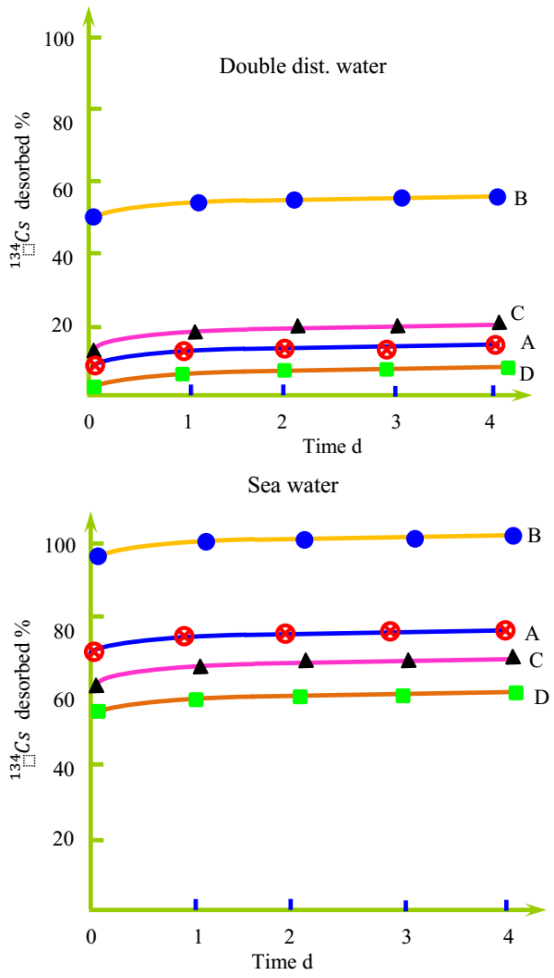
Desorption of ^{89}Sr and ^{134}Cs from denoted size fractions plotted as % desorption against time are shown in figures **2** and **3**, respectively and summarized in table **2**. The release experiments were continued until equilibrium was reached. These time periods were about 4 days for both ^{89}Sr and ^{134}Cs.

Table (2): Desorption % of ^{89}Sr and ^{134}Cs from contaminated different size fraction of North East Libya Mediterranean shore sediment (NELMSS) either by doubly distilled water or seawater

	^{89}Sr(after 4d)		^{134}Cs(after 4d)	
NELMSS Size fraction	Double distilled water	sea water	Double distilled water	sea water
Natural Sediment	17.20	85.60	17.11	75.50
Sand	75.50	76.40	56.90	96.30
Silt	20.75	87.30	20.63	75.60
Clay	10.1	79.0	11.0	65.2



Figure(2): Effect of contact time and salinity on the desorption of ^{89}Sr, in (a) doubly distilled water, (b) sea water media from different NELMSS size functions. Natural sediment (A = ⊗), sand (B = ●), silt (C = ▲), clay (D = ■).



Figure(3): Effect of contact time and salinity on the desorption of ^{134}Cs in (a) doubly distilled water, (b) sea water media from different NELMSS size functions. Natural sediment (A = ⊗), sand (B = ●), silt (C =▲), clay (D = ■).

Conclusion

The release curves showed that equilibrium for both ^{89}Sr and ^{134}Cs for NELMSS size fractions is attained much more quickly in case of seawater than in the doubly distilled water.

The desorption of ^{89}Sr and ^{134}Cs are almost similar to each other both in their values and shapes of doubly distilled water or seawater. It was also found that all NELMSS size fractions release ^{89}Sr and ^{134}Cs in sequence opposite to the sorption sequence sand > silt > natural sediment >clay. Also the desorption of ^{89}Sr and ^{134}Cs by sand fraction is in general much stronger than that by any other sediment size fraction. [Hasan, M.M., et. al., (2014), and Higashi, H., et. al. (2015)].

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