# Accumulation of $^{210}$Po and $^{210}$Pb in mussels: implications for environmental bio monitoring programmes

**Fernando P. Carvalho, João M. Oliveira**
Nuclear and Technological Institute (ITN)
Department of Radiological Protection and Nuclear Safety
E.N. 10, 2686-953 Sacavém, Portugal
E-mail: carvalho@itn.pt

## INTRODUCTION

Mussels are common marine molluscs found in most coastal areas and estuaries in a wide range of latitudes. They have been used as sentinel organisms for measuring contaminants and comparing concentrations of radionuclides, heavy metals and organic contaminants in marine environments (Gosling, 1992). Monitoring discharges of land based industries, including NORM (naturally occurring radioactive materials) industries into aquatic environments has been performed with the help of mussels also (Ryan *et al.*, 1999; Connan *et al.*, 2007).
The range of radionuclide concentrations, including those of the uranium series, in mussels vary widely round the year in the same station and amongst sampling stations. Fluctuation of concentrations might be due to a number of reasons, both external as well as internal, *i.e.*, related with the physiology and growth of mussels. However, correct interpretation of the results must be achieved in order to draw the relevant conclusions from environmental monitoring programs.
This paper reports monthly concentrations of $^{210}$Po and $^{210}$Pb in mussels and the relationship of these concentrations with a number of parameters in an attempt to better understand the source of variation of radionuclide concentrations round the year.

## MATERIALS AND METHODS

For a 17-month period mussel samples were monthly collected, during low tide, always at the same rocky pier of Costa da Caparica beach, South of Tagus River mouth. During the sampling period physical-chemical parameters of seawater, such as temperature, salinity and turbidity were measured with a portable multi parameter probe.

Water samples were collected into large polyethylene drums, transported to the laboratory and immediately filtered through 0.45μm pore size, 142 mm diameter membrane filters. Filtered water was acidified with $HNO_3$ to pH<2 and saved for radionuclide analysis. Filters with particulate matter were weighed, dried in the oven at 60ºC, and weighed again, before analysis of $^{210}$Po and $^{210}$Pb. Separate water samples were taken for determination of chlorophyll a and phaeopigments according to standard methods.

Mussels (*Mytilus galloprovincialis*) were collected from the rock surface in high number and specimens selected in the laboratory. For the purpose of the study on allometric variation of radionuclide concentrations, the shell length of mussels was measured with a Caliper rule and mussels separated into several size classes, 40 individuals in each (shell length classes 1.5-1.9, 2.5-2.9, 3.0-3.4, 3.5-3.9, 4.0-4.4, 4.5-4.9 cm). For the investigation of seasonal variation of radionuclide concentrations, monthly samples of the size class 3.5-3.9 cm were collected. Every month, 40 mussels with shell length in this interval were selected at random, soft

tissues dissected, pooled, and than dried, homogenized, and aliquots used for radionuclide analysis.

Po-210 radionuclide analysis was performed using about 0.5 g dry tissues, with addition of $^{209}$Po as internal isotopic tracer. Samples were fully dissolved with acids and polonium plated onto silver discs as described elsewhere. The sample solution was saved and stored during 6-12 months and, after a new addition of $^{209}$Po tracer, evaporation and treatment of dry residue with $HNO_3$, polonium was plated again and measured by alpha spectrometry. Pb-210 was calculated from the $^{210}$Po measurement in this second polonium determination. Measurements of polonium alpha particles emitted by the Ag discs were performed with low background 450 $mm^2$ ion implanted detectors from ORTEC EG&G connected to an OCTETE Plus alpha spectrometer. Quality assurance of analytical results was ensured by analysis of IAEA certified reference materials and participation in inter comparison exercises with good results.

## RESULTS AND DISCUSSION

*Allometric variations*

Total weight of mussels, as well as soft tissues dry weight, increased as a power function of the shell length linear variable. Po-210 concentration in mussels' soft tissues was not constant. Instead, it displayed a clear decrease with increasing shell length, or with increasing body weight. This result underlines the importance of keeping constant the size of mussels sampled in monitoring programs in order to allow comparisons of radionuclide measurements as well as temporal and spatial trend analyses.

