

## $^{210}\text{Po}$ AND $^{210}\text{Pb}$ INTAKE BY THE PORTUGUESE POPULATION: THE CONTRIBUTION OF SEAFOOD IN THE DIETARY INTAKE OF $^{210}\text{Po}$ AND $^{210}\text{Pb}$

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**Abstract**—Through analysis of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the diet, the average ingestion rate for the Portuguese population is estimated at 1.2 and 0.47 Bq d<sup>-1</sup> per capita for  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ , respectively. Detailed analysis of foods indicate that seafood alone contributes up to 70% of the  $^{210}\text{Po}$  ingestion rate, whereas cereals, vegetables, and meat altogether contribute 79% of the  $^{210}\text{Pb}$  ingestion rate. Consumption of seafood, both in terms of quantities (kg d<sup>-1</sup> per person) and preferential consumption of certain marine species, is the cause of the relatively high intake of  $^{210}\text{Po}$  and high  $^{210}\text{Po}$ : $^{210}\text{Pb}$  ratio in the diet in comparison with other countries. Other  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  sources, namely inhalation of surface air and cigarette smoke, contribute only a small percentage of the absorption of these radionuclides in the blood. Estimated total body burdens of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in adult men, 70 Bq, are 3.5 times higher than estimates for humans living in normal radioactivity regions and consuming a reference diet. Average whole body effective doses for the adult from the Portuguese population are estimated at about 85  $\mu\text{Sv y}^{-1}$  from  $^{210}\text{Po}$  and 170  $\mu\text{Sv y}^{-1}$  from  $^{210}\text{Pb}$  absorbed with the diet. Effective dose from  $^{210}\text{Po}$  in the diet may vary from 25  $\mu\text{Sv y}^{-1}$  in a person consuming no seafood to 120  $\mu\text{Sv y}^{-1}$  in a heavy consumer of sardines, to 1,000  $\mu\text{Sv y}^{-1}$  in a hypothetical heavy consumer of molluscs. *Health Phys.* 69(4):469–480; 1995

**Key words:**  $^{210}\text{Po}$ ;  $^{210}\text{Pb}$ ; diet; effective dose

### INTRODUCTION

EVALUATION OF human population exposure to radioactivity has provided, and continues to provide, impetus for a large number of studies which are especially motivated by artificial modifications of the natural radiation environment and by potential biological effects of low level radiation (CEC 1990; UNSCEAR 1993).

A large contribution to the radiation dose received by humans comes from naturally-occurring uranium series radionuclides accumulated in the body, namely

$^{210}\text{Pb}$ ,  $^{210}\text{Bi}$ , and  $^{210}\text{Po}$  (UNSCEAR 1982, 1993). The internal radiation dose from these radionuclides follows the dose from  $^{40}\text{K}$ , whose concentration in tissues is homeostatically controlled. In contrast to  $^{40}\text{K}$ ,  $^{210}\text{Pb}$ , and  $^{210}\text{Po}$  in human tissues display variable concentrations. Food is the main source of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in the human body (Jaworowski 1969; Parfenov 1974; Holtzman 1978; UNSCEAR 1977, 1982). However, because the majority of studies on the intake of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  are based on analyses of diets with little or no inclusion of seafood, a common conclusion has been the existence of a  $^{210}\text{Po}$ : $^{210}\text{Pb}$  ratio lower than or near to unity in the human diet. Based on those studies, it is frequently assumed for dose assessment purposes that  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  are in radioactive equilibrium in the diet. Recently, following a review of concentration data, it was suggested that an average intake of 0.15 Bq d<sup>-1</sup> of  $^{210}\text{Po}$  and 0.09 Bq d<sup>-1</sup> of  $^{210}\text{Pb}$  would apply for the average individual in regions of normal radioactivity background (UNSCEAR 1993). Known exceptions to this intake rate are cases such as the Laplanders who consume reindeer and caribou meat with high  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  levels, and populations living in high radioactivity areas (UNSCEAR 1982, 1988, 1993; Thomas 1994). Moreover, as acknowledged by UNSCEAR (1982, 1993), populations consuming large quantities of seafood are expected to have a higher than average  $^{210}\text{Po}$  intake.

Portuguese consume relatively large quantities of seafood, 60 kg y<sup>-1</sup> per capita, which may be compared with 72 kg y<sup>-1</sup> in Japan, 21 kg y<sup>-1</sup> in the USA, and 20 kg y<sup>-1</sup> in the UK (gross quantities based on live weights, FAO 1993). Therefore, analyses of a variety of seafoods were performed in order to evaluate the ingestion of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  by the Portuguese population. Other food sources and inhalation sources were also evaluated in order to assess the total intake of these radionuclides. From this information, total body burdens of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  as well as whole body effective radiation doses are estimated for the adult human.

### MATERIALS AND METHODS

Samples of the main categories of food were purchased at markets in the Lisbon area. Only the edible uncooked parts were analyzed after appropriate cleaning

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and treatment for consumption. Therefore, results for fish samples refer to fish filet (muscle tissue), fruit samples refer to fruit pulp, leafy vegetables such as cabbage refer to washed cabbage leaves, and so on. Most of the marine organisms sampled were directly obtained from fishing vessels. In general, 1 to 4 samples were analyzed for each variety or species of terrestrial and animal produce, whereas for seafoods generally a higher number of replicate samples were analyzed. For a comprehensive presentation of data, the results were averaged over food groups, each one including a given number of biological species or varieties. Concentration of radionuclides are given in  $\text{Bq kg}^{-1}$  fresh weight as available for consumers. Uncertainty values given are  $\pm 1$  standard deviation (SD) of the arithmetic mean, unless stated otherwise. Dry:wet weight ratios, based on samples oven dried at  $80^\circ\text{C}$ , are given to facilitate comparison with published data.

Inhalation of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  was calculated through the use of concentrations measured in surface air at Sacavem, a small suburb of Lisbon. These measurements were made over a 4-year period encompassing all the seasons of the year on large volume air samples filtered through Whatman #42<sup>†</sup> paper filters (Carvalho 1995).

The three most popular cigarette brands produced in the country were analyzed for  $^{210}\text{Po}$ . Mainstream smoke, i.e., the smoke aspirated throughout the cigarette into the mouth, was sampled using a puffing device connected to a peristaltic pump. Smoke was aspirated via a glass tube with a cotton plug and drawn through a large volume wash bottle containing a 2 M HCl-ethanol solution which efficiently trapped the smoke. This solution, the plug, and two hot  $\text{HNO}_3$  washes to recover radioactivity adsorbed on the glassware were pooled for  $^{210}\text{Po}$  analysis. For each cigarette brand the experiment was repeated 5 times.

Polonium analyses were performed starting with a standard addition of a known activity of  $^{209}\text{Po}$ , as an internal isotopic tracer for radiochemical yield, followed by complete dissolution of the sample in mineral acids. Polonium isotopes were plated onto a silver disc in 0.5 M HCl solution in the presence of ascorbic acid using a technique modified from Flynn (1968). In order to ensure the complete removal of polonium isotopes, after plating the solution was cleaned of any polonium traces with a scrap of silver foil immersed for several hours. Several tests demonstrated that no co-deposition of  $^{210}\text{Pb}$  occurred on the silver disc under the conditions employed. Six to 12 mo later, following a new standard addition of  $^{209}\text{Po}$  tracer, a second polonium plating was made in order to determine  $^{210}\text{Pb}$  through the in growth of  $^{210}\text{Po}$ .

The measurement of polonium isotopes plated on silver discs was performed with silicon surface barrier detectors, 450  $\text{mm}^2$ , R type, 100  $\mu\text{m}$  depletion depth,

connected to a pulse height analyzer.<sup>‡</sup> Analytical blanks averaged  $0.2 \pm 0.1$  mBq and  $0.3 \pm 0.3$  mBq for  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ , respectively. Concentrations of radionuclides, always well above analytical blank values, were calculated through the Bateman equations for the sample collection date which was considered to be the consumption date. After propagation of the analytical and counting errors, the one-sigma relative uncertainty of  $^{210}\text{Po}$  concentration in each sample was between 5% and 10%. Analytical quality control of the entire procedure was regularly performed through participation in IAEA inter-comparison exercises using sample matrices such as sediments, fish, and cockles of unknown concentrations, with results remaining within  $\pm 1$  SD of the after exercise disclosed reference value.

