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Baseline

Occurrence of ^{210}Po in periwinkle (*Littorina undulata*, Gray, 1839) collected from Kudankulam (Gulf of Mannar (GOM), Southeast coast of India)

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ABSTRACT

Polonium-210 activity concentration was analysed in the whole body tissue of periwinkle *Littorina undulata* collected from intertidal rocky shore along Kudankulam coast. We carried out the study for a period of 12 months (2011–2012) focusing on three seasons. ^{210}Po was found non-uniformly distributed among the periwinkles depending on the allometry. The ^{210}Po accumulation showed a significant difference between seasons ($p < 0.05$). Smaller sized winkles registered higher activity of ^{210}Po compared to larger ones ($p < 0.05$). The overall activity range of ^{210}Po varied from 13.5 to 58.9 Bq/kg (wet). The activity of ^{210}Po was also quantified in seawater and intertidal sediments to calculate the biological concentration factor (BCF) and radiation dose rate. The dose rate to the winkles was performed using ERICA Assessment Tool and it was within the prescribed limit. The intake of ^{210}Po through periwinkles delivered an effective dose in the range of 2.2–9.6 μSv/y to human beings.

Among naturally occurring radionuclides, ^{210}Po, in marine biota attains great significance due to its radiotoxic properties. ^{210}Po ($t_{1/2}$ = 138.4 days) is a naturally occurring radionuclide formed by the beta decay of its grandparent ^{210}Pb ($t_{1/2}$ = 22 years) via $_{210}$Bi of the ^{238}U decay chain. The concentration of ^{210}Po in marine food has received much interest from the marine scientific community because of the high radioactive dose it delivers to marine organisms than anthropogenic radionuclides released into coastal waters (Aarkrog et al., 1997; Khan and Wesley, 2011a). ^{210}Po has a tendency to get accumulated in the edible portions of marine organisms, and is considered to be the most important contributors of radiation dose received by humans via fish and shellfish consumption (Mishra et al., 2009; Strok and Smodis, 2011). The ^{210}Po accumulated in marine organisms is generally known to derive from the food chain (Carvalho and Fowler, 1994; Carvalho, 2011), and significant differences are noted in its accumulation in different species. Therefore, assessment of radioactive elements in the natural environment and their effects on living organisms has become one of the most important issues in environmental monitoring programmes around nuclear installations in recent years.

Marine molluscs form an important component of the marine food web. They act as sentinels and considered one of the abundant primary consumers in biogenic habitats. The common periwinkle *Littorina undulata* is a grazing intertidal mollusc with restricted mobility and are especially sensitive to pollution and habitat degradation because they reside in the shore environment where chemical contaminants accumulate. In general, molluscs have the potential to accumulate significant amounts of radionuclides which may be biomagnified in the food chain to higher trophic levels including human consumers (Karouna-Renier et al., 2007). They also serve as a potential bioindicator species in coastal areas by reflecting the effects of natural and anthropogenic stressors (Gerhart and Bert, 2009).

Although many studies on the level of ^{210}Po in various molluscs have been conducted in the marine environment, sufficient data are not available for periwinkles along the southern coast of India. Previous studies have focused on commonly available fishes, crustaceans and molluscs and not on the particular species. Studies on individual species helps in setting a bioindicator for a particular region, thus helps in future safety action plans. Therefore, we investigated to gain a better knowledge on; (i) the distribution of ^{210}Po in the soft tissue of periwinkles based on season, site and size, (ii) to study the internal dose rate received by periwinkles, and (iii) to estimate the effective dose received by human beings consuming the molluscs. This study is considered as a baseline data for this coast, since a mega nuclear power hub is getting its criticality (2 × 1000 MWe).

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Samples of periwinkle, *L. undulata* were collected along the Kudankulam coastal region at three sites. This region is located in the southeastern coast of peninsular India, in the distal end of the Gulf of Mannar Biosphere Reserve (GOMBRe) (Fig. 1). The coastal zone is intertidal endowed with a rich diversity of marine organisms. Periwinkle, *L. undulata* belongs to the family Littorinidae and habitat in marine rocky intertidal regions. They attain a maximum size of 20 mm. Invariably, they occupy the highest tidal levels on rocky shores commonly clustering in rock crevices during day light, becoming active at night and crawling on the rocks. They have operculum that completely seals off the aperture of the shell helping to reduce water loss (Maruthamuthu et al., 1987). It grazes on encrusted algae (Alyakrinskaya, 2005). Collections of periwinkles are very much based on availability and seasonal abundance. Random sampling of periwinkles (n = 150) were handpicked from each stations, station 1 (Rasthacaud), station 2 (Perumanal, near reactor site to Kudankulam) and station 3 (Idinthakarai). All the samples were collected in three different seasons: pre-monsoon (July–September), monsoon (October–February) and post-monsoon (March–June) during July 2011–June 2012. In the laboratory, the specimens were maintained in seawater for 3 days for depuration. After mortality, the samples were washed thoroughly with tap water to remove the external debris, sand and silt. The morphometry of the animal was measured with a calliper scale and weighed. Specimens from each sampling stations were separated into different groups based on their shell length (mm). The whole body tissue is removed by dissection from all the specimens representing each group (due to smaller size, we could not able to separate the organs), weighed and dried in an oven at 80–90 °C for 48 h. The dried samples were homogenised and taken for analysis. The ^{210}Po activity of seawater (100 l) and sediments (3 kg) were also collected for estimating biological concentration factor (BCF) and radiation dose rate.

Polonium-210 concentration in seawater was determined by pre-concentration to $Fe(OH)_3$ (Jia et al., 2001; Khan and Wesley, 2011b). One hundred litres of seawater sample was filtered through a 0.45 μm filter paper and the filtrate is acidified with concentrated HCl to pH 1. Then 1 ml of ^{208}Po tracer and 25 mg of iron carrier was added to the sample. The sample was stirred using a mechanical stirrer for 30 min and allowed to stay for an hour. The pH was raised to 9 by adding concentrated NH_4OH. The sample was again stirred for an hour and kept overnight. The supernatant was decanted, and the precipitate collected after three washings. The solution was heated on a hot plate at 80–90 °C to incipient dryness and redissolved in 50 ml of 0.5 N HCl and filtered. ^{210}Po in the filtrate is allowed to deposit in a silver disc and alpha counted.

Ten grams of sediment sample together with 1 ml of 0.02 Bq ^{208}Po tracer, 30 ml of concentrated HNO_3 and 15 ml of 40% H_2O_2 were added to a 150 ml Erlenmeyer flask. The flask was constantly heated at 80–90 °C for 48 h. Before drying, 30 ml of concentrated HNO_3 and 10 ml of 40% H_2O_2 were added and the step was repeated until the emanation of brown fumes stopped the colour of the residue changed from brown to pale white. The solution is evaporated to incipient dryness and an additional 20 ml of H_2O_2 was added. The solution was evaporated to fuming to destroy the organic matter and the step repeated until a colourless residue was obtained. Three 10-ml portions of concentrated HCl were consecutively added to change the solution medium and evaporated to dryness. The residue was finally dissolved with 50 ml of 0.5 M HCl and filtered through a 0.1-μm Millipore filter paper and taken in a 50-ml beaker.

The dried homogenised mollusc sample together with 1 ml of 0.02 Bq ^{208}Po tracer, 20 ml of concentrated HNO_3 are added to an Erlenmeyer flask, which was then slowly heated on a hot-plate until the reaction starts. Due to foaming, the reaction rate is controlled by cooling. The flask was heated until the brown fumes disappeared on addition of another 20 ml of concentrated HNO_3. This step was repeated until the cloudy solution became clear. The solution was heated to incipient dryness, and then 15 ml of 40% H_2O_2 was added carefully. The flask were again heated and, if the solution became darker or vigorous reaction found it was allowed to react in a lower temperature. The solution was evaporated to dryness and another 15 ml of 40% H_2O_2 was added drop by drop to completely destroy the organic matter. Subsequent analyses were the same as those for sediment described above.

To the 50 ml filtered solution, 7 ml of 20% hydroxylamine hydrochloride and 7 ml of 25% sodium citrate solution and 0.2 g ascorbic acid was added. The pH of the solution was adjusted to 1.5–2 with 1:5 ammonia. The solution was stirred on a hot plate by using a magnetic stirrer. A Perspex holder with a silver disc was immersed into the solution. The ^{210}Po deposition is continued for 6 h at a temperature 85–90 °C, then disc is removed, washed with distilled water and acetone, dried and alpha counted. Second plating was also carried out immediately to completely remove the polonium and tracer and the results added. ^{210}Po was determined using an alpha probe (Nucleonix make, efficiency 35% using ^{241}Am standard, minimum detectable limit 0.5 Bq, counting time 6000 s). The calculation is done as per the formula:

$$A_{\text{Po-210}} = \frac{N\mathit{cps} \times 100 \times S \times M}{E \times A} \times \mathit{Cf} \times \mathit{Rf}$$

where $A_{\text{Po-210}}$, is the activity of ^{210}Po (Bq/kg wet), N*cps*, is the Net counts per second (cps), S is the volume conversion from g to kg, M is the moisture ratio for converting dry to wet weight, E is the efficiency of the counting system, A the sample size (taken for analysis in dry weight), Cf is the correction factor (time between the collection of sample and counting), and Rf is the recovery of ^{209}Po tracer.

The results were corrected for radiochemical recovery, and it ranged from 74% to 81%. The internal quality control was checked using certified reference material (CRM) procured from IAEA (IAEA

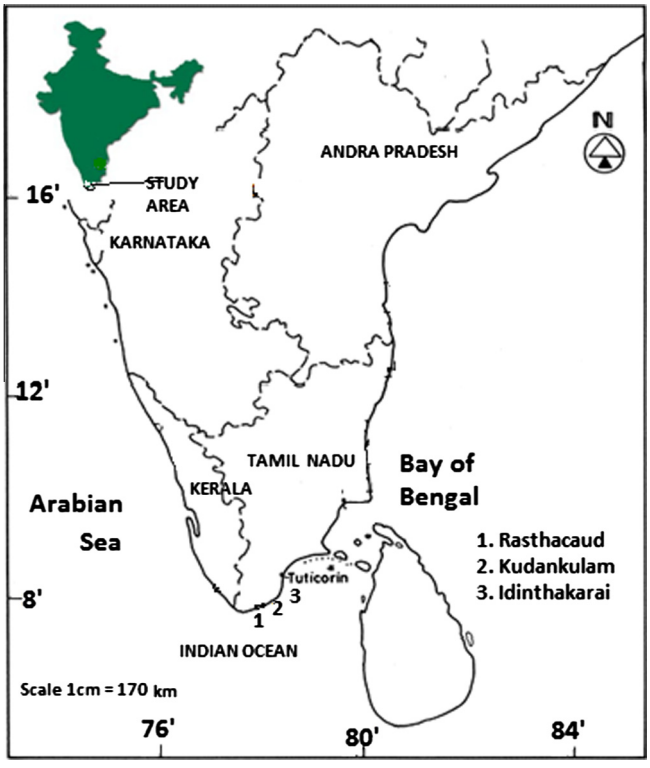

Fig. 1. Map showing the study area.