As the mussels' size class 3.5-3.9 cm in shell length was selected for the long term investigation, in one monthly sample the 40 mussels were individually analysed in order to investigate inter individual variation. In this sample, $^{210}$Po average concentration was 970±330 Bq $kg^{-1}$ (dry weight). The distribution of results was log-normal and the geometric mean of $^{210}$Po concentration was 921(+280;-215) Bq $kg^{-1}$. Amazingly, in spite of the narrow range of shell lengths selected, within this 3.5-3.9 cm interval mussels' soft tissues still displayed a significant trend of decreasing radionuclide concentration with increasing shell length ($p<0.05$). In the same sample, $^{210}$Po concentration in individual mussel soft tissues significantly decreased also with increasing soft tissues weight (body size) ($p<0.001$), confirming that larger individuals have lower $^{210}$Po concentrations on a weight basis.

*Environmental parameters*

Water temperature measurements indicated a smooth seasonal fluctuation with minimum temperatures in January, 12.8ºC, and maximal in June, with 22.7ºC. Water temperature followed air temperature fluctuation closely with the exception of July and August months with cooler sea water temperatures due to the summer coastal upwelling. Salinity of water at the sampling station displayed fluctuations dependent upon rainfall and freshwater discharges by the Tagus River. Nevertheless, during the study period there have been no dramatic changes and average salinity measured in low tide was 32±2‰ (n=17) for the entire period.
Interestingly, chlorophyll-a and phaeopigment determined in the coastal seawater revealed higher values in February-March in both years. This was due to phytoplankton bloom and means increased amounts of food available to mussels. Over the 17-month period, concentrations of particulate matter in sea water fluctuated around a mean value of 14±5 g $m^{-3}$ (n=17) with 26% of combustible organic matter in average. Therefore, organic particles in seawater were available throughout the year but phytoplankton cells, the main food item to mussels, were generally less abundant in Summer and Autumn.

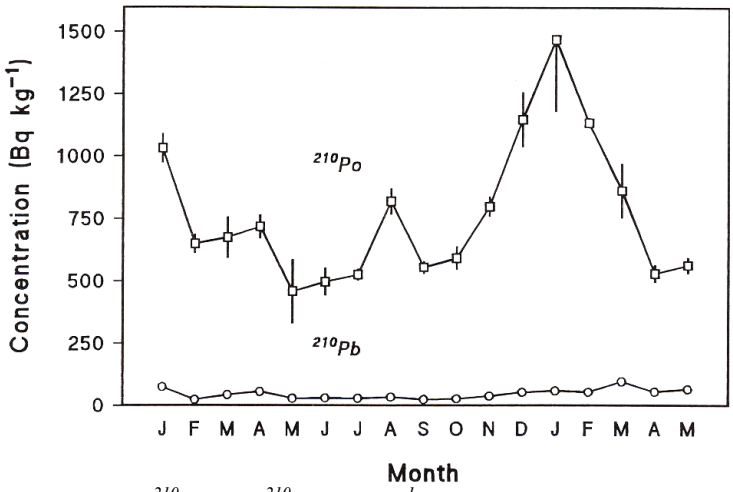


*Figure1. Monthly concentrations of $^{210}Po$ and $^{210}Pb$ (Bq $kg^{-1}$ dry weight) in mussels' soft tissues. Mussels' size: shell length comprised between 3.5 and 3.9 cm.*

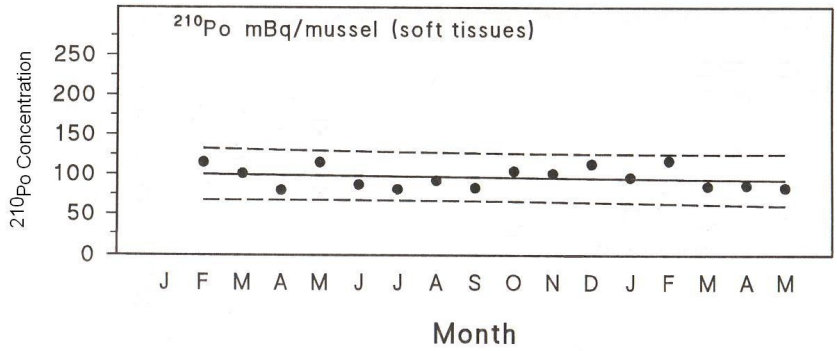


*Figure 2. Monthly $^{210}Po$ concentration per mussel during the study period. Lines represent the average and 95% confidence interval of the mean $^{210}Po$ concentration value.*

### *$^{210}Po$ and $^{210}Pb$ in seawater and suspended matter*

As mussels are filter feeders and ingest organic particles and plankton cells in suspension, the concentrations of these radionuclides were measured on monthly samples of suspended matter collected over the study period. The average concentration of dissolved radionuclides (n=17) was 0.45±0.14 Bq $m^{-3}$ and 0.54±0.28 Bq $m^{-3}$ for $^{210}Po$ and $^{210}Pb$, respectively. There was no seasonal fluctuation of radionuclide concentrations in water. Instead, they were fairly constant throughout the study, despite the seasonal fluctuation of $^{210}Po$ and $^{210}Pb$ concentrations in the atmosphere and in surface depositions at this area (Carvalho, 1995). In suspended matter, radionuclide concentrations were 140±40 Bq $kg^{-1}$ and 78±32 Bq $kg^{-1}$ dry weight (n=17) for $^{210}Po$ and $^{210}Pb$, respectively. Again, no clear fluctuation related to the season of the year was observed.

Clearly, radionuclides in solution and in suspended matter are the source of these radionuclides to mussels. The ingestion of $^{210}Po$ bound to particulate organic matter seems to be the most relevant transfer pathway (Carvalho and Fowler, 1994). However, as mussels avoid ingestion/digestion of sediment particles, and have preference for phytoplankton cells, these might be the main vector of radionuclide transfer to mussels.

*$^{210}$Po and $^{210}$Pb in mussels*

Radionuclide concentrations calculated on mass basis (Bq kg$^{-1}$ dry weight) from monthly measurements in mussels are displayed in Figure 1. $^{210}$Po concentrations fluctuated seasonally, with peaks of higher concentrations in Winter (January) and lower concentrations in late Spring-early Summer (May-June) and again in September-October. In mussels of this size class, concentration values ranged from 460 Bq kg$^{-1}$ to 1470 Bq kg$^{-1}$ (dry weight), averaging 767±283 Bq kg$^{-1}$ while $^{210}$Pb fluctuated from 23 to 96 Bq kg$^{-1}$ (dry weight) averaging 46±20 Bq kg$^{-1}$.

The type of fluctuations observed with $^{210}$Po concentrations are commonly interpreted as a seasonal effect. However, no seasonal fluctuation of $^{210}$Po and $^{210}$Pb in water and suspended matter was observed and, therefore, there has been no seasonality in the supply of these radionuclides to mussels.

Using the same data and the individual records of weight and length of mussels, the mussel Condition Index was computed for all samples (Gosling, 1992). Furthermore, $^{210}$Po was plotted as $^{210}$Po *per mussel* instead of $^{210}$Po per kg. This is shown in Figure 2. Values of $^{210}$Po content per mussel remained reasonably stable throughout the study period, while Condition Index values fluctuated. This plot suggests that concentration variations are due not to variation of mussels' $^{210}$Po body burden but to fluctuation of mussel's body weight. As $^{210}$Po does not bind to lipids, but to proteins, the seasonal fluctuation of lipid content in mussels might induce a fluctuation of the result expressed on body weight.

## CONCLUSIONS

Based on results reported herein, the main conclusions are:

1. In order to allow for comparing results over time and space, mollusc samples should be of a constant number of specimens and specimens always of the same size class.
2. It seems unlikely that seasonal signals, such as $^{210}$Po and $^{210}$Pb in atmospheric depositions, will reflect significantly in radionuclide concentrations displayed by marine mussels.
3. Mussels physiological condition may have a large effect on radionuclide content expressed on body weight and, if relevant for data interpretation, mussel's Condition Index should be determined.

Sampling programs must take into account the effect of age (body size) and physiological status (Condition Index) of molluscs on radionuclide concentrations measured.

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