The daily absorption rate ( $\text{Bq d}^{-1}$ ) from the diet into the blood,  $A_1$ , was calculated (after Magno et al. 1970; UNSCEAR 1977; Holtzman 1978) as

$$A_1 = I_1 \cdot f_1, \quad (1)$$

where

$I_1$  = the radionuclide ingestion rate through diet intake ( $\text{Bq d}^{-1}$ ) and  
 $f_1$  = the radionuclide gut transfer factor.

The daily absorption rate through inhalation into the blood,  $A_2$ , was calculated (Magno et al. 1970; UNSCEAR 1977) as follows:

$$A_2 = I_2 \cdot D_5 \cdot (f_2 + f_1 \cdot f_3), \quad (2)$$

where

$I_2$  = the radionuclide inhalation rate ( $\text{Bq d}^{-1}$ )  
 $D_5$  = the fraction of inhaled aerosol deposited in the lungs (0.50)  
 $f_1$  = the radionuclide gut transfer factor  
 $f_2$  = the fraction of aerosol activity deposited in the lungs absorbed through the lung wall (0.33); and  
 $f_3$  = the fraction of aerosol deposited in the lungs which was transferred into the digestive tract (0.67).

The total body burden,  $TBB$ , of each radionuclide, taking into consideration ingestion and inhalation, was calculated by

$$TBB = (A_1 + A_2) \cdot T_{ef} \cdot (\ln 2)^{-1} \quad (3)$$

with  $T_{ef}$  being the radionuclide effective half-life in the human body.

According to experimental evidence,  $^{210}\text{Po}$  absorbed into the blood is not significantly transferred to the bone (Torvik et al. 1974). In the human body,  $^{210}\text{Po}$  turns over rapidly in soft tissues, with a biological half-life of 50 d and an effective half-life of 37 d. Excretion of  $^{210}\text{Po}$  is well-described by a single exponential, and the whole organism may be regarded as a single compartment

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(Moroz and Parfenov 1972; Bernard 1979; Jaworowski 1969). The metabolism of  $^{210}\text{Pb}$  is much more complex, and its transfer from the blood to internal organs has been described through a multi-compartment model (Bernard 1977; ICRP 1979). Turnover of  $^{210}\text{Pb}$  in soft tissues is described by shorter biological half-lives than  $^{210}\text{Po}$  (Torvik et al. 1974). From results of  $^{210}\text{Pb}$  and stable lead analysis in human tissues, it is well established that lead is a bone-seeker, with 70% of lead in the organism accumulating in the bone and 30% in soft tissues. Approximately the same distribution has been found in humans from populations exposed to low as well as to high environmental levels of  $^{210}\text{Pb}$ . Elimination of  $^{210}\text{Pb}$  from the bone follows an exponential process with a long effective half-life (approximately 3,300 d) (Parfenov 1974; Holtzman 1978).

The selection of adequate radionuclide gut transfer factors for the model is a more difficult matter. Well accepted values applicable to absorption of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  with the diet are not available. According to published reports, gut absorption of  $^{210}\text{Po}$  ranges from 0.06 to 0.80 (Hill 1967; Moroz and Parfenov 1972; Ladinskaya et al. 1973; Hunt and Allington 1993). The lowest absorption values were determined with polonium inorganic compounds likely to be found at workplaces, but do not apply to the more absorbable organic polonium-complexes likely present in the food (Kendall et al. 1988; Phipps et al. 1991; Hunt and Allington 1993). We selected a cautious value of 0.35 based on Ladinskaya et al. (1973) and assumed that it applies to the intake of  $^{210}\text{Po}$  from diet by the adult. Published values for the absorption of  $^{210}\text{Pb}$  through the gut also spread over a relatively wide range, i.e., 0.04 to 0.14, and even to 0.65 under special test conditions (ICRP 1979). We chose 0.08 based on the review of data by Holtzman (1978), which is in close agreement with the  $^{210}\text{Pb}$  gut transfer factor of about 10% reported by Spencer et al. (1977).

Annual individual effective dose values were calculated by using dose per unit intake conversion factors recommended by the ICRP (1991):  $0.2 \mu\text{Sv Bq}^{-1}$  for  $^{210}\text{Po}$  and  $1 \mu\text{Sv Bq}^{-1}$  for  $^{210}\text{Pb}$ . It should be noted, however, that ICRP recommended factors are more suited for professional exposures and derived through the use of gut transfer factors different of those we selected above.

## RESULTS AND DISCUSSION

### Radionuclide concentrations in foods and drinking water

Radionuclide concentrations in agricultural products in food groups I and II were between  $0.02$  and  $0.78 \text{ Bq kg}^{-1}$  for  $^{210}\text{Po}$  and from  $0.03$  to  $0.71 \text{ Bq kg}^{-1}$  for  $^{210}\text{Pb}$  (Table 1). Horticultural products (group II) which include common cabbages, tomatoes, and fruits contained apparent lower concentrations than foods in group I, due to much higher water content. In all these foods, a relatively large spread in concentration values was found

even between samples of the same food type. The exceptional potential of some vegetables to contribute greatly to the dietary intake of  $^{210}\text{Pb}$  is particularly noteworthy. This is the case of watercress, which contained up to  $0.3 \pm 0.03 \text{ Bq kg}^{-1}$  of  $^{210}\text{Po}$  and  $9.7 \pm 0.8 \text{ Bq kg}^{-1}$  of  $^{210}\text{Pb}$  (wet wt), and of wild mushrooms with  $2.6 \pm 0.3 \text{ Bq kg}^{-1}$  of  $^{210}\text{Po}$  and  $4.1 \pm 0.3 \text{ Bq kg}^{-1}$  of  $^{210}\text{Pb}$  (wet wt). Wild mushrooms, however, were not included in Table 1 due to their minor contribution to the average diet.

In general, concentrations of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  measured in agricultural products (Table 1) fall in the overall range of values reported in the literature for other countries (Takata et al. 1968; Ladinskaya et al. 1973; Khandekar 1977; Holtzman 1978; Kametani et al. 1981; Smith-Briggs and Bradley 1984; Watson 1985; UNSCEAR 1993). Most of the samples analyzed in the course of this work originated in fields near Lisbon, a sedimentary plain with a normal radioactivity background of  $30 \text{ Bq kg}^{-1}$   $^{226}\text{Ra}$  (dry wt). However, some of the agricultural products supplied to markets in Lisbon may originate in the north and central portion of the country with soils 2–3 times higher in  $^{226}\text{Ra}$  concentrations. Although plants grown in higher radioactivity soils frequently display higher radionuclide concentrations (Vasconcellos et al. 1987), evidence has been presented suggesting that radionuclide accumulation in plants may depend more upon soil type (organic matter, pH, ion exchange capacity, etc.) than upon radioactivity in the soil (Simon and Ibrahim 1987). The use of phosphate fertilizers which can add radionuclides of the uranium series to the soil, eventually in chemical forms more available for absorption by plants, may also cause noticeable differences in radionuclide concentrations measured in the same plant species (Santos et al. 1990). Therefore, a relatively wide spread in  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  concentrations measured in samples of the same plant species seems unavoidable even within a small geographical area. Nevertheless, the large variation computed in some food groups, such as vegetables and fruits (Table 1), is largely due to the variation among varieties of food pooled in the group.

In most of the agricultural products analyzed, namely cereals and vegetables (Table 1, food groups I and II),  $^{210}\text{Po}:^{210}\text{Pb}$  concentration ratios were much lower than unity. In particular, leafy vegetables displayed  $^{210}\text{Po}:^{210}\text{Pb}$  ratios between 0.1 and 0.2, closely reflecting the  $^{210}\text{Po}:^{210}\text{Pb}$  ratio in atmospheric deposition (0.15) rather than the  $^{210}\text{Po}:^{210}\text{Pb}$  ratio in soils ( $\sim 1$ ) measured in the Lisbon region (Carvalho 1995).  $^{210}\text{Po}:^{210}\text{Pb}$  concentration ratios in non-leafy vegetables (roots, tomatoes) were also lower than unity ( $\sim 0.1$ ); however, the concentrations are below those measured in leafy vegetables (Table 1). This is in accord with results from experimental research on the uptake of  $^{210}\text{Pb}$  and stable lead by plants which demonstrated that foliar uptake is more intensive than uptake from soils via the roots (Chamberlain 1983). During radionuclide uptake from soil,  $^{210}\text{Po}$  seems to be excluded from absorption through



**Table 1.** Concentrations of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in foods and average radionuclide ingestion rate per capita in the Portuguese population. For each food type, mean  $\pm$  1 SD of measured concentration values is given; for marine produce, the weighted mean and range is shown. (see also Table 3);  $n$  = number of food varieties or species combined in the group. Consumption data are based on published national statistics.

