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RADIOACTIVE AND STABLE ELEMENT DISTRIBUTION IN MARINE BIOSPHERE OFF TARAPUR COAST[1,2]

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ABSTRACT

The distribution of natural radioactive elements like Uranium, Thorium, Radium isotopes and also some stable element distributions were measured in sea water, biological samples and sediment leaches from the samples collected off Tarapur coast during 1966, post-monsoon period. Studies were also made to obtain the seasonal fluctuations for all these elements in all the three matrices. Copper and Phosphate-phosphorus values were significantly lower in post-monsoon period than in the pre-monsoon period and this was attributed to uptake of these elements by phytoplankton blooms during this period. High values of Ra^{228} relative to its parent nuclide Th^{232} have been observed (factor of 13 to 20) in sea water and this may be due to the easy migration of Radium from sediments or from land run off. An attempt has been made to explain the Ra^{228}/Th^{232} activity ratio observed in biological species and sediment leaches. Some of the commonly available fishes have been analysed for major and trace element distribution and it was found that enriching capacity for various nuclides varies from species to species and also from season to season and this observation will have to be taken into account while computing the discharge levels of these radioisotopes.

INTRODUCTION

THE two main sources of radioactivity in the oceans are 1) fall-out due to nuclear explosions and 2) the deliberate and controlled release of liquid and solid wastes from nuclear energy industry. Moderately long-lived nuclides such as Fe-55, Mn-54, Zn-65 and Co-60 and very much long lived fission product nuclides such as Cs-137 and Sr-90 are the significant ones, contributing to the marine radioactivity. Several workers (Templeton, 1961; Pritchard, 1960; and Schaefer, 1961) reviewed the fate of these nuclides when discharged into the sea. These radionuclides once introduced into the sea are subjected to dilution, dispersion, removal through pick-up by silt and marine organisms and physical transport which will reduce the radioactive concentration levels by enormous factors. However, the removal of activity by silt acts as a concentration process and may thus be responsible for the transfer of large amount of activity to benthic organisms which consume silt as their basic food (Lowman *et al.*, 1966). From an applied point of view, investigations on these lines are of use for the consideration of local environment and biological controls in the accumulation of radioactive materials by organisms, some of which may be used as food by man or which constitute a link in the food webs that eventually lead to man (Lowman *et al.*, 1965).

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The objective of this work is to present the stable element distribution that may be used as the basis for the understanding of the fate of these radionuclides introduced into the sea (Wallanschek and Lutzen, 1964), since radionuclides discharged into the sea follow the same paths as the stable element present in the sea (Phleps, 1966). With the objective of understanding the relative distribution of element in seawater and fishes and the role the sediments play in transferring activities to benthic organisms, seawater, sediments and marine organisms have been collected from Tarapur coast 3 years before commencement of Tarapur Power Reacter in 1969. Efforts were also made to look for indicator organisms for specific elements.

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MATERIALS AND METHODS

Sampling: The location of the stations (all near shore) from which the water and sediment samples were collected is shown in Fig. 1.

