STUDIES ON SORPTION AND RELEASE OF SOME RADIONUCLIDES BY FRESH WATER SUSPENDED MATTER, EGYPT

Part III — Strontium—89

By

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ABSTRACT

The effect of some parameters on the sorption and release of Sr-89 by suspended matter of Ismailia Canal was investigated. The uptake of Sr-89 increased to an equilibrium state by increasing the concentration of the suspended matter to 500 mg/l. Organic content of suspended matter did not show any appreciable effect on the uptake of Sr-89. Sorption of Sr-89 decreased at higher concentrations of the strontium and calcium while it increased sharply at K^+ concentration of 10^{-2} m.eq./ml. Maximum uptake of Sr-89 occurred at pH 10 after one day of contact.

Filtered canal water and other extraction solutions easily removed Sr-89 from the suspended matter. Minimum percentage release values of Sr-89 occurred at pH 5.

INTRODUCTION

Several physico-chemical processes cause significant effects on radionuclides discharged into fresh water bodies. Such processes may render the radioactivity in solution to become associated with bottom sediments, organic and aquatic organisms of the receiving aquatic environment (IAEA, 1971).

Considerable research was carried out to study the transport of Sr-89 and Sr-90 in both fresh water and marine systems (DAVIS 1958; FRANK, 1963; REYNOLDS & GLOYNA, 1964; CHURCHIL el al, 1965; PICKERING et al. 1966; COLLET et al 1968; PURUSHOTHAMAN, 1971; BENES, 1972 and LERMAN, 1972). It was concluded that the most important factors affecting the uptake of this radionuclide by sediments were the cation exchange capacities of the sediments and the concentration in the water, competing cations and pH. The strontium exchange capacity of particulate matter increased with decreased particle-size and with increasing the suspended particulate concentration (WILDUNG) et al . 1972).

The main objective of this investigation is to study the effect of some parameters on the sorption and release of Sr-89 by fresh water suspended matter.

MATERIAL AND METHODS

The suspended matter used for this investigation was collected from Ismailia Canal at the site of the Egyptian Atomic Energy Establishment as previously described (ISHAK and KHALIL, 1974). The procedures used for the sorption and release studies were also described in detail (ISHAK and KHALIL, 1974) Strontium-89 spike was used in the form of a carrier-free $SrCl_2$ solution in 6 N HCl.

RESULTS

Sorption Studies

Suspended matter concentration :

The uptake of Sr-89 by the suspended matter in relation to its concentration is shown in Table (1) and Fig. (1-A). It is noticed that while the distribution coefficient (Kd) values decreased by increasing the concentration of suspended matter, the percentage uptake of Sr-89 increased with increased concentrations. Maximum uptake occurred after a contact time of one hour at low concentrations of the supended mater and after 3 days contact time at higher concentrations.

Organic content of suspended matter :

Table (2) and Fig. (1-B) illustrate the uptake of Sr-89 by original and organic-free suspeneded matter at a concentration of 500 mg/l for each and at pH 8. It is clear that the percentage uptake (Q) and distribution coefficient (Kd) values of Sr-89 were almost the same for both original and organic-free suspended matter. Also, increasing the activity of Sr-89 in solution did not appreciably change the rate of strontium uptake by the suspended matter either in the organic or in the organic-free form.

Carrier ion concentration :