Food groups	Consumption per capita $\text{kg d}^{-1}$	Dry:wet weight	$n$	Concentration ( $\text{Bq kg}^{-1}$ wet)		Ingestion rate ( $\text{Bq d}^{-1}$ )	
				$^{210}\text{Po}$	$^{210}\text{Pb}$	$^{210}\text{Po}$	$^{210}\text{Pb}$
I. Cereals							
wheat bread	0.199	0.71	1	$0.14 \pm 0.01$	$0.26 \pm 0.02$	0.028	0.052
rice	0.047	0.80	1	$0.23 \pm 0.02$	$0.27 \pm 0.10$	0.011	0.013
maize	0.100	0.40	1	$0.049 \pm 0.002$	$0.066 \pm 0.002$	0.005	0.007
Potato	0.278	0.26	2	$0.18 \pm 0.07$	$0.18 \pm 0.09$	0.05	0.05
Sugar	0.082	—	1	$0.78 \pm 0.03$	$0.71 \pm 0.03$	0.064	0.058
Grains <sup>a</sup>	0.015	0.08	3	$0.09 \pm 0.07$	$0.09 \pm 0.08$	0.001	0.001
II. Horticultural products	0.309						
leafy vegetables <sup>b</sup>	50%	0.07	4	$0.054 \pm 0.093$	$0.33 \pm 1.15$	0.008	0.051
non-leafy vegetables <sup>c</sup>	50%	0.06–0.46	3	$0.023 \pm 0.026$	$0.23 \pm 0.33$	0.003	0.035
Fruits <sup>d</sup>	0.176	0.06–0.45	8	$0.045 \pm 0.060$	$0.035 \pm 0.063$	0.008	0.006
III. Animal produce							
beef	0.031	0.28	2	$0.86 \pm 0.04$	$0.55 \pm 0.02$	0.027	0.017
pork	0.027	0.28	1	$0.67 \pm 0.05$	$0.43 \pm 0.02$	0.018	0.012
chicken	0.043	0.29	2	$0.15 \pm 0.10$	$0.12 \pm 0.05$	0.006	0.005
other	0.028	0.28	2	$0.29 \pm 0.10$	$0.29 \pm 0.10$	0.008	0.008
chicken eggs	0.014	0.25	2	$0.25 \pm 0.09$	$0.14 \pm 0.12$	0.004	0.002
cow milk	0.202	—	1	$0.28 \pm 0.03$	$0.24 \pm 0.01$	0.056	0.048
cheese	0.012	0.52	3	$0.90 \pm 0.04$	$0.77 \pm 0.10$	0.011	0.009
IV. Marine produce							
fish (fresh) <sup>e</sup>	0.100	0.23	51	6.0 (0.6–13.5)	0.3 (0.08–0.69)	0.60	0.03
dry cod	0.011	0.80	1	$1.8 \pm 0.1$	$0.50 \pm 0.02$	0.020	0.006
crustaceans <sup>f</sup>	0.001	0.25	7	20 (4–75)	1.2 (0.15–2.8)	0.002	0.001
bivalve molluscs <sup>g</sup>	0.003	0.15	6	75 (6–152)	3.6 (0.5–16)	0.22	0.01
cephalopods <sup>h</sup>	0.005	0.20	5	2 (1.1–45)	0.4 (0.3–1.6)	0.013	0.002
V. Beverages							
table wine and beer	0.345		2	$0.12 \pm 0.04$	$0.13 \pm 0.03$	0.041	0.045
tap water	0.500		1	$(0.21 \pm 0.02) 10^{-3}$	$(0.20 \pm 0.01) 10^{-3}$	0.0001	0.0001
Ingestion rate:	2.5					1.2	0.47

<sup>a</sup> Peas, beans, chick peas.

<sup>b</sup> Cabbage, lettuce, spinach, watercress.

<sup>c</sup> Tomato, carrot, turnip.

<sup>d</sup> Orange, apple, banana, strawberry, grapes, peach, melon, olives.

<sup>e</sup> Based on concentrations in muscle tissue, Table 3.

<sup>f</sup> Edible part of shrimp, prawns, crabs, Table 3.

<sup>g</sup> Soft tissues of clams, razor clams, cockle, oyster and mussels, Table 3.

<sup>h</sup> Soft tissues (mantle and arms) of squid, octopus, cuttlefish, Table 3.

roots, or at least is less taken up by plants than  $^{210}\text{Pb}$  (Simon and Ibrahim 1987). In addition, after uptake through the roots,  $^{210}\text{Po}$  is little translocated to the aerial organs of plants (Chamberlain 1983; Santos et al. 1990). Therefore,  $^{210}\text{Pb}$  measured in the pulp of fruits (Table 1) would likely correspond closer to the  $^{210}\text{Pb}$  actually taken up from soil than concentrations measured in leaves exposed to atmospheric deposition. The near-equilibrium  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  concentrations measured in fruits as well as in some cereals, grains, and potatoes, may result from radioactive decay of accumulated  $^{210}\text{Pb}$  during the life span of the plant and time lag between harvest and consumption.

In contrast to agricultural products,  $^{210}\text{Po}$  concentrations in meat and foods of animal origin such as eggs, milk, and cheese (Table 1, group III), were higher than those of  $^{210}\text{Pb}$ , with  $^{210}\text{Po}$ : $^{210}\text{Pb}$  ratios varying from 1.0 to 1.8. Although viscera (liver and kidneys from cattle) have not been analyzed, concentrations in these animal

products are likely to be higher than in beef and to display  $^{210}\text{Po}$ : $^{210}\text{Pb}$  ratios between 2 and 4 (Bunzl et al. 1979). On the other hand, the consumption of frozen meat result in a decrease in excess  $^{210}\text{Po}$  activity relative to  $^{210}\text{Pb}$  in the diet due to  $^{210}\text{Po}$  radioactive decay during the storage period. Therefore, stored animal products (frozen, canned, dried) may display  $^{210}\text{Po}$ : $^{210}\text{Pb}$  concentration ratios lower than those in the same foods analyzed fresh. Meat from wild game, although representing a non-negligible animal protein source, was not considered in the average diet due to lack of statistical information on its consumption. However, concentrations of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in this meat may be higher than those in domestic cattle. For example, a sample of meat from wild woodcock (dry:wet wt = 0.28) contained  $27 \pm 2 \text{ Bq kg}^{-1}$  of  $^{210}\text{Po}$  and  $0.96 \pm 0.02 \text{ Bq kg}^{-1}$  of  $^{210}\text{Pb}$  (wet wt). It may also be noted that radionuclide concentrations in cheese were about 3 times higher than those measured in milk (Table 1). However, this is not surprising taking



into account that 3–5 L of milk are normally used to produce 1 kg of cheese. In general, our results for animal products demonstrate  $^{210}\text{Po}$  concentrations higher than those of  $^{210}\text{Pb}$ , in contrast with previous studies which frequently assumed radioactive equilibrium.

Water and beverages (Table 1, group V) have the lowest  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  concentrations determined in all foods analyzed. The concentration given for water is for tap water at Lisbon (1/5 of the population of the country), and compares well with concentrations measured in filtered samples of surface waters in the region. This concentration can be applied to the average diet because most of the population (about 2/3) lives in towns fed with water from surface reservoirs. Nevertheless, exceptional situations do exist. This is the case of villages in the north and center of the country where water is supplied from mountain springs and aquifers in granitic rocks, which are known to bear high concentrations of dissolved radionuclides of the uranium series. In the water supply of a village in the mountain area, measured  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  concentrations were 100 times higher than in the Lisbon water. Furthermore, consumption of mineral water from spring sources may further elevate the ingestion rate of uranium decay product radionuclides (Bettencourt et al. 1988). However, even taking this into account, the radionuclide intake from water would still remain relatively low in comparison with other foods.