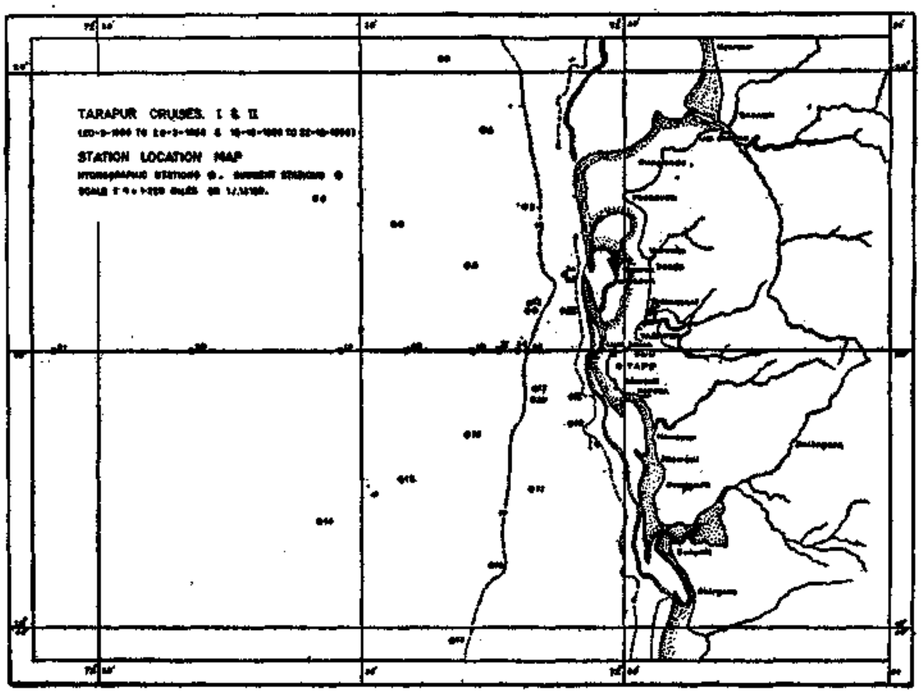

Fig. 1. Station location map.

The water samples were collected from about 1 m depth in polyethylene carboys (20 litres). 'Grab' samples of sediments were collected from these stations. The biological samples were collected from fresh hauls from nearby regions.

Processing: The water samples were filtered through Whatman No. 42 filter paper and preserved in polyethylene carboys. The biological samples were oven-dried at 105°C and then ashed at 500°C. Sediment samples were oven-dried at 105°C. A known weight of the dried samples (16-32 g) were taken and leached with 8 litres of N/20 HCl, 2.25 litres of 5% EDTA, 1N Ammonium citrate (NH_4 Cit) and 1N Ammonium acetate NH_4 Ac) as per the procedures described by Sarma *et al.* (1968). Ammonium acetate leach will give an idea of the exchangeable component of the labile part whereas N/20 HCl leach will represent the acid soluble component of the labile part of the elements in sediments including the amount present as hydrous-oxide form. EDTA and Ammonium citrate leach may give an idea of the amount element present as the labile form that can be complexed with these reagents.

Major, Minor and trace elements and natural radioactive elements in all these samples were estimated according to Sarma *et al.* (1968).

RESULTS AND DISCUSSIONS

The distribution of the elements in the three matrices viz., water, sediment and biological species is give elementwise The values are expressed in μg atom/ml in the case of seawater μg atom/gm of sediment and μg atom/gm of wet tissue in the case of biological species. The minimum and maximum values for each element in sediment are shown in brackets.

The average concentration of each individual element in seawater is calculated separately for post-monsoon and pre-monsoon periods. While calculating the concentration Factor (CF) in the sediments and biological species, the average concentration of the element obtained for the seawater samples collected during the same cruise, is used. Defining the concentration Factor (CF) for biological species as the ratio of the concentration of the element per gram of wet weight of the soft tissue to the elemental concentration per ml of seawater, the CF values for all biological species were calculated. Similarly the CF value in the sediment was defined as the ratio of the concentration of the element per gram of the sediment (dried) to the elemental concentration in seawater per ml. While calculating the CF values from the various sediment leaches the elemental concentration in each particular leach for all the sediment samples collected in the same cruise were averaged out. These averaged values were used for calculating the CF values in the sediment leaches using the average seawater concentration of the respective elements in the samples collected in the same cruise.

The biological samples were collected both in the pre-monsoon and post-monsoon times from the fresh hauls in the same region off Tarapur coast. The ash content of the biological samples (post-monsoon) are given in Table 1.

TABLE 1. *Ash content of biological samples*

Sample No.	Species	Ash %
	(POST-MONSOON)	
1.	*Pampus* sp. (Pomfret)	1.57
2.	*Arius* sp. (Cat fish)	1.31
3.	*Harpodon nehereus* (Bombay Duck)	1.27
4.	*Otolithus brunneus* (Koth fish)	2.78
5.	*Pseudosciaena dicanthus* (Ghol)	1.21
6.	*Panulirus polyphagus* (Lobster)	1.78
7.	*Cynoglossus* sp. (Flat fish)	1.43

TABLE 2. *Major and minor element content in seawater (μg atom/ml)*

Element	Seawater				
	Station numbers				
	22T	23T	24T	25T	Average
K	9.90	10.32	10.36	10.74	10.33
Mg	54.0	52.9	—	53.9	53.6
Ca	9.4	9.8	—	9.7	9 6
Sr	0.045	0.045	0.034	0.043	0.042
Fe	1.6(-4)+	1.6(-4)	—	1.6(-4)	1.6(-4)
Cu	2.4(-5)	ND	4.7(-5)	ND	3.6(-5)
Zn	—	6.25(-4)	4.06(-4)	8.10(-4)	6.10(-4)
Mn	1.40(-4)	0.90(-4)	—	1.20(-4)	1.15(-4)
Co	*3.40(-6)		*5.16(-6)		4.30(-6)
SO_4-S	28.30	28.20	28.50	28.00	28.25
Zr	**0.47(-5)				0.47(-5)
Ce	**5.70(-6)				5.70(-6)
U	1.11(-5)	1.14(-5)	1.07(-5)	1.24(-5)	1.14(-5)
Th	**1.08(-6)				1.08(-6)
Ra-226	1.51(12)	1.69(-12)	2.51(-12)	1.01(-12)	1.68-(12)
Ra-228	6.71(-15)	7.99(-15)	6.89(-15)	10.34(-15)	7.98(-15)
Cs	3.76(-6)	6.02(-6)	3.31(-6)	6.77(-6)	4.97(-6)
PO_4-P	2.25(-3)	1.32(-3)	3.20(-3)	1.60(-3)	2.10(-3)

*Composite sample of 22T and 23T, 24T and 25T

**Composite sample of four

+1.6(-4) read as 1.6×10^{-4}, likewise for others.

In Table 2 are given the major and trace element contents of seawater after monsoon. Of the major elements (K, Ca, Mg and Sr) 10 to 15% lowering in Ca and Mg contents was observed in the post-monsoon time compared to that in the pre-monsoon (Sarma *et al.*, 1968). The lowering of calcium, magnesium and strontium values after monsoon can be understood as due to the large dilution of the coastal waters and a slow rate of mixing with the open ocean waters. The Mg/Ca ratio remains unchanged. The potassium content of the post-monsoon samples is agreeing with the open ocean values reported in the literature (Sverdrup *et al.*, 1942), whereas, the pre-monsoon values are about 10% lower (9.3μg at/ml).

In post-monsoon, the Cu values are at least a factor of 10 lower than the pre-monsoon values and the two of the 4 samples (Post-monsoon) which did not show any detectable amounts of Cu have lower PO_4-P content compared to the other samples. As the samples were collected during the peak time of productivity (October-November) in this region, it may not be unreasonable to suggest that significant amounts of Cu and PO_4-P were used up by the phytoplankton. Even though Cu and Zn are known to be biologically active elements, Cu is taken up by the phytoplankton in significant amounts relative to Zn, depleting the Cu content of the sea water during the bloom periods, hardly affecting the Zn content of sea water during this time.

The high observed Th values in the post monsoon time can be attributed to the influx of this element from land run-off. Most of the other trace element concentrations (Zn, Mn, Co, Ce, Ra and Cs and PO_4 - P) during post-monsoon are close to that of pre-monsoon values within the observed variation from sample to sample.

TABLE 3. *Major and trace element content in Biological samples (µg atom/gm of wet weight)*

Elements	Ghol	Lobster	Pomfret	Cat fish	Bombay Duck	Flat fish	Koth fish
K	116.50	133.20	115.00	114 00	95.50	99.50	99 50
Mg	7.32	14 82	6.34	0.29	13 94	13.77	37.00
Ca	74 00	90.00	23.00	3.50	85.00	209.00	225.00
Sr	0 011	0.040	—	0.012	0.018	—	—
Fe	0.17	0.07	0.24	0.04	0.04	0.14	0 25
Cu	7.84(–3)+	151.0(–3)	14.00(–3)	11.00(–3)	4.50(–3)	9.68(–3)	35.6(–3)
Zn	—	0.22	—	0 13	—	—	0 05
Mn	0.16(–2)	8.30(–2)	0.93(–2)	0.59(–2)	1.14(–2)	0.87(–2)	2.06(–2)
Co	2.50(–4)	6.50(–4)	6.40(–4)	13.40(–4)	2.50(–4)	6.30(–4)	5.60(–4)
SO_4 .S	24.9*						
Zr	<1.3(–4)	—	<1.3(–4)	<1.30(–4)	—	—	—
Ce	2 70(–3)	2.25(–3)	2.74(–3)	1.45(–3)	—	—	—
U	2 18(–5)	2.83(–5)	2.12(–5)	1.29(–5)	1.25(–5)	7.50(–5)	2.74(–5)
Th	11.70(–5)	9.70(–5)	13.90(–5)	7.06(–5)	—	—	—
Ra^{226}	5 36(–12)	7.12(–12)	61.9(–12)	6.38(–12)	4.95(–12)	—	12.30(–12)
Ra^{228}	0.59(–14)	6.76(–14)	6 03(–14)	0 93(–14)	0.48(–14)	—	1.41(–14)
Cs	4.98(–4)	0.81(–4)	2.95(–4)	1.40(–4)	—	0.46(–4)	1.02(–4)
PO_4 .P	9 25*						

*Analysis is done one wet samples digested with concentra.ed H NO_3

+7.84(–3), read as 7.84×10^{-3}, likewise for others

In Table 3, are given the major and trace element content in biological samples and in Table 4 are given average elemental distributions in the various leachates of the sediments from post-monsoon times. The CF values for biological species are given in Table 5. Table 6 gives the CF values for various sediment leaches.