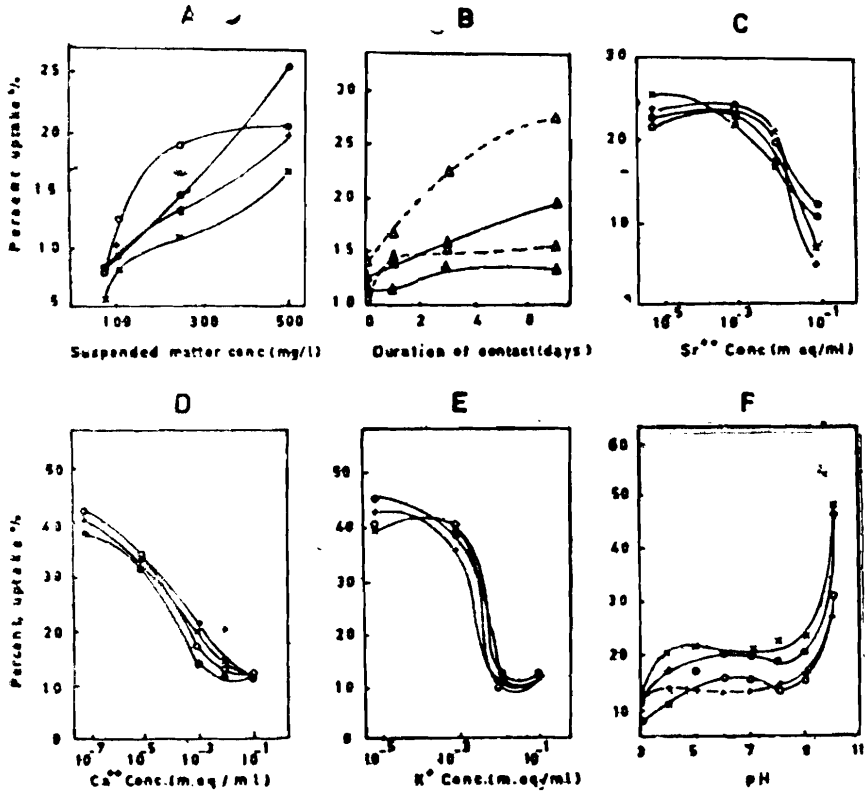
The pattern of Sr-89 uptake by suspended matter at different strontium carrier concentrations ranging between 10^{-5} to 10^{-1} m.eq/ml is shown in Table (3) and Fig. (1-C). The concentration of the suspended matter was 500 mg/1 and the experiments were carried out at pH 8. The percent uptake increased to maximum values at concentrations below 10^{-3} m.eq/ml. The distribution coefficient (Kd) and percentage uptake (Q) of Sr-89 decreased by increasing the strontium carrier concentration higher than 10 -3 m.eq./ml. Slight and variable effects on the distribution coefficient and percent uptake were observed at the different experimental contact times (Fig. 1-C).

Calcium and Potassium ions concentration :

The effect of calcium and potassium ions on the uptake of Sr-89 by the suspended matter concentration of 500 mg/1 at pH 8 is shown in Tables (4,5) and Figs. (1-D,E). It is apparent that increasing the contaot time for each experimental concentration did not show significant increase of either Kd or Q values for both calcium and potassium ions. In the presence of calcium ions, a gradual decrease of the distribution coefficient (Kd) and percentage uptake values of Sr-89 occurred by increasing the concentration of Ca^{+}+ from 10 to 10 m.eq./ml. However, for k^{+} ions maximum increase of the uptake uptake value of Sr-89 occurred at concentrations ranging between 10^{-2} and 10^{-3} m.eq/ml.

pH and contact time :

The influence of the pH of the medium on the distribution coefficient (Kd) and percentage uptake (Q) of Sr-89 by the suspended matter at a concentration of 500 mg/1 is shown in Table (6) and Fig. (1-F). The experimental results indicate that the effect of pH is rather slight within the pH values ranging from 3 to 9. However, at pH 10, a sharp increase of sorption of Sr-89 was observed. Maximum uptake of Sr-89, at the various pH values, occurred after 3 days of contact time (Fig. 1 F).



Release studies :

Extraction Solutions :

The effect of the experimental extraction solutions on the percentage release of Sr-89 from suspended matter is presented in Table (7) and Fig. (2). It is obvious that the high percentage release of strontium occurred during the early hours of contact by these extraction solutions. For the different solutions, maximum percent release values of Sr-89 ranged between 80 and 93% after days of contact.