#### Radionuclide concentrations in seafood

Concentrations of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in seafoods (Table 1, group IV) justify a more detailed presentation. Table 2 displays results for edible flesh of selected marine species and allows the immediate perception of the variability of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  concentrations in different species. Concentrations of these radionuclides, especially those of  $^{210}\text{Po}$  in fish muscle, varied over a wide range both among species and among individuals of

the same species. For example, separate analyses of lateral muscle from 12 sardines (*Sardina pilchardus*, 57.5 g mean individual weight) from the same catch in August demonstrated  $^{210}\text{Po}$  values ranging from 3.5 to 16.1 Bq kg<sup>-1</sup> ( $6.8 \pm 3.7$  Bq kg<sup>-1</sup> wet wt). Moreover, in the same species the concentration of  $^{210}\text{Po}$  may display important seasonal fluctuation. For instance, the average concentration given above for sardines sampled in summer, is one tenth of the  $^{210}\text{Po}$  concentration measured in samples collected in winter (Table 2). Beyond this variation, small pelagic plankton feeding fish (e.g., sardines, sardinellas, and anchovies) tend to accumulate more  $^{210}\text{Po}$ , whereas large top predators such as tuna, blue-marlin, and sharks display lower concentrations. Also, bottom fish such as sole and hake accumulate less  $^{210}\text{Po}$  than pelagic plankton feeders (Table 2). In all samples of seafood analyzed, concentrations of  $^{210}\text{Po}$  were higher than those of  $^{210}\text{Pb}$  (Table 2). In fact, in fish muscle  $^{210}\text{Po}$ : $^{210}\text{Pb}$  concentration ratios were typically about 20 (range 3–66). An overall picture and interpretation of  $^{210}\text{Po}$  concentration levels in marine organisms based on food web relationships has been reviewed by Carvalho (1988). Recently, it has been experimentally demonstrated that  $^{210}\text{Po}$  in marine species is almost exclusively absorbed from food, and, at least in crustaceans, the digestive absorption of  $^{210}\text{Po}$  is higher than that of  $^{210}\text{Pb}$  causing the high  $^{210}\text{Po}$ : $^{210}\text{Pb}$  ratios usually found (Carvalho and Fowler 1993, 1994).

Taking account of differences in radionuclide concentrations in marine species while grouping them in a small number of biological groups relevant in the diet, the grouping criteria of FAO was followed (FAO 1993). A typical radionuclide concentration value per group was obtained as the arithmetic mean of concentrations for the species in a group, unless the catches of one or two species are dominant in the group. In this case the weighted mean concentration was calculated (Table 3).

Table 2. Concentrations of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  (Bq kg<sup>-1</sup> wet wt) in common seafoods. All samples analyzed are from the North-Eastern Atlantic, off Portugal.

Species collection date, body weight, no. individuals	Dry:wet weight	Muscle	
		$^{210}\text{Po}$	$^{210}\text{Pb}$
Sardine, <i>Sardina pilchardus</i> 30.01.87, w = $51 \pm 2.5$ g, n = 4	(0.230)	$66 \pm 2$	$1.00 \pm 0.02$
Anchovy, <i>Engraulis encrasicolus</i> 06.02.91, w = $5.5 \pm 1$ g, n = 6	(0.230)	$9.4 \pm 0.3$	$0.42 \pm 0.01$
Mackerel, <i>Scomber japonicus</i> 28.01.87, w = $120 \pm 26$ g, n = 4	(0.259)	$19 \pm 1$	$0.63 \pm 0.04$
Horse-mackerel, <i>Trachurus trachurus</i> 24.01.84, w = 200 g, n = 2	(0.221)	$5.2 \pm 0.2$	$0.10 \pm 0.02$
Bigeye tuna, <i>Thunnus obesus</i> 25.01.87, w = 31 kg, n = 1	(0.330)	$3.05 \pm 0.09$	$0.46 \pm 0.02$
Hake, <i>Merluccius merluccius</i> 24.01.84, w = 0.256 kg, n = 2	(0.200)	$6.7 \pm 0.3$	$0.15 \pm 0.01$
Red sea bream, <i>Pagellus bogaraveo</i> 24.01.84, w = 0.275 kg, n = 1	(0.270)	$2.41 \pm 0.09$	$0.84 \pm 0.02$
Common sole, <i>Solea vulgaris</i> 06.02.91, w = 0.150 kg, n = 1	(0.186)	$1.38 \pm 0.04$	$0.14 \pm 0.01$
Skate, <i>Raja undulata</i> 23.01.84, w = 0.894 kg, n = 1	(0.229)	$0.73 \pm 0.03$	$0.115 \pm 0.004$
Squid, <i>Loligo forbesi</i> 02.01.89, w = 2 kg, n = 1	(0.250)	$1.61 \pm 0.04$	$0.41 \pm 0.01$
Common shrimp, <i>Crangon crangon</i> n = 12	(0.295)	$49 \pm 1.5$	$1.11 \pm 0.03$
Clam, <i>Ruditapes decussatus</i> n = 6		soft tissues	
	(0.20)	$152 \pm 19$	$2.9 \pm 0.1$
Cockle, <i>Cerastoderma edule</i> n = 6		soft tissues	
	(0.11)	$94 \pm 3$	$1.32 \pm 0.06$
Mussel, <i>Mytilus galloprovincialis</i> n = 12		soft tissues	
	(0.14)	$132 \pm 5$	$2.6 \pm 0.1$



**Table 3.** Representative concentrations of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  by groups of seafood in the diet of Portuguese. Freshwater and diadromous fish were added to marine fish to compute ingestion rates. In some groups, species dominating the consumption are underlined.

FAO code	Group of species	% in the annual catch (1990)	No. of species analyzed	Typical concentration in muscle (Bq kg <sup>-1</sup> wet)			
				$^{210}\text{Po}$	(range)	$^{210}\text{Pb}$	(range)
1+2	Freshwater + diadromous fishes	0.8	6	1.0	(0.19–2.3)	0.12	(0.03–0.42)
	Marine fishes:						
31	Flounders, halibuts, <u>soles</u>	4.6	2	1.7	(0.7–2.8)	0.08	(0.03–0.14)
32	<u>Cods</u> , <u>hakes</u> , haddockes	10.1	7	3.4	(0.3–6.7)	0.30	(0.04–0.60)
33	Redfishes, basses, congers	11.9	2	0.8	(0.07–1.8)	0.19	(0.01–0.31)
34	Jacks, <u>breams</u> , groupers	7.5	6	2.1	(0.52–3)	0.69	(0.2–1.0)
35	<u>Sardines</u> , anchovy, horse mackerel	29.3	5	13.5	(3–66)	0.37	(0.1–1.4)
36	<u>Tunas</u> , bonitos, billfishes	4.3	5	4.9	(3–8)	0.54	(0.3–0.83)
37	<u>Mackerels</u> , snoeks, cutlassfishes	8.0	5	6.1	(1.7–19)	0.20	(0.03–0.63)
38	Sharks, rays	6.2	6	0.6	(0.12–1.7)	0.08	(0.02–0.13)
39	Miscellaneous marine fishes	9.1	7	1.8	(0.7–3.7)	0.17	(0.01–1.45)
	Mean in fresh fish, weighted by catches:			6.0		0.30	
4	Crustaceans	0.7	7	20	(4–75)	1.26	(0.15–2.8)
5	Molluscs: bivalves and gastropods	2.1	6	75	(6–152)	3.6	(0.5–16)
5	Molluscs: cephalopods	5.4	5	2	(1.1–45)	0.4	(0.3–1.6)

In the following step, a single representative value for "fresh fish" was computed through weighting each group by its percentage in the annual fish catch. In this way, the final value takes into account the proportions of main species in each fish group as well as the percentage of fish groups in the diet of the Portuguese. Molluscs and crustaceans also represent a large proportion of the dietary intake of  $^{210}\text{Po}$  (Table 1) due to the high concentrations usually measured in these organisms (Table 2). Furthermore, some fish products, such as liver and gonads, display  $^{210}\text{Po}$  concentrations much above those in fish filet (Carvalho 1988); however, these tissues are not generally consumed. Since the seafoods analyzed in this study are from the northeast Atlantic, most of the values with minor adjustments may also be applied to other European countries. Values for the FAO group 35 (sardines, anchovies, and herring) would need adjustment in order to take into account that northern European countries consume herring but not sardine, thus lowering the average  $^{210}\text{Po}$  concentration of the group to about 1–2 Bq kg<sup>-1</sup>. On the other hand, Mediterranean countries do not consume herring but instead eat anchovies and sardines and nearly the same average concentration value in Table 3 would be obtained.

#### Intake of radionuclides with the diet

The per capita ingestion rate for each radionuclide (Bq d<sup>-1</sup>) was calculated using the national average consumption of foods. Average ingestion rates are 1.2 Bq d<sup>-1</sup> and 0.47 Bq d<sup>-1</sup> for  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ , respectively, with a corresponding  $^{210}\text{Po}$ : $^{210}\text{Pb}$  ratio in the diet of 2.6 (Table 1). Relative contributions of food groups to the Portuguese diet are shown in Fig. 1. It is noteworthy that the largest contribution (70%) to the ingestion of  $^{210}\text{Po}$  comes from seafood in spite of the small percentage it represents in the diet (5%). The largest contribution of  $^{210}\text{Pb}$  arises from cereals, vegetables, and meat (79%),

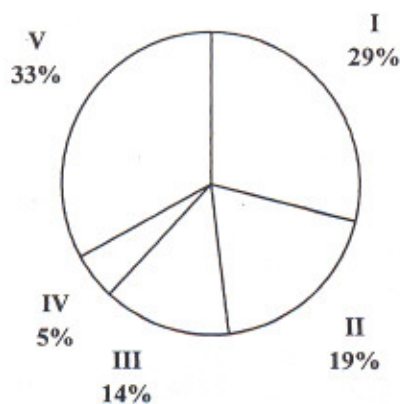
whereas seafood contributes only to 10% of the total  $^{210}\text{Pb}$  ingestion.

It is clear that the high ingestion rate of  $^{210}\text{Po}$  by Portuguese as well as the elevated  $^{210}\text{Po}$ : $^{210}\text{Pb}$  ratio in the diet are due to the high consumption of seafood, and in particular to the types of marine species more heavily consumed. Moreover, the data for seafood and also fresh animal products (Table 1) indicate that the common assumption of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  equilibrium in the diet does not hold for the Portuguese population. These data contrast with results of the dietary intake of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in other countries when diet is a continental type (Watson 1985; Smith-Briggs et al. 1986).

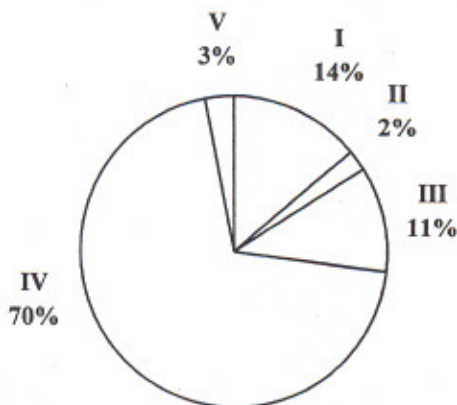
Considering only the consumption of seafood, the average ingestion rates of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  are 0.82 Bq d<sup>-1</sup> and 0.047 Bq d<sup>-1</sup>, respectively. These rates compare well with the per capita ingestion rates from seafood recently calculated for Japan, i.e., 0.48–0.69 Bq d<sup>-1</sup> for  $^{210}\text{Po}$  and 0.022–0.042 Bq d<sup>-1</sup> for  $^{210}\text{Pb}$  (Yamamoto et al. 1994), although the composition of the seafood diets is different.

The ingestion rate of  $^{210}\text{Pb}$  by the Portuguese population, 0.47 Bq d<sup>-1</sup>, is higher than  $^{210}\text{Pb}$  ingestion rates (0.05 to 0.22 Bq d<sup>-1</sup>) reported for several West-European and North-American countries (Watson 1985; Smith-Briggs et al. 1986) and is nearer to  $^{210}\text{Pb}$  dietary intake rates reported for Japan (0.63 Bq d<sup>-1</sup>, Takata et al. 1968) and the former URSS, Rostov on Don (0.23 Bq d<sup>-1</sup>, Ladinskaya et al. 1973). The differences in  $^{210}\text{Pb}$  ingestion rates are due, at least partially, to the composition of diets. For example, leafy vegetables which give a significant contribution to  $^{210}\text{Pb}$  in the diet of the Portuguese are not included in reports on the diet of the British (Smith-Briggs and Bradley 1984; Smith-Briggs et al. 1986). Nevertheless, some foods of terrestrial origin, which contribute most of the  $^{210}\text{Pb}$  intake rate by the Portuguese, still contain relatively high  $^{210}\text{Pb}$  concentra-

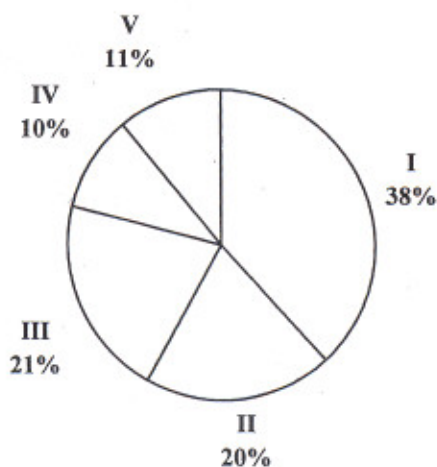




### CONSUMPTION OF FOODS



### INGESTION OF $^{210}\text{Po}$



### INGESTION OF $^{210}\text{Pb}$

Fig. 1. Percent contribution of foods to diet and to  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  ingestion rates by the Portuguese population. Food groups: I cereals, potatoes and grains, II vegetables and fruits, III meat and animal products, IV seafoods, V water and beverages.

tions in comparison with comparable foods for other diets (Watson 1985). Taking into account that the food samples we analyzed are from a region of normal radioactivity soils with low atmospheric  $^{210}\text{Pb}$  deposition, it is therefore unlikely that measured  $^{210}\text{Pb}$  concentrations in terrestrial foods are simply related with  $^{210}\text{Pb}$  concentrations in soils or with  $^{210}\text{Pb}$  atmospheric flux. Most likely, without additional knowledge of soil type and the use of fertilizers, single-parameter based models, such as universal concentration values or universal soil-to-plant transfer coefficients, would not be able to accurately estimate  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  concentrations in plants and in the human diet. Similar difficulties likely apply to concentrations of these radionuclides in meat because of different livestock feeds which may be the cause, and would help explain, the wide variation in published concentration data (UNSCEAR 1993). Moreover, it is likely that food processing and cooking may modify radionuclide concentration in foods, eventually decreasing them in comparison with concentrations measured in products analyzed fresh as we did.

### Radionuclide concentrations in inhaled air

Mean concentrations of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in surface air measured in the surroundings of Lisbon are  $31 \times 10^{-6}$  and  $181 \times 10^{-6} \text{ Bq m}^{-3}$  for  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ , respectively (Carvalho 1995). Thus, inhalation of  $20 \text{ m}^3$  of air per day (ICRP 1979) would lead to inhalation rates of  $6.2 \times 10^{-4} \text{ Bq d}^{-1}$  and  $3.6 \times 10^{-3} \text{ Bq d}^{-1}$  for  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ . These values are lower than estimated average inhalation rates for human populations in the north hemisphere (UNSCEAR 1993). They are, however, fully justified by the origins of radon and radon progeny in the atmosphere of the region (Carvalho 1995).

### Radionuclide concentrations in cigarette smoke

For a significant portion of the population, cigarette smoke is an additional source of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  intake through inhalation. Concentrations of  $^{210}\text{Po}$  measured in tobacco produced in Portugal, ranging from 2.8 to  $37 \text{ mBq g}^{-1}$ , vary with the cigarette type and are likely due to the different varieties of tobacco and manufacturing procedures used (Table 4). Due to the time lag between the harvest of tobacco leaves and manufacture of cigarette (a few months to 2 years)  $^{210}\text{Po}$  in cigarettes may approach secular equilibrium with  $^{210}\text{Pb}$ .  $^{210}\text{Po}$  concentrations are, however, unexceptional and reported values range from  $3 \text{ mBq g}^{-1}$  in India to  $36 \text{ mBq g}^{-1}$  in the U.S. (Holtzman 1978; Watson 1985). Volatilization of  $^{210}\text{Po}$  is evident in the low  $^{210}\text{Po}$  activity measured in cigarette ash and tips (butts) compared with the total  $^{210}\text{Po}$  present in the unburned cigarette (Table 4). In cigarettes with filters, 7–12% of the initial  $^{210}\text{Po}$  in the tobacco was retained by the filter, whereas in cigarettes without a filter, 40% of the initial  $^{210}\text{Po}$  was found in the butt plus ash (the butt here was an ~1.5 cm cigarette tip containing unburned tobacco). For the three brands analyzed, 50% or more of the initial  $^{210}\text{Po}$  in the cigarette was not carried with the mainstream of smoke (inhaled), but was



Table 4. Concentration of  $^{210}\text{Po}$  in cigarettes and cigarette smoke.

Cigarette type	Tobacco per cigarette (dry g)	$^{210}\text{Po}$ mBq (g tobacco) $^{-1}$	$^{210}\text{Po}$ (mBq per cigarette)				Daily inhalation of $^{210}\text{Po}$ mBq per 20 cigarettes	
			Total	Inhaled smoke (mainstream)		Residue (tip + ash)		
				mBq	(% Total)	mBq		(% Total)
Blended, with filter	0.748	37	28.9	1.52	(5)	2.1	(7)	30
Blended, without filter	0.793	15	12.2	1.37	(11)	4.9	(40)	27
Light, with filter	0.940	2.8	2.6	0.97	(37)	0.3	(12)	19

dispersed in the atmosphere. Therefore, a person that smokes one pack per day (20 cigarettes) of a blended type cigarettes may inhale about  $0.03 \text{ Bq d}^{-1}$  of  $^{210}\text{Po}$ . This activity is 48 times higher than the daily inhalation of atmospheric  $^{210}\text{Po}$  by a non-smoker. These results confirm previous reports on the inhalation of  $^{210}\text{Po}$  from cigarette smoke which is assumed to contain  $^{210}\text{Pb}$  at about half the concentration of  $^{210}\text{Po}$  (Holtzman 1978; Mussalo-Rauhamaa and Jakkola 1985; Watson 1985).

### Absorption into the blood

Based on the radionuclide ingestion rates determined above, the digestive absorption from the diet into the blood (eqn 1) is  $0.42 \text{ Bq d}^{-1}$  for  $^{210}\text{Po}$  and  $0.038 \text{ Bq d}^{-1}$  for  $^{210}\text{Pb}$ . For the population in the Lisbon area, absorption rates from surface air into the blood (eqn 2) are  $1.7 \times 10^{-4} \text{ Bq d}^{-1}$  for  $^{210}\text{Po}$  and  $6.9 \times 10^{-4} \text{ Bq d}^{-1}$  for  $^{210}\text{Pb}$ . Persons smoking one pack per day absorb into the blood  $8 \times 10^{-3} \text{ Bq d}^{-1}$  of  $^{210}\text{Po}$  from inhalation of cigarette smoke, i.e., about 50 times more than non-smokers. Absorption of  $^{210}\text{Pb}$  from cigarette smoke is likely to be much lower than absorption of  $^{210}\text{Po}$ .

Cigarette smoke was reported to significantly increase lung exposure to  $^{210}\text{Po}$  and confirmed by  $^{210}\text{Po}$  measurements in lung tissues (Jaworowski 1969; UNSCEAR 1982). Nevertheless,  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  absorbed into the blood through inhalation of surface air and cigarette smoke contribute only little to the total absorption of these radionuclides in internal organs (Fig. 2). Evidence of this small contribution through lung absorption was provided by concentrations measured in internal tissues of smokers that do not markedly differ from those of non-smokers (Hill 1965; Blanchard 1967; Holtzman 1978; Gilbert et al. 1988).

Therefore, the diet and absorption through the digestive tract are the main source and principal pathway of intake for  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  by the Portuguese population (Fig. 2).

### Radionuclide body burdens and radiation dose

Using the intake rates determined above and selected metabolic parameters, the total body burdens of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  may be estimated through eqns (1–3), for the non-smoker adult in the Portuguese population.

Assuming a constant  $^{210}\text{Po}$  intake through ingestion of food,  $1.2 \text{ Bq d}^{-1}$ ,  $^{210}\text{Po}$  in the body may reach equilibrium in about 120 d at a cumulative deposit of 22 Bq.  $^{210}\text{Po}$  absorbed from ingested food turns over in the

body with an effective half-life of 37 d (Jaworowski 1969; Moroz and Parfenov 1972; Bernard 1979). However, in internal organs there is also *in vivo* production of  $^{210}\text{Po}$  through radioactive decay of  $^{210}\text{Pb}$ .

The whole body burden of  $^{210}\text{Pb}$  at equilibrium is estimated to be 70 Bq, with 50 Bq accumulated in the bone and 20 Bq in soft tissues. The absorbed  $^{210}\text{Pb}$  is preferentially accumulated in bone (70%) and turns over in this tissue with a long effective biological half-life of 3,300 d, due to immobilization in the mineral matrix (Jaworowski 1969; Bernard 1977; UNSCEAR 1977; Holtzman 1978). The majority of experimental evidence indicates that  $^{210}\text{Po}$  in the bone arises through radioactive decay of  $^{210}\text{Pb}$  and mostly remains immobilized in bone structure. This process leads to  $^{210}\text{Po} : ^{210}\text{Pb}$  ratios close to 1, which have frequently been measured in samples of human bone (Blanchard 1967; Jaworowski 1969; Takizawa et al. 1990). It is also believed that part of this  $^{210}\text{Po}$  may be transferred to other tissues and excreted. We assume that a fraction of 0.2 of  $^{210}\text{Po}$  formed in the bone may be transferred to soft tissues (Parfenov 1974). The exchangeable  $^{210}\text{Po}$  that arises from decay of the  $^{210}\text{Pb}$  accumulated in soft tissues and  $^{210}\text{Po}$  transferred from bone, are also eliminated from the body with a  $T_{ef} = 37 \text{ d}$ . At equilibrium, the activity of exchangeable polonium ( $A_{Po}$ ) in the body ( $^{210}\text{Po}$  directly absorbed through the gut and lungs plus  $^{210}\text{Po}$  from the decay of  $^{210}\text{Pb}$  in soft tissues and  $^{210}\text{Po}$  translocated from bone) may be calculated through the balance equation

$$I_1 \cdot f_1 + \lambda_{Po} \cdot A_{Pb(st)} + 0.2 \lambda_{Po} \cdot A_{Pb(sk)} - (\lambda_{Pb(st)} + \lambda_{Po}) \cdot A_{Po} = 0, \quad (4)$$

where  $A_{Pb}$  is the inventory of  $^{210}\text{Pb}$  in soft tissues (*st*) and skeleton (*sk*),  $\lambda_{Po}$  is the radioactive decay rate constant of  $^{210}\text{Po}$  ( $0.00501 \text{ d}^{-1}$ ),  $\lambda_{Po(b)}$  is the biological elimination rate constant of  $^{210}\text{Po}$  ( $0.0138 \text{ d}^{-1}$ ), and  $I_1$  and  $f_1$  are as given in eqn (1). Calculation gives  $A_{Po} = 30 \text{ Bq}$ . This inventory of exchangeable  $^{210}\text{Po}$  in the body is comprised of 22 Bq (70%) from  $^{210}\text{Po}$  intake with the diet, 5 Bq (20%) from the decay of  $^{210}\text{Pb}$  in soft tissues, and 3 Bq (10%) translocated from the bone. The total body burden of  $^{210}\text{Po}$  is, therefore, estimated at about 70 Bq, with 30 Bq of exchangeable  $^{210}\text{Po}$  and 40 Bq trapped in mineralized bone produced *in situ* through  $^{210}\text{Pb}$  decay.  $^{210}\text{Po}$  immobilized in the bone turns over with a long



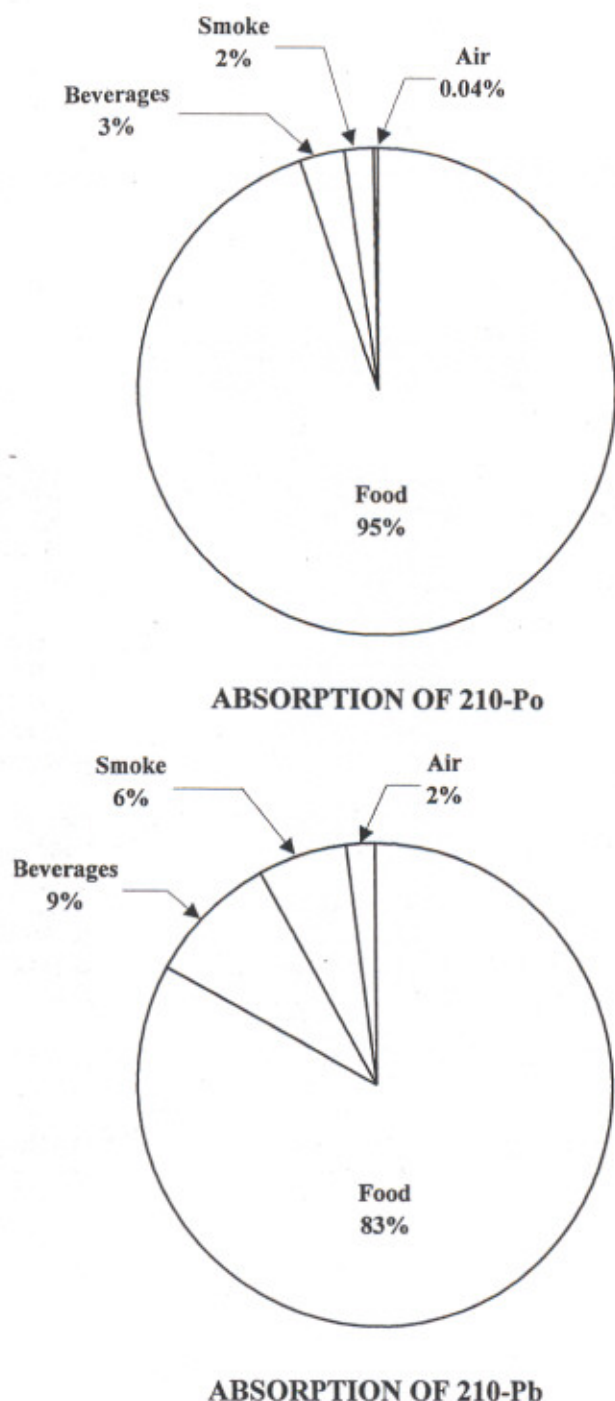


Fig. 2. Percent contribution of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  sources to the activity absorbed in the blood through ingestion and inhalation. It was assumed that  $^{210}\text{Pb}$  concentration in cigarette smoke (20 cigarettes per day) is half of the  $^{210}\text{Po}$  concentration.

biological half-life comparable to that of  $^{210}\text{Pb}$ , and both depend on the remodeling rate of bone (Jaworowski 1969). These results imply that  $^{210}\text{Po}$  due to absorption from the diet, 22 Bq, although contributing to the majority of the exchangeable  $^{210}\text{Po}$  in the body (30 Bq), accounts only for 30% of the  $^{210}\text{Po}$  TBB. The remaining

70% originates from the decay of  $^{210}\text{Pb}$  in the body. The  $^{210}\text{Po}$ : $^{210}\text{Pb}$  ratio in the whole body is, therefore, expected to be around unity, whereas the  $^{210}\text{Po}$ : $^{210}\text{Pb}$  ratio in the exchangeable fraction is approximately 1.5.

These estimates of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  total body burdens for the adult from the Portuguese population, 70 Bq, may be compared with the TBBs computed for the average Reference Man using the same metabolic parameters and model calculations as above. Assuming intake rates of  $0.15 \text{ Bq d}^{-1}$   $^{210}\text{Po}$  and  $0.08 \text{ Bq d}^{-1}$   $^{210}\text{Pb}$  in the reference diet (UNSCEAR 1993), the radionuclide body burdens in this average person would be 19 Bq  $^{210}\text{Po}$  and 24 Bq  $^{210}\text{Pb}$ . These computed values are in accord with TBBs of about 18 - 20 Bq  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  measured in human tissues in a population with low intake ( $0.05 \text{ Bq d}^{-1}$ ) of each radionuclide (Blanchard and Moore 1971; Holtzman 1978). In a similar way, an adult from the Arctic regions consuming reindeer and caribou meat at a median ingestion rate of  $7 \text{ Bq d}^{-1}$   $^{210}\text{Po}$  and  $1 \text{ Bq d}^{-1}$   $^{210}\text{Pb}$  (UNSCEAR 1977), would attain 235 Bq  $^{210}\text{Po}$  and 155 Bq  $^{210}\text{Pb}$  body burdens. Blanchard and Moore (1970) reported 130 Bq for the  $^{210}\text{Pb}$  TBB in a population of Alaskan Eskimos, based on direct measurements of radionuclides in tissue samples. The reasonable agreement of model calculations above with reported measurements suggests that the gut transfer factors we selected may be reasonable. Moreover, body burdens of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the average adult from the Portuguese population would be 3-3.5 times higher than those in the reference adult. Nevertheless, they remain far below values for Arctic populations and people living in areas of high natural radioactivity. The higher than average  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  ingestion rates by the Portuguese are the reason for the total body burden of these radionuclides. It may be noted, however, that although the ingestion rate of  $^{210}\text{Po}$  is about 8 times higher than the reference intake suggested by UNSCEAR (1993), the  $^{210}\text{Po}$  body burden is not higher in the same proportion. This is due to the short effective half-life of  $^{210}\text{Po}$  from the diet in the body and to the relatively large contribution of  $^{210}\text{Po}$  from  $^{210}\text{Pb}$  immobilized in the bone.

Based on the average radionuclide intakes (Table 1), the whole body effective dose computed (ICRP 1991) for the adult from the Portuguese population is of about  $85 \mu\text{Sv y}^{-1}$  from  $^{210}\text{Po}$  and  $170 \mu\text{Sv y}^{-1}$  from  $^{210}\text{Pb}$  in the diet (including the contributions by  $^{210}\text{Bi}$  and  $^{210}\text{Po}$  formed in the body). These values may be compared with the 11 and  $32 \mu\text{Sv y}^{-1}$  respectively for  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the average person (UNSCEAR 1993) and with 510 and  $365 \mu\text{Sv y}^{-1}$  computed as above for the Arctic populations. Recent dose calculations for inhabitants of Alaska have concluded that effective doses due to  $^{210}\text{Po}$  from the diet are  $444 \mu\text{Sv y}^{-1}$  in Alaskans eating caribou meat and of  $64 \mu\text{Sv y}^{-1}$  in British residents that did not (Thomas 1994).

Studies on populations with high consumption of seafood are very scarce in the literature. It is therefore



interesting to further evaluate the implication to the dose from  $^{210}\text{Po}$  in the diet in relationship with the variation in individual dietary habits.

A person not consuming seafood ingests  $0.34 \text{ Bq d}^{-1} \text{ }^{210}\text{Po}$  with the diet (Table 1), therefore nearly the same activity as for  $^{210}\text{Pb}$ . The total body burden of  $^{210}\text{Po}$  in such a person would be  $66 \text{ Bq}$ , slightly below the  $^{210}\text{Pb}$  TBB. The whole body effective dose due to  $^{210}\text{Po}$  from the diet is thus estimated to be  $25 \mu\text{Sv y}^{-1}$ . A hypothetical heavy consumer of sardines may find fresh sardines on the market during 4–5 months per year (late spring and summer) and consume  $0.24 \text{ kg d}^{-1}$  during that period. Using the concentration of  $^{210}\text{Po}$  measured in sardines sampled in summer,  $6.8 \text{ Bq kg}^{-1}$  (wet wt), the adult total body burden of  $^{210}\text{Po}$  would attain  $135 \text{ Bq}$  at the end of the summer period. The effective dose to an individual from  $^{210}\text{Po}$  in this diet would be of  $120 \mu\text{Sv y}^{-1}$ , i.e., about 5 times higher than in a person consuming no seafood. Another hypothetical, but more unlikely, heavy consumer of  $0.2 \text{ kg d}^{-1}$  of molluscs (e.g., clams and mussels) would ingest  $15 \text{ Bq d}^{-1} \text{ }^{210}\text{Po}$  and his  $^{210}\text{Po}$  TBB would be  $320 \text{ Bq}$ . The corresponding whole body effective dose is computed to be about  $1,000 \mu\text{Sv y}^{-1}$ . In the three cases considered above, the TBB and dose to humans from  $^{210}\text{Pb}$  in the diet would practically remain unchanged because seafood gives little contribution to the intake of this radionuclide. Moreover, the increased TBB of  $^{210}\text{Po}$  in heavy consumers of seafoods would decrease back to the average level computed in the Portuguese population a short time after ceasing the extraordinary ingestion of seafood. On the other hand, any increased ingestion of  $^{210}\text{Pb}$  most likely would occur in relationship with agricultural products and give rise to increased and long lived body burdens of  $^{210}\text{Pb}$  due to the accumulation of this radionuclide in the bone.

## CONCLUSIONS

Results from measurements of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  in the diet of the Portuguese population highlight the contribution of seafood (70%) in the dietary intake of  $^{210}\text{Po}$ . However, seafood does not contribute significantly to the ingestion of  $^{210}\text{Pb}$ . Instead, cereals, vegetables, and meat are the main sources of ingested  $^{210}\text{Pb}$  (79%). In contrast with results from dietary studies in other countries,  $^{210}\text{Po}:$  $^{210}\text{Pb}$  in the Portuguese diet is 2.6, clearly above unity.

As expected, inhalation of atmospheric  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  results in a minor contribution to the total intake of these radionuclides. Inhalation of cigarette smoke definitely increases the exposure of lungs to  $^{210}\text{Po}$  by a factor of about 50 when compared with atmospheric  $^{210}\text{Po}$ . However, the contribution of cigarette smoke and atmospheric inhalation to the total  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  absorbed into internal tissues is small (likely less than 5%) in comparison with the intake of these radionuclides from the diet.

The average total body burdens of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  calculated for an adult in the Portuguese population,  $70 \text{ Bq}$  for  $^{210}\text{Po}$  and  $70 \text{ Bq}$  for  $^{210}\text{Pb}$ , are about 3.5 times higher than estimates for an adult in regions of normal radioactivity and assumed to ingest the reference diet proposed by UNSCEAR (1993). The higher than average  $^{210}\text{Pb}$  body burden is due to the contribution of agricultural and animal products to the diet and does not result from the consumption of seafood. It is noteworthy that the high  $^{210}\text{Po}$  intake rate with seafood does not contribute to the build up of a total  $^{210}\text{Po}$  body burden in humans above that of  $^{210}\text{Pb}$ , due to the short effective half-life of  $^{210}\text{Po}$  from the diet in the body. Nevertheless,  $^{210}\text{Po}$  ingested in the diet and thus especially with seafood, accounts for a radiation dose higher than in humans consuming no seafood. Average consumers of seafood from the Portuguese population may receive an effective dose of about  $85 \mu\text{Sv y}^{-1}$ , i.e., 3.5 times higher than a person consuming no seafood. Nevertheless, they are still far below intake rates and radiation doses reported for populations in Arctic regions (UNSCEAR 1993; Thomas 1994). Hypothetical heavy consumers of seafood, especially molluscs, may be exposed to radiation doses from  $^{210}\text{Po}$  much above the average  $85 \mu\text{Sv y}^{-1}$ . However, this exposure would closely follow changes in the diet intake because  $^{210}\text{Po}$  is rapidly eliminated from the body.

Estimates of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  body burdens and effective doses in the average adult from the Portuguese population are a direct result of the higher than average dietary intake rates of those radionuclides. However, it should be pointed out that analyses of foods were performed on fresh produces (market-basket survey) and, therefore, that the possible effect of cooking on the concentration of radionuclides is not taken into account. At present, no data on such effect are available to enable the discussion on implications to dietary intake rates of  $^{210}\text{Po}$  and  $^{210}\text{Pb}$ . Computation of radionuclide body burdens and dose to humans depend very much upon the gut transfer factors used. It is important to note that most of the experimentally determined gut transfer factors in use for  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  were obtained under conditions (chemical compounds used, administration route, etc.) that may not apply to the absorption of radionuclides from the diet (Kendall et al. 1988; Hunt and Allington 1993). Hence, further research applied to the actual dietary intake of these radionuclides would be highly desirable.

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## REFERENCES

- Bernard, S. R. Dosimetric data and metabolic model for lead. *Health Phys.* 32:44–46; 1977.
- Bernard, S. R. A metabolic model for polonium. *Health Phys.* 36:731–732; 1979.



- Bettencourt, A. O.; Teixeira, M. M. R.; Faisca, M. C.; Vieira, I. A.; Ferrador, G. C. Natural radioactivity in Portuguese mineral waters. *Radiat. Protect. Dosim.* 24:139–142; 1988.
- Blanchard, R. L. Concentrations of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in human soft tissues. *Health Phys.* 13:625–632; 1967.
- Blanchard, R. L.; Moore, J. B.  $^{210}\text{Po}$ ;  $^{210}\text{Pb}$  in tissues of some Alaskan residents as related to consumption of caribou or reindeer meat. *Health Phys.* 18:127–134; 1970.
- Bunzl, K.; Kracke, W.; Kreuzer, W.  $^{210}\text{Pb}$ ;  $^{210}\text{Po}$  in liver and kidneys of cattle—I. Animals from an area with little traffic or industry. *Health Phys.* 37:323–330; 1979.
- Carvalho, F. P.  $^{210}\text{Po}$  in marine organisms: a wide range of natural radiation dose domains. *Radiat. Protect. Dosim.* 24:113–117; 1988.
- Carvalho, F. P. Origin and concentrations of  $^{222}\text{Rn}$ ,  $^{210}\text{Pb}$ ,  $^{210}\text{Bi}$  and  $^{210}\text{Po}$  in the surface air at Lisbon, Portugal, at the Atlantic edge of the European continental landmass. *Atmospheric Environ.* 29; 1995.
- Carvalho, F. P.; Fowler, S. W. An experimental study on the bioaccumulation and turnover of polonium-210 and lead-210 in marine shrimp. *Mar. Ecol. Prog. Ser.* 102:125–133; 1993.
- Carvalho, F. P.; Fowler, S. W. A double-tracer technique to determine the relative importance of water and food as sources of polonium-210 to marine prawns and fish. *Mar. Ecol. Prog. Ser.* 103:251–264; 1994.
- Commission of the European Communities. The radiological exposure of the population of the European Community from radioactivity in North European waters Project "Marina." Report EUR 12483. Radiation Protection No. 47: 566. Luxembourg, CEC; 1990.
- Chamberlain, A. C. Fallout of lead and uptake by crops. *Atmospheric Environ.* 17:693–706; 1983.
- Flynn, W. W. The determination of low levels of polonium-210 in environmental materials. *Anal. Chim. Acta* 43:221–227; 1968.
- Food and Agriculture Organization. Fishery statistics, catches and landings 1991. Rome: FAO Fisheries Series No. 40; 1993. Gilbert, G. E.; Bishop, C. T.; Casella, V. R.; Aguirre, A. G. Radionuclides of U, Th, Po and Pb in residents of central Ohio and coal miners of West Virginia. *Health Phys.* 55:571–574; 1988.
- Hill, C. R. Polonium-210 in man. *Nature* 208:423–428; 1965.
- Hill, C. R. Routes of uptake of  $^{210}\text{Po}$  into human tissues. In: *Radioecological concentration processes. Proceedings of an International Symposium in Stockholm, 1966.* Oxford: Pergamon Press; 1967:297–302.
- Holtzman, R. B. Application of radiolead to metabolic studies. In: Nriagu, J. O., ed. *The biogeochemistry of lead in the environment (Part B).* Amsterdam: