ASSESSMENT OF ^{210}Po AND ^{210}Pb IN MARINE BIOTA OF THE MALLIPATTINAM ECOSYSTEM OF TAMILNADU, INDIA

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Abstract:

To provide baseline data on background radiation levels for the future assessment of the impact of nuclear and thermal power stations, a systematic study was carried out in the Mallipattinam ecosystem of Tamilnadu, India. Mallipattinam is located between the Kudankulam and Kalpakkam nuclear power plants and near to Tuticorin thermal power plant. Water, sediments, seaweeds, crustaceans, molluscs, and fish were collected to measure the concentrations of ^{210}Po and ^{210}Pb. The concentrations of ^{210}Po and ^{210}Pb in most samples are comparable to values reported worldwide. In fish, the concentrations of ^{210}Po and ^{210}Pb are in the range of 16-190 Bq.kg^{-1} and 8-153 Bq.kg^{-1}, respectively. The concentration factors of ^{210}Po and ^{210}Pb for the biotic components ranges from 10^3 to 10^6.

Keywords: Polonium; Lead; Palk Strait; Biota

1. Introduction:

Numerous sources of radiation (both natural and man made) can lead to internal and external human exposures. According to Clayton and Bradley (1995), about 18% of the average internal dose to humans is caused by the ingestion of ^{210}Po, along with its precursor ^{210}Pb. Lead-210 forms naturally in sediments and rocks that contain ^{238}U, as well as in the atmosphere as a by-product of radon gas. Within 10 days of its creation from radon, ^{210}Pb falls out of the atmosphere (Pietrzak-Flis and Skowronska-Smolak, 1995; Karali et al., 1996). It accumulates on the surface of the Earth, where it is stored. In general, the concentrations of ^{210}Po and ^{210}Pb are relatively low in meat and milk products, intermediate in vegetables and cereals, and much higher in most marine organisms.

The best known high background radiation area in India is the southwest coast, i.e., Kerala. The southeast coast also has some sparse distribution of high levels of radiation. On the east coast, two nuclear power plants are situated: one is functioning at Kalpakkam, and the other is under construction at Kudankulam. Therefore, a systematic study was undertaken to measure the activity of ^{210}Po and ^{210}Pb in abiotic and biotic components on the east coast. As a continuation of our previous work (Suriyanarayanan et al., 2008), we are now reporting the results from the Mallipattinam ecosystem. The study area, Mallipattinam, is situated in the Palk Strait, about 350 km south of Chennai (Madras). It is a muddy shore that serves as a protective shelter for many invertebrates. Radioactivity studies remain fragmentary at this station, and hence the present investigation was launched to determine the activity of ^{210}Po and ^{210}Pb.

2. Materials and Methods

Samples of water, sediment, seaweeds, crustaceans, molluscs and fish were collected and washed thoroughly with distilled water to free them from attached sand/silt. The soft tissues and muscles were separated from the shells, exoskeletons or bones of animals. The wet weights of the samples were recorded, and the samples were then dried in an oven at 110℃ overnight to obtain the dry weights.

2.1. Water:

2.1.1 ^{210}Po analysis: Approximately 50 L of water was filtered through Whatman 42 filter paper and acidified with concentrated HCl to pH 1. A Fe^{3+} carrier (500 mg) was added and ^{210}Po was collected on $Fe(OH)_3$ by slow addition of concentrated ammonium hydroxide with rapid stirring until reaching pH 9. Two repeated precipitations were carried out to completely capture ^{210}Po. The precipitate was dissolved in 0.5 N HCl, and ^{210}Po was deposited on both sides of a polished

silver planchette by electrochemical deposition. Alpha counting was carried out according to the procedure of Flynn (1968) and Iyengar (1983).

2.1.2. ^{210}Pb analysis: Water (~100 L) was filtered through Whatman 40 filter paper and passed through $Fe(OH)_3^-$ impregnated acrylic fiber (25 g) packed in a glass column at a flow rate of 50-60 ml/min (Krishnaswamy et al., 1972). After complete passage of the water, the column was rinsed with distilled water and the fiber transferred to a 500 ml beaker containing hot 8 N HCl. The fiber was kept immersed until it was bleached white. The leachate was evaporated to dryness on a hot plate, and the residue was dissolved in 2 M HNO_3 and made up to 50 ml. This solution was then used for the determination of ^{210}Pb via its beta-emitting daughter (Iyengar, 1983; Kamath et al., 1964).

2.2. Sediment and biological materials:

Five to twenty grams of dry weight sediment and the biological materials were homogenized and transferred to a 400 ml beaker and repeatedly digested with concentrated HNO_3:H_2O_2 (1:1) oxidizing mixture until a white ash was obtained.

