

## FATE OF MAJOR RADIONUCLIDES IN THE LIQUID WASTES RELEASED TO COASTAL WATERS\*

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In the liquid radwaste released to coastal waters from the nuclear power stations, fission products - radiocesium and radioiodine - and activation product -  $^{60}\text{Co}$  are reported as the critical radionuclides (1,2). Behaviour of these nuclides in the sea water depends on the ionic or colloidal state of radionuclide. Part of the discharged radionuclides get absorbed as soon as it comes in contact with suspended silt of sea water and slowly settle to the bottom. Radionuclides are biologically accumulated by marine organisms and also by the algae and ultimately reach human body through consumption of sea foods. In case of sea foods like crabs and prawns significant portion of radionuclides are in the non-edible part which reduces naturally the intake by man. The behaviours of  $^{60}\text{Co}$  in sea water has been reviewed by Fukai & Murray (3) in detail.

The study presented here has been carried out in the coastal waters at the Tarapur Atomic Power Station (TAPS) operating from 1969. Controlled release of low level liquid wastes are made to the coastal waters. The water movement studies (1) have shown the oscillating nature of Tarapur near shore water with slow mixing with deeper sea waters. The coastal waters have silt varying from 50 to 200 mg/litre during non-monsoon period, but it goes as high as 1000 mg/litre during monsoon. At TAPS the liquid effluent is injected to the condenser coolant sea water at the outfall where it gets thoroughly mixed and then this diluted effluent flows out in open canal along the coast and joins the tidal waters. The activity discharged builds up in the oscillating coastal water body and gets distributed in sea water, silt, algae, fish and other foods. The chemical state of the critical nuclides in sea water, silt absorption and desorption and biological uptake of radionuclides by the marine organisms in the near shore region are described in this paper.

### FATE OF THE NUCLIDES AT THE STATION OUTFALL

The liquid waste before release to the condenser coolant is adjusted to pH of 7.5 to 8.5 and the sea waters receiving this effluent has pH of 8.0. Dialysis experiments were carried out on the radwaste sample and also on the sea water mixed radwaste using cellulose tube membrane of 4.8 millimicron pore size. In case of  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  and  $^{131}\text{I}$  in radwaste nearly 100% passed through the dialyser membrane but in case of  $^{60}\text{Co}$  only 15 to 20% passed through the membrane. Filtered sea water after spiking with radwaste and dialysis showed almost same result for  $^{137}\text{Cs}$  and  $^{131}\text{I}$ . Only 5 to 10% of  $^{60}\text{Co}$  in filtered sea water passed through the dialyser. Thus, I and Cs which are in ionic state in sea water pass through the membrane but Co

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being hydrolysed completely at pH 8, more than 90% is likely to be in hydrous oxide particles of size more than the pore diameter (4.8 millimicron) of the dialyser. In case of cobalt Fukai & Murray (3) have reported negligible formation of cobalt hydroxide in sea water contrary to our observation.

#### IMMEDIATE SILT ADSORPTION BY SUSPENDED SILT

Liquid radwaste (10 ml) of known composition was mixed with fresh sea water (2 litres) stirred for a minute and filtered through millipore filter paper. The silt with adsorbed radionuclides was retained on the paper which was quantitatively counted by gamma spectrometry. The sea water silt content was 82 mg/litre during the experiments. Observed instantaneous silt adsorptions were  $^{137}\text{Cs}$  1.5 to 5%,  $^{131}\text{I}$  1 to 3% and  $^{60}\text{Co}$  80 to 99.5%.

#### EXCHANGE OF SILT ADSORBED RADIONUCLIDES

Samples of silt deposited at the bottom of discharge canal and nearby coastal areas were studied for the exchange of activity of the radiocesium and radiocobalt adsorbed on them. Silt at Tarapur coastal area contains about 85% of particles of size 50 microns or smaller. Coarse and fine silt having adsorbed  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  were leached with 1 M ammonium acetate. The total activity on silt and in ammonium acetate leach were determined by gamma spectrometry. It was observed that only 0.5% of  $^{60}\text{Co}$  was leached from fine silt and about 5% was leached from coarse silt by amm. acetate. 30 to 40% of total  $^{137}\text{Cs}$  was leached from both coarse and fine silt by amm. acetate. Thus, cobalt adsorbed on silt is not easily available for exchange reaction where as cesium is labile to a significant extent. The exchangeability is seen decreasing with particle size. Organisms get their food through deposited silt and the labile activity in the silt would be easily available for biological uptake.

#### ACCUMULATION OF RADIONUCLIDES IN MARINE ALGAE

In the coastal environment around the radwaste release point, there is an abundant growth of algae (sea weeds) during the non-monsoon months. From the average radionuclide contents of sea water and algae, the concentration factors calculated for the three varieties of weeds are given in Table 1.

TABLE 1. Radionuclide concentration factors observed for Marine algae in the Tarapur environment.

Species	Concentration factor = $\frac{\text{pCi/kg (algae)}}{\text{pCi/litre sea water}}$		
	Range (average) for the nuclides		
	$^{131}\text{I}$	$^{134+137}\text{Cs}$	$^{60}\text{Co}$
1. Sargassum	354 - 2295 (1098)	76 - 195 (115)	235 - 920 (567)
2. Ulva Lactuca	77 - 226 (153)	3.6-33.5 (20)	65 - 292 (175)
3. Entromorpha	20 - 436 (192)	51 - 502 (275)	73 - 358 (215)

Fukai & Murray (3) have reported the concentration factor for marine algae as 2000 from the analysis of stable cobalt. The (CF) obtained here from  $^{60}\text{Co}$  concentrations is 4 to 10 times less.

#### ACCUMULATION IN ORGANISMS AND SEA FOODS

Crabs, prawns/shrimps, shell fish, onchidium (gastropod) and small fishes are the main coastal organisms caught in the Tarapur near shore region. The edible soft tissues and non-edible shells and scales were tested separately. Significant amount of radionuclide accumulations on the non-edible portions were observed. The percentage of radionuclide in the edible tissue portion of the organisms are given in Table 2.

TABLE 2. Percentage of radionuclide activity in the edible portion of the coastal sea food.

Sea food variety	Percent of edible tissue to the total wt.	Percentage activity in the edible tissue of sea food		
		$^{131}\text{I}$	$^{137}\text{Cs}$	$^{60}\text{Co}$
1. Shrimps	32.0	12.0	78.0	40.0
2. Prawns	60.0	8.0	47.0	35.0
3. Crabs	81.0	70.0	79.3	79.8
4. Shell fish	11.0	Not analysed	81.0	85.8

Significant amount of activity present in the non-edible portion is a favourable factor decreasing the human intake through sea food.

#### CONCENTRATION FACTORS FOR SOFT TISSUES OF ORGANISMS

Radionuclide concentration factors (CF) in the sea foods help in radiation exposure evaluation. In the near shore environment of Tarapur Power Station, the CFs for the radionuclides in the coastal organisms were determined under natural conditions and the results obtained are shown in Table 3.

TABLE 3. Concentration factors of radionuclides in marine organisms in the near shore environment.

Organisms	Concentration factor = $\frac{\text{pCi/kg}}{\text{pCi/litre}}$		
	$^{131}\text{I}$	$^{137}\text{Cs}$	$^{60}\text{Co}$
1. Prawns	11 to 68	6 to 41	10.5
2. Onchidium	13.2	22.5	$1.36 \times 10^5$
3. Oysters	29.7	26.7	40.0
4. Bombay Duck	11.2	6.5	15.5
5. Crabs	31 to 93	8 to 51	47 to 136

The near shore fishes taking up radiocobalt from coastal water medium have concentration factors ranging from 10 to 50 only, compared to  $10^2$  to  $10^3$  reported (3,4) for marine environment from the study of stable nuclides.

#### CONCLUSION

When  $^{131}\text{I}$ ,  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  are released to coastal waters,  $^{60}\text{Co}$  gets almost completely and irreversibly adsorbed on the silt,  $^{131}\text{I}$  and  $^{137}\text{Cs}$  get reversibly adsorbed to a fractional extent.

All the three nuclides are picked up by the crustaceans and benthos to a higher extent than fishes in the near shore region. The radionuclides considered reach the population mainly through sea food items.

The concentration factors observed for these radionuclides in coastal waters are low compared to general CFs reported (4) for sea waters. This may be due to the fact that near shore waters have about 10 to 50 times more inactive trace element content compared to off-shore waters.

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