An examination of Table 5 reveals that the CF values in biological samples vary over a wide range from sample to sample and from element to element. The CF values for major elements vary from 1 to 20 whereas Sr and Mg are, in fact, discriminated against in almost all the cases. The relatively high CF values of 2.6 for Sr in Bombay Duck should not be taken too seriously as some small bones which were difficult to remove might have contributed to the Sr content of the Bombay Duck. Fe, Cu, Zn, Mn, Ce, Th and Co are enriched in all biological species analysed, their CF values varying from 50 to 10,000. There is a significant enhancement in the CF values for Fe and Cu in the post-monsoon time, whereas, Mn, Co and U are showing much less CF values relative to pre-monsoon. These large variations in the CF values from season to season have to be taken into account while computing the discharge levels of radioisotopes of these elements. The CF values for U in the biological samples is varying from 0.7 to 65.

From Table 6, it is seen that trace elements such as Fe, Cu, Mn, Co, Zr, Ce, Th and to a certain extent Ra are showing very high CF values in sediments, much more than the biological species. It is worthwhile to note that the CF value for PO_4-P in sediments is definitely higher in post-monsoon time as in biological material also.

Table 4. *Distribution of elements in sediment leaches (µg atom/gm)*

Element	Sediment leaches			
	Hcl	EDTA	Amm Cit	Amm. Acet
K	157	73	69	68
	(147–166)	(64–80)	(61–82)	(67–69)
Mg	740	220	291	247
	(678–789)	(199–231)	(213–361)	
Ca	629	449	391	415
	(469–750)	(434–473)	(388–393)	(401–429)
Sr	1.60	0.71	0.56	0.40
	(1.31–1.88)	(0 60–0 80)	(0.49–0.69)	(0.30–0.57)
Fe	67 2	18 0	33.0	0.38
	(54.1–77.6)	(12.2–23.0)	(26.2–37.7)	
Cu	1.02	0.53	0.55	0.07
	(0.66–1.37)	(0.49–0 59)	(0.48–0.61)	(0.06–0.09)
Zn	—	0.075	0.266	0.63
		(0.06–0.086)	(0.262–0.271)	(0.54–0.74)
Mn	6.45	3.71	3.94	2.97
	(6.16–6.73)	(3.58–3.80)	(3.35–3.89)	(2.85–3.09)
Co	0.11	0.05	0.17	0.014
	(0.10–0.11)	(0.04–0.06)	(0.09–0.29)	(0.013–0.016)
SO_4 -S	62 3	49.7	18.1	45.2
	(61.5–68.6)	(46.3–52.3)	(16.4–19.5)	(38.9–57.4)
Zr	1.97(–2)*	7.5(–2)	2.59(–2)	1.16(–2)
	(1.86–2.19)(–2)**	(4.4–9.2)(–2)	(1.86–3.29)(–2)	(0.33–2.50)(–2)
Ce	15 0(–2)	13.0(–2)	15.0(–2)	3.0(–2)
	(14.0–16.0) (–2)	(12.0–15.0)(–2)		(2.1–4.0)(–2)
U	20.3(–4)	13.0(–4)	11.0(–4)	5.4(–4)
	(18.5–23.0)(–4)	(12.0–14.0)(–4)	(9.2–13 0)(–4)	(4.6–6.7)(–4)
Th	1.35(–3)	1.75(–3)	4.48(–3)	0.2(–3)
	(0 40–2.16)(–3)	(1.21–2.02)(–3)	(3.77–5.66)(–3)	
Ra^{226}	1.12(–9)	0.64(–9)	—	0.12(–9)
	(0.89–1 50)(–9)	(0.43–0.84)(–9)	—	(0.09–0.27)(–9)
Ra^{228}	8 8(–12)	4.70(–12)	—	1.30(–12)
	(6 8–10.3)(–12)	(2.8–6.0)(–12)		(1.1–1.5)(–12)
PO_4 -P	12 33	4.11	4.20	4.06
	(11 6–12 8)	(3.84–4.29)	(3.68–4.61)	(0.74–9.97)

*1.97 (–2), read as 1.97×10^{-2} likewise for others

**(1.86–2.19)(–2), read as 1.86×10^{-2}, likewise for others

In almost all the cases, N/20 HCl leach values were giving high values. It is well known that sediments act as good scavengers for hydroxides. At a pH of 2, a good part of the hydrous-oxides dissolve except a few which do not go into solution easily such as Zr, Ti, Nb and Ta. At pH 2, a good number of metallic complexes are unstable, so that some of the trace elements present as organometallic complexes may also be contributing to the high valves of the elements in N/20 HCl leach.

Table 5. *Average concentration factors of major and trace elements for biological samples*

Sample	Cs	K	Mg	Ca	Sr	Fe	Cu	Zn	Mn	Co	PO_4-P	SO_4S	Zr	Ce	U	Th	Ra^{226}	Ra^{228}
(PRE-MONSOON, March 1966)																		
Panulirus polyphagus **(Lobster)**	68	19.0	0.6	1.3	—	497	1040	251	290	4315	—	—	—	—	—	—	—	—
Pseudosciaena diacanthus **(Ghol)**	18	—	—	—	—	250	10	41	—	1762	—	—	—	—	—	—	—	—
Limpet *(Patella)*	7	6.4	0.8	2.0	—	6095	106	148	607	3320	770	—	—	—	65	—	—	—
***Pampus* sp. (Pomfret)**	44	11.0	0.1	0.5	1.0	333	14	51	61	1928	1380	1.00	80	200	46	—	—	—
***Aplysia* sp. (Sea hare)**	—	22.0	1.6	4.8	1.6	6637	480	262	484	10140	—	—	700	530	20	—	—	—
(POST-MONSOON, October 1966)																		
Pseudoscianea diacanthus **(Ghol)**	104	9.4	0.14	7.7	0.25	1063	221	—	10	59	5130	0.90	—	474	1.9	109	3.2	0.7
Panulirus polyphagus **(Lobster)**	17	10 8	0.28	9.4	0.94	438	4254	355	55	153	—	—	—	395	2.5	90	4.2	8.4
***Pampus* sp. (Pomfret)**	60	9.3	0.12	2.4	—	1469	394	—	62	151	—	—	—	481	1.9	129	7.1	7.5
***Arius* sp. (Cat fish)**	28	9.2	—	0.4	0.29	263	310	217	39	315	—	—	—	254	1.1	65	3.8	1.2
Harpodon nehereus **(Bombay Duck)**	—	7.7	0.26	8 9	2.60	256	127	—	76	60	—	—	—	—	1.1	—	3.0	0.6
Cynoglossus sp. **(Flat fish)**	9	8.0	0.26	21.8	—	844	273	—	58	148	—	—	—	—	0.7	—	—	—
Otolithus brunneus **(Koth fish)**	21	8.0	0.69	23.4	—	1563	1014	77	137	132	—	—	—	—	2.4	—	7.3	1.8

TABLE 6. *Average concentration factors of major and trace elements for sediments*

Sample	K	Mg	Ca	Sr	Fe	Cu	Zn	Mn	Co	PO_4–P	SO_4–S	Zr	Ce	U	Th	Ra^{226}	Ra^{228}
(PRE-MONSOON, March 1966)																	
HCl leach	13.6	19	41	14	1.80(5)*	0.33(4)	940	7.60(4)	13.00(4)	2164	1.9	2.9(3)	2.8(4)	290	2.5(4)	1273	—
EDTA leach	5.3	3	36	8	0.80(5)	0.17(4)	230	5.00(4)	4.60(4)	987	2.8	2.6(3)	0.2(4)	50	1.1(4)	520	—
Amm.cit. leach	7.2	7	37	5	0.60(5)	1.10(4)	400	2.60(4)	15.00(4)	850	2.0	7.4(3)	—	420	—	274	—
Amm.acet. leach	8.5	5	17	5	480	160	620	1.20(4)	5.20(4)	260	2.8	1.9(3)	—	88	5.6(3)	130	—
(POST-MONSOON, October 1966)																	
HCl leach	12.7	13.9	66	38	4.20(5)	2.90(4)	—	4.3(4)	2.60(4)	5900	2.3	4.2(3)	2.6(4)	178	1700	667	1100
EDTA leach	5.9	4.1	47	17	1.10(5)	1.50(4)	122	2.3(4)	1.90(4)	2000	1.8	1.6(4)	2.3(4)	112	1620	381	610
Amm.cit.leach	5.6	5.5	41	15	2.10(5)	1.60(4)	434	2.40(4)	3.90(4)	2000	0.6	5.5(3)	2.6(4)	97	4148	—	—
Amm.acet.leach	5.4	4.6	43	10	2.50(5)	0.20(4)	1026	2.00(4)	0.30(4)	1900	1.6	2.5(3)	0.5(4)	47	187	107	163

*1.8(5), read as 1.8×10^5, likewise for others.

For elements such as Th, Ce, Zr and U which are known to form very stable complexes with EDTA and ammonium citrate, the labile part of these elements is close to that of HCl leach. With elements of intermediate stability with EDTA and ammonium citrate, such as Ca, Mg, Ra, Fe, Mn, etc. the HCl values were 2-3 times that of EDTA and ammonium citrate leaches. In many cases, ammonium acetate values were much lower than HCl leach values, except in cases, where ammonium acetate is known to form complexes with the cations.

The average Sr/Ca ratios in the various leaches were not varying from season to season except in the case of ammonium acetate leach. It is to be noted in all these leaches the Sr/Ca ratio is much lower than that of the seawater ratio which implies that Sr is discriminated relative to Ca in the sediments.

TABLE 7. *Natural radioactive nuclides in marine environment*

Sample	Ra^{226}	Ra^{228}	Th^{232}	(Ra^{228}/Ra^{226})	(Ra^{228}/Th^{232})
	(dpm kg^{-1})				
Seawater 22	0.74	0 795		1.074	12.90
Seawater 23	0.828	0.947	*6.16(-2)	0.784	15.37
Seawater 24	1.230	0.816		0.663	13.24
Seawater 25	0.495	1.226		2.475	19.90
HCl leach	549	1042	102.36	1.90	10.18
EDTA leach	314	577	99.93	1.84	5.78
Amm. cit. leach	—	—	203.20	—	—
Amm. acet. leach	88	154	115.30	1.75	1.34
Pseudosciaena diacanthus (Ghol)	2.63	0.698	6.70	0.266	0.104
Panulirus polyphagus (Lobster)	3.49	8.011	5 54	2.294	1 446
Pampus sp. (Pomfret)	5.83	7 145	7.93	1.226	0.901
Arius sp. (Cat fish)	3 13	1.103	4.03	0.352	0.274
Harpodon nehereus (Bombay Duck)	2.43	0.569	—	0.234	—
Cynoglossus sp. (Flaf fish)	—	—	—	—	—
Otolithus brunneus (Koth fish)	6.03	1.710	—	0.282	—

*Analysis is done on a composite sample.

The Ra^{226} and Ra^{228} activities have an uncertainity of $\pm$ 15% (statistics only) whereas Ra^{228}/Ra^{226} and Ra^{228}/Th^{232} are uncertain upto $\pm$ 20% (statistics only)

In Table 7, are given the Ra^{226}, Ra^{228} and Th^{232} activities found in water samples, sediment leaches and biological samples and also their ratios. As can be seen from this Table, the activity ratio of Ra^{228} to its parent Th^{232} varies from 13 to 20 and is much higher than that expected from the decay of Th^{232}. Similar high values for this ratio were also observed by Moore and Sackett (1964) in the Carribean, Atlantic and Pacific Coastal waters. The Ra^{228}/Ra^{226} activity ratio in sea water varies significantly from sample to sample. The abnormally high Ra^{228} content was either due to land run off or due to preferential leaching from the sediments. The Ra^{228}/Ra^{226} activity ratio in the lobster *Panulirus polyphagus* was close to the value found in various sediment leaches, whereas most of the fish samples were giving a much lower value except the Pomfret *Pampus* sp. The mobility of *Panulirus polyphangus* is highly localised and thus the activity ratio in lobster showing a value close to that of the sediment value is quite understandble. Since fishes move over large areas in the sea, the Ra^{228}/Ra^{226} ratio found in fishes, should represent an averaged Ra^{228}/Ra^{226} value over its domain of movement. Since the Ra^{228}/Ra^{226} ratio in fishes is much lower than the activity ratio found in the near coastal waters, it suggests that the Ra^{228}/Ra^{226} ratio away from the coast may be quite small, which leads us to conclude that a significant amount of Ra^{228} is coming from land run off.

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