2.2.1. ^{210}Po analysis: The ash was evaporated repeatedly with concentrated HCl to convert the ash into chloride medium. Then the ash was dissolved in 0.5 N HCl, and the ^{210}Po electrochemically deposited on a silver planchette (Suriyanarayanan et al., 2008). The deposition efficiency of ^{210}Po using this method varied from 95-100%, with an average efficiency of 98 ± 2%. A Radiation Counting System (ECIL-RCS 4027-A) was used with an ZnS(Ag) detector (ECIL-SP 647-A) having a background of 0.1-0.2 cpm and a counting efficiency of 25-28% for a ^{239}Pu standard source.

2.2.2. ^{210}Pb analysis: One to three grams of ash was leached with 2 N HNO_3, then filtered and made up to 50 ml. Lead-210 determination used the method of Suriyanarayanan et al., (2008).

The counting system comprised a gas flow-type GM counter with argon as the counting gas and isopropyl alcohol vapor as the quenching agent. A coincidence-anticoincidence technique was used to reduce the background (Mishra and Kerala Varma, 1963). A 15 cm thick lead shield substantially reduced background. The background was 1.5-2.0 cpm, with a counting efficiency of 40% for ^{40}K β-energy. The sample purity was checked by ^{210}Bi decay over a 120 h period, and the final activity was calculated by applying the necessary corrections for decay, chemical recovery (80 ± 4%), date of sampling, etc. Spiked experimental recoveries on water and biological samples using this method yielded an overall efficiency of 90%.

3. Results and Discussion:

Table 1 presents the activity concentrations of ^{210}Po and ^{210}Pb in water, sediments, seaweeds, sea grass, crustaceans and molluscs of the Mallipattinam ecosystem. All the ^{210}Po and ^{210}Pb results are expressed in $Bq.kg^{-1}$ (Fresh weight) and the compared results are also in same fresh/wet weight basis. In water, our values are lower than the reported values from the nearby coastal water of Mandapam (Somasundaram et al., 1998), Kalpakkam (Iyengar, 1982), and Point Calimere (Suriyanarayanan et al., 2008). Saito et al. (2003) reported that the ^{210}Pb and ^{210}Po levels in the Cananeia-Iguape Estuary of Brazil vary from 2.1 to 6.2 $mBq.l^{-1}$ and from 1.6 to 4.1 $mBq.l^{-1}$, respectively, and attributed this high activity to the presence of phosphatic rocks in that region. Taken together, it is clear that Mallipattinam coastal water does not possess high activity of ^{210}Po and ^{210}Pb.

In sediment, our ^{210}Po and ^{210}Pb activities (Table 1) were less than those of other sites, including Kalpakkam (Iyengar et al., 1983), the Red Sea (Sirelkhatim et al., 2008), the Bay of Ghazaouet on the Algerian coast (Noureddine and Baggoura, 1997), and the Bay of Bengal (Sharif et al., 1994).

The concentrations of ^{210}Po in seaweeds *Sargassum wightii* and *Grateloupia filicina* were 26 and 10 Bq.kg^{-1}, respectively, whilst the ^{210}Pb concentration was 2 Bq.kg^{-1} in both. Because seaweeds are rich in protein, a higher activity of ^{210}Po was observed rather than ^{210}Pb. The observed concentrations of ^{210}Po and ^{210}Pb in seaweeds of the Mallipattinam ecosystem are comparable with the reported values from the Mandapam (Somasundaram et al., 1998) and Tuticorin (Krishnamookthy et al., 2008) ecosystems. Miura et al. (1999) reported that the ^{210}Po value for the seaweed *Gelidium amansii* was 10 Bq.kg^{-1}, which is also comparable with our results. For sea grass (*Cymadocea serrulata*), the activities of ^{210}Po and ^{210}Pb were 11 and 2 Bq.kg^{-1}, respectively. The former value is lower than that reported by Krishnamookthy et al., (2008) (15.4 Bq.kg^{-1}) from the nearby Mandapam ecosystem.

Among crustaceans, one prawn (*Penaeus indicus*) and three crabs (*Portunus pelagicus*, *Portunus sanguinolentus* and *Ocypoda*) were selected for measurement of ^{210}Po and ^{210}Pb concentrations. The concentrations in prawns of Mallipattinam were higher than those reported in Kalpakkam (Iyengar et al., 1983), Mumbai (Mishra et al., 2009), Goa (Avadhani et al., 2001) and the Sao Paulo region of Brazil (Saito et al., 2003). In crabs, the ^{210}Po activity in muscle and exoskeleton ranged between 265-324 and 55-96 Bq.kg^{-1}, respectively, while the ^{210}Pb activity ranged between 3-5 Bq.kg^{-1} in muscle and 3-10 Bq.kg^{-1} in exoskeleton. The measured ^{210}Po activity was much higher than that recorded on the Kalpakkam coast (Kannan et al., 2001).

In molluscs, five species (*Strombus canarium, Tonna dolium, Sepia elliptica, Scapharca inaequivalvis,* and *Gafrarium dispar*) were taken for measurement of ^{210}Po and ^{210}Pb activities (Table 1). Among the five species, *G. dispar* possessed the highest ^{210}Po activity (415 Bq.kg^{-1} in soft tissue). In all varieties of crustaceans and molluscs, the ^{210}Po activity was higher in soft tissue than in the shell/exoskeleton, whereas the ^{210}Pb activity was vice versa. Lead is a bone

seeker and has a long residence time in the skeleton. The higher concentration of lead in the skeleton may be linked to its capacity to replace calcium. Similar results have been reported by Asokan et al., (1992).

Fish play a significant role in the transfer of radionuclides to humans. Nine of the most abundant fish varieties in the Mallipattinam ecosystem were collected; the ^{210}Po and ^{210}Pb activities are shown in Table 2. Among the varieties, *Sardinella longiceps* (oil sardine, plankton feeder) recorded the highest ^{210}Po activity in its muscle (190 Bq.kg^{-1}), whereas *Sphyraena barracuda* (ray-finned fish, carnivore) recorded the highest ^{210}Pb activity (97 Bq.kg^{-1}) in its bone. Both the ^{210}Po and ^{210}Pb activities of fish on the Mallipattinam coast are very high compared with values reported in the literature. In Syria, Al-Masri et al., (2000) reported the ^{210}Po activity in *Sardinella sp.* with a range of 10.65-18.45 Bq.kg^{-1}, which is very low compared with those obtained here. However, Khan et al., (2007) reported that the *S. longiceps* has high natural gamma radiation activity in the coast around Kudankulam of the Gulf of Mannar, which is just 200 km south of Mallipattinam. Plankton feeders tend to accumulate higher ^{210}Po than the carnivores, omnivores, and detritus feeders, which is the case in the present study. Polonium-210 levels in the muscles of marine fish samples obtained in the present study were comparable with those obtained by Bangera and Rudran (1995) (520-25,000 mBq.kg^{-1}) and Kannan et al., (2001) (1100-29,600 mBq.kg^{-1}). In all fish, the concentrations of ^{210}Po in muscle were higher than in bones. Concentrations of ^{210}Po in muscle are important for humans, who consume mostly the muscles of fish. This accounts for the high rate of dose transfer to humans (Iyengar, 1983).

The ^{210}Po and ^{210}Pb concentration factors for our marine organisms ranged between 10^3 and 10^5. Generally, higher concentration factors were seen in mollusc species than in seaweeds and crustaceans, confirming that they could serve as good indicators of radionuclide pollutants in

a marine environment. Similar results were found by Saito et al., (2003) in Sao Paulo, Brazil, where molluscs were found to have a higher concentration factor. The concentration factors for fish are in the range of 1-50 ($x10^4$), which is higher than the range reported for fish by Saito et al., (2003) and the MARDOS Project (1994). From these results, it is clear that the ^{210}Po and ^{210}Pb concentrations in water, seaweeds, sea grass, crustaceans, and molluscs of the Mallipattinam ecosystem are comparable with those reported elsewhere, except for plankton feeder fish, which had higher concentrations than reported values.

4. Conclusion:

Polonium-210 and Lead-210 concentrations were measured in the biota of the Mallipattinam ecosystem to obtain a better understanding of the regional ingestion dose as well as to provide a baseline to assess any changes that may occur as a consequence of the development of nuclear facilities in this area. The ^{210}Po and ^{210}Pb activities were found to be low in water, sediments, and biota, except for some varieties of fish such as *S. longiceps*. The soft tissues of crustaceans and molluscs were found to accumulate contaminants at concentrations significantly higher than those of ambient water. The differences in the concentrations of ^{210}Po and ^{210}Pb in biota are probably due to differences in lifestyle and feeding habits.

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Table 1: The ^{210}Po and ^{210}Pb activities and concentration factors of biota from the Mallipattinam ecosystem.

Name of samples	Number of samples	Activity (Bq.kg^{-1}F.W)		Concentration factor[a]	
		^{210}Po	^{210}Pb	^{210}Po	^{210}Pb
Water	12	0.4	0.6	-	-
Sediments	8	4	1	-	-
Seaweeds					
Sargassum wightii	8	26 ± 2	2 ± 1	7 x 10^4	3 x 10^3
Grateloupia filicina	8	10 ± 1	2 ± 1	3 x 10^4	3 x 10^3
Sea grass					
Cymadocea serrulata	8	11 ± 1	2 ± 1	3 x 10^4	3 x 10^3
Crustaceans					
Prawn					
Penaeus indicus	8				
Muscle		119 ± 6	6 ± 1	3 x 10^5	10 x 10^3
Exoskeleton		77 ± 4	13 ± 2	2 x 10^5	2 x 10^4
Crabs					
Portunus pelagicus	4				
Muscle		265 ± 9	5 ± 1	7 x 10^5	8 x 10^3
Exoskeleton		55 ± 2	3 ± 1	1 x 10^5	8 x 10^3
Portunus sanguinolentus	4				
Muscle		300 ± 4	3 ± 1	7 x 10^5	6 x 10^3
Exoskeleton		96 ± 2	10 ± 2	2 x 10^5	2 x 10^5
Ocypoda	4				
Muscle		324 ± 12	3 ± 1	8 x 10^5	6 x 10^3
Exoskeleton		90 ± 1	5 ± 1	2 x 10^4	8 x 10^3
Molluscs					
Gastropods					
Strombus canarium	8				
Soft tissue		11 ± 2	2 ± 1	3 x 10^4	3 x 10^3
Shell		3 ± 1	3 ± 1	8 x 10^3	6 x 10^3
Tonna dolium	8				
Soft tissue		132 ± 16	2 ± 1	3 x 10^5	3 x 10^3
Shell		4 ± 1	4 ± 1	10 x 10^3	7 x 10^3
Cephalopod					
Sepia elliptica	8				
Soft tissue		45 ± 2	27 ± 2	1 x 10^5	5 x 10^4
Shell		31 ± 2	31 ± 2	8 x 10^4	5 x 10^4
Scapharca inaequivalvis	8				
Soft tissue		225 ± 12	9 ± 3	6 x 10^5	2 x 10^4
Shell		4 ± 1	10 ± 1	10 x10^3	2 x 10^4
Gafrarium dispar	8				
Soft tissue		415 ± 23	12 ± 6	1 x 10^6	2 x 10^4
Shell		3 ± 1	16 ± 1	7 x 10^3	3 x 10^4

[a] Concentration factor = ^{210}Pb activity in biota (Bq.kg^{-1}F.W)/^{210}Pb activity in water (Bq.l^{-1})

Table 2: The ^{210}Po and ^{210}Pb activities and concentration factors in selected fishes of the Mallipattinam ecosystem.

Name of samples	Number of samples	Activity (Bq.kg^{-1}F.W)		Concentration factor[a]	
		^{210}Po	^{210}Pb	^{210}Po	^{210}Pb
Argyrosomus amoyensis	12				
Muscle		16 ± 1	5 ± 1	4 x 10^4	8 x 10^3
Bone		8 ± 1	6 ± 1	2 x 10^4	10 x 10^3
Apolectus niger	12				
Muscle		24 ± 1	2 ± 1	6 x 10^4	3 x 10^3
Bone		22 ± 2	3 ± 1	6 x 10^4	5 x 10^3
Synaptura commersoniana	10				
Muscle		38 ± 2	16 ± 1	9 x 10^4	3 x 10^4
Bone		24 ± 2	17 ± 1	6 x 10^4	3 x 10^4
Lutianus johnii	12				
Muscle		38 ± 2	17 ± 2	9 x 10^4	3 x 10^4
Bone		26 ± 2	19 ± 2	7 x 10^4	3 x 10^4
Liza vaigiensis	12				
Muscle		41 ± 2	1 ± 0.5	1 x 10^4	2 x 10^3
Bone		21 ± 2	4 ± 1	5 x 10^4	7 x10^4
Sphyraena barracudda	10				
Muscle		44 ± 1	10 ± 1	1 x 10^5	2 x 10^4
Bone		40 ± 3	97 ± 5	1 x 10^4	2 x 10^4
Tachysurus jella	10				
Muscle		44 ± 2	3 ± 1	1 x 10^4	5 x 10^3
Bone		21 ± 2	4 ± 1	5 x 10^4	6 x 10^3
Eleutheronema tetradactylum	10				
Muscle		116 ± 7	1 ± 0.5	3 x 10^5	2 x 10^3
Bone		62 ± 4	14 ± 3	2 x 10^4	2 x 10^4
Sardinella longiceps	12				
Muscle		190 ± 5	13 ± 1	5 x 10^5	2 x 10^4
Bone		153 ± 8	44 ± 5	4 x 10^5	7 x 10^4

[a] Concentration factor = ^{210}Pb activity in biota (Bq.kg^{-1} F.W)/^{210}Pb activity in water (Bq.l^{-1})