TABLE 1. **Relationship between Concentration of Suspended Matter and its Uptake (Q) of Sr-89 and Relative Distribution Coefficient (K_d) for Different Contact Times***

Suspended Matter concentration (mg/l)	Duration of Contact							
	one hour		one day		three days		seven days	
	K_d (ml/mg)	Q	K_d (ml/mg)	Q	K_d (ml/mg)	Q	K_d (ml/mg)	Q
500	0.49±0.03	19.45±1.04	0.39±0.01	16.48±0.33	0.70±0.07	25.63±2.19	0.50±0.01	20.31±0.38
250	0.62±0 04	13.25±0.70	0.50±0 01	11.12±0.24	0.72±0.11	14.64±1.85	0.86±0.04	17.47±1.01
110	0.99±0.01	9.89±0.12	0.80±0.05	8.18±0.56	0.92±0.11	9.20±1.09	1.31±0.21	12.38±1.79
80	1.08±0.15	7.91±1.04	0.71±0.12	5.43±0.85	1.03±0.28	7.73±2.02	1.11±0.04	7.97±0.25

* **Values presented are averages of three experiments ± standard deviation.**

TABLE 2. **Relationship between the Type of Suspended Matter (organic and inorganic) on Percentage Uptake (Q) of Sr-89 and Distribution Coefficient (K^d) for Different Contact Times***

Solution Activity (cpm/ml)	Type of suspended Matter	Duration of Contact							
		one hour		one day		three days		seven days	
		K_d(ml/mg)	Q	K_d(ml/mg)	Q	K_d(ml/mg)	Q	K_d(ml/mg)	Q
3500	original**	0.29±0 02	12.56±0.41	0.32±0.04	13.75±0.99	0.37±0.06	15.49±1.37	0.48±0.04	19.46±1.34
	organic-free	0.33±0.05	13.87±1.94	0.39±0.07	16.35±2.25	0.50±0.13	22.43±4.80	0.72±0.06	27.23±1.45
7000	original**	0.26±0.03	11.27±0.61	0.25±0.04	11.01±1.06	0.32±0.02	13.95±0.38	0.32±0.07	13.80±1.43
	organic-free	0.23±0.01	10.25±0.20	0.33±0.04	14.43±1.28	0.35±0.04	15.36±1.35	0.37±0.03	15.49±0.82

* Values presented are averages of three experiments ± standard deviation.

** The organic fraction = 21.4% of the suspended matter.

Table 3.—**Relationship between the Carrier Ion Concentration and Percentage Uptake (Q) of Sr-89 by Suspended Matter and Distribution Coefficient (K_d) for Different Contact Times***

Carrier ion concentration (m.eq./ml)	Duration of Contact							
	one hour		one day		three days		seven days	
	Kd (ml/mg)	Q	Kd (ml/mg)	Q	Kd (ml/mg)	Q	Kd (ml/mg)	Q
10^{-1}	0.12±0.00	5.35±0.13	0.15±0.01	6.78±0.23	0.24±0.01	10.77±0.19	0.29±0.03	12.15±0.91
10^{-2}	0.58±0.12	21.53±3.52	0 41±0.01	17.12±0.48	0 44±0.01	17.68±0.38	0.49±0.06	19.88±1.55
10^{-3}	0.67±0.08	24.47±1.67	0.56±0.04	22.48±1.30	0.61±0.01	23.73±0.29	0.64±0.06	24.02+1.64
10^{-5}	0.64±0.09	23.73±2.29	0.66±0.26	25.43±1.61	0.60±0.04	22.90±1.17	0.57±0.13	21.62±3.76

* Values presented are averages of three experiments ± standard deviation.

TABLE 4.—Relationship between the Calcium Ion Concentration and Percentage Uptake (Q) of Sr-89 by Suspended Matter and the Distribution Coefficient (K_d) for different Contact Times*

Ca++ Concentration (m.eq./ml)	Duration of Contact							
	One hour		One day		three days		seven days	
	K_d (ml/mg)	Q	K_d (ml/mg)	Q	K_d (ml/mg)	Q	K_d (ml/mg)	Q
10^{-1}	0.25±0.02	10.95±0.79	0.26±0.02	11.68±0.67	0.26±0.04	11.35±1.56	0.28±0.03	12.05±0.71
10^{-2}	0.50±0.05	19.73±1.66	0.31±0.04	13.17±1.33	0.26±0.00	11.42±0.10	0.29±0.04	12.43±1.45
10^{-3}	0.56±0.03	21.30±1.21	0.49±0.03	19.60±0.57	0.30±0.04	13.09±1.20	0.42±0.07	17.29±2.19
10^{-5}	1.06±0.41	34.07±8.77	0.95±0.1	31.88±4.41	0.93±0.18	31.45±4.85	1.01±0.19	33.82±4.25
10^{-7}	1.46±0.32	42.50±7.18	1.35±0.11	40.50±3.92	1.18±0.21	38 01±5.01	1.41±0.08	41.65±2.03

* Values presented are averages of three experiments ± standard deviation.

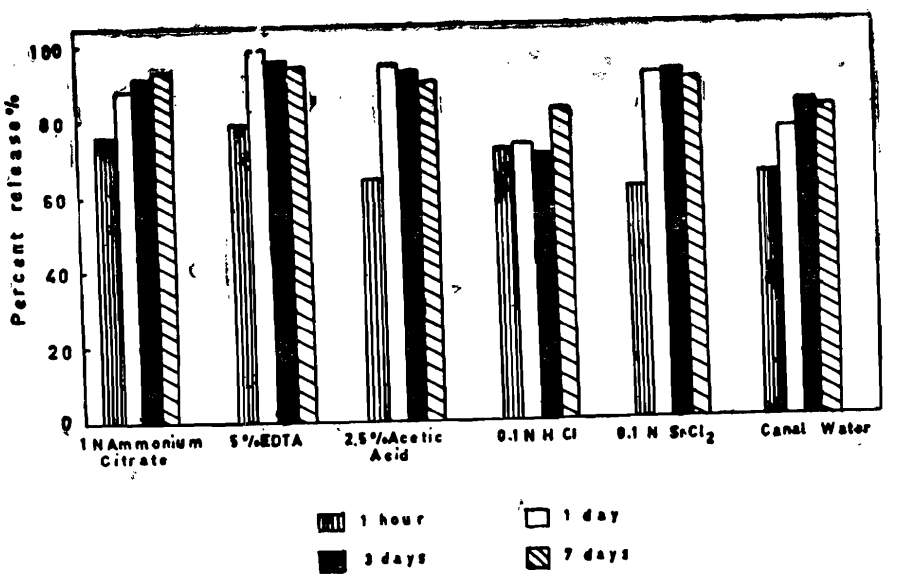


Fig. 2. Effect of some extraction solutions on percentage release of Sr-89 from suspended matter

TABLE 5.— **Relationship between the Potassium Ion Concentration and Percentage Uptake (Q) of Sr-89 by Suspended Matter and the Distribution Coeffcient (Kq) for Different Contact Times*.**

Concentration (m.eq./ml)	Duration of Contact							
	one hour		one day		three days		Seven days	
	K_d (ml/mg)	Q	K_d (ml/mg)	Q	K_d (ml/mg)	Q	K_d (ml/mg)	Q
10^{-1}	0.27	11.82	0.26	11.70	0.29	12.55	0.27	11.95
10^{-2}	0.25	11.02	0.25	11.35	0.27	11.74	0.22	9.70
10^{-3}	1.07	35.60	1.42	40.30	1.19	37.50	1.40	40.59
10^{-5}	1.51	42.56	1.37	39.33	1.61	45.25	1.40	40.10

* Each value represents an average of two experiments.

TABLE 6.— **Relationship between the pH Values of the Medium and Percentage Uptake(Q) of Sr-of P-89 by Suspended Matter and Distribution Coefficient (Kd) for Different Contact Times***

pH	Duration of Contact							
	One hour		One day		Three days		Seven days	
	K_d (ml/mg)	Q	K_d (ml/mg)	Q	K_d (ml/mg)	Q	K_d (ml/mg	Q
3	0.25	11.22	0.34	13.82	0.28	12.20	0.18	7.90
4	0.34	14.45	0.45	20.40	0.41	17.00	0.25	11.10
5	0.33	14.15	0.43	21.66	0.42	16.90	0.32	13.65
6	0.30	13.18	0.49	19.65	0.50	20.05	0.40	16.57
7	0.33	14.00	0.44	20.50	0.52	20.65	0.37	15.45
8	0.36	15.25	0.47	22.25	0.47	19.02	0.33	14.08
9	0.39	16 30	0.50	23.20	0.52	20.20	0.36	15.35
10	0.73	26.80	1.07	48.00	1.72	45.65	0.90	31.10

* Each value represents an average of two expcriments.

TABLE 7. **Effect of Different Extraction Solutions on the Percentage Release of Sr-89 from Suspended Matter***

Extraction Solutions	Duration of Contact			
	One hour	One day	Three days	Seven days
1 N Ammonium citrate	76.29±1.71	88.01±0.67	92.43±1.66	93.18±1.11
5% EDTA-Na_2 . .	79.36±2.09	98.83±0.76	95.48±5.07	93.62±5.56
2.5% Acetic acid .	64.29±3.48	93.75±2.48	93.22±2.61	88.97±6.65
0.1 N HCl	72.41±0.71	72.58±1.94	70.08±0.37	81.75±0.70
1 N HCl	80.25±0.28	76.11±0.70	73.17±0.10	82.67±0.64
0.1 N $SrCl_2$	60.70±1.56	91.28±2.28	92.38±1.48	85.27±1.22
Canal water	63.77±5.35	76.18±0.30	84.18±0.52	80.66±0.23

* Values presented are averages of three experiments ± standard deviation.

pH and contact time :

The percentage release of Sr-89 from suspended matter is shown in Table (8). The results indicate that pH - values ranging between 4 and 10 had a slight effect on the release of Sr-89 from suspended matter after seven days, (Fig. 3), However, at pH 3, Canal water removed almost all the radioactive strontium from the suspended matter. After one hour of contact and above pH 4; the percentage release of strontium decreased almost to 65%.

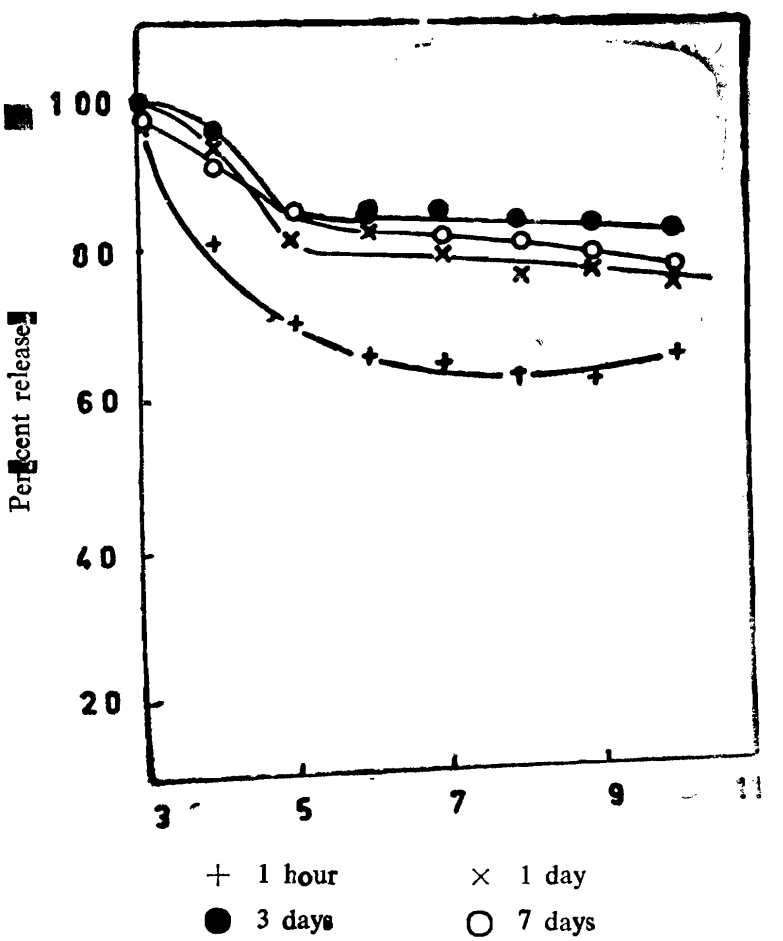


Fig. 3. Effect of pH and contact time on percentage release of Sr-89 from suspended matter

DISCUSSION

The percentage uptake of Sr-89 increased with increased concentration of suspended matter but did not exceed 25%. Almost the same results were obtained by REYNOLDS (1963) who indicated that the distribution coefficient (Kd) gave a fair representation of the distribution of the radioactive strontium between the sediment and the water. The low sorption values of Sr-89 by the suspended matter obtained in this study may be due to the fact that Sr^{++} does not form an intergral part of the clay mineral but remains as part of the extramineral aggregates as pointed out by GANAPATHY et al (1968).

TABLE 8. **Relationship between pH value of the medium and percentage release of Sr-89 by suspended matter for different contact times***

pH	Duration of contact			
	One hour	One day	Three days	Seven days
3	100.00	98.93	98.05	96 58
4	80.42	93.71	95.80	90.27
5	69.62	80.85	83.88	83.96
6	65.95	82.38	85.25	84.28
7	66 25	79.49	86.25	82.02
8	63.77	76.18	84.18	80.66
9	63.55	79.25	84.42	78.73
10	67.52	75.47	83.27	77.46

* Each value represents an average of two experiments.

The Kd values for both the original and organic-free suspended matter were almost the same at all the experimental contact periods with an average of 0.4 ml/mg. Similar results were obtained by

CLANTON (1963) who found that the original sediment samples removed more strontium than the organic-free ones. CLANTON (1968) suggested that the naturally organic fraction contributes to the overall sorption capacity of the sediments even though some of the organic molecules may block exchange sites on the clay minerals against exchange with the radionuclides.

The results of this investigation showed that in the presence of Ca^{++} ions, the percentage uptake and distribution coefficient values of Sr-89 by suspended matter gradually decreased by the increase of the concentration of calcium ions. On the other hand, in the presence of K^{+} ions, the decrease of uptake values occurred sharply at a concentration ranging from 10^{-3} to 10^{-2} m.eq./ml. REYNOLDS (1963) and MAHMOUD et al (1970) found that the divalent cations were most effective in inhibiting Sr-89 sorption by suspended matter or soil. BEETEM (1963) showed that high increase in the concentration of the monovalent ions resulted in a reduction of calcium and strontium absorption.

The maximum uptake of Sr-89 by suspended matter at Ismailia Canal was obtained at pH 10, and reached 48% after one day. This result agree with those obtained by (REYNOLDS, 1963; MORTON, 1961 and TAMURA & JACOBS, 1960) who found that Sr-89 uptake was very sensitive, particularly, in the pH range of natural water. SCHRODEOER et al (1962) and KOKOTOV (1961) also reported similar results.

All the experimental extraction solutions removed a high percentage (about 85%) of the Sr-89 contaminating suspended matter. The results obtained are similar to those reported for the Ismailia Canal bottom sediments (MAHMOUD et al., 1968). They also found that water removed Sr-89 from bottom sediments. GANAPATHY et al. (1968), showed that the leaching agents EDTA, acetic acid and HCl, leached out strontium present as labile constituents from the sediment. GOLDBERG & ARRHENIUS (1958) found that the loss of strontium by 5% EDTA solution was 100% for the mineral particles whose diameter were more than 32 μ.

Rising the pH value was followed by a decrease in the percentage release of Sr-89 to a minimum value, after which it remained constant. The maximum release was observed at pH 3 and reached

99% after 7 days. The results of the present investigation agree with those obtained by PICKERING et al. (1966), who showed that in strong acidic solutions, radioactive strontium was released while in strongly basic solutions, Sr-90 was not removed.

SUMMARY

Sorption and release of strontium-89 by fresh water suspended matter of Ismailia Canal were investigated using the batch technique.. Increased concentrations of suspended matter were followed by increase in the uptake of strontium to equilibrium state at a concentration of 500 mg/1.

A slight increase of activity sorbed by suspended matter was observed up to seven days of contact. Organic content of suspended matter had little effect on the uptake of Sr-89. Sorption of Sr-89 remained constant at low carrier concentrations while it showed a decreasing effect at higher inactive strontium concentrations. Strontium uptake by suspended matter was gradually decreased by the increase of Ca^{++} concentration, but it decreased sharply at K^{+} concentrations ranging between 10^{-3} to 10^{-2} m. eq./ml. Raising the pH value increased the uptake of Sr-89 by suspended matter to a peak value at pH 10.

The experimental extraction solutions of ammonium citrate, acetic acid and HCl were efficient in releasing the radiostrontium from contaminated suspended matter. Canal water released 80% of Sr-89 after seven days. Rising the pH value decreased the percentage release of Sr-89 to a minimum value at pH 5 after which it remained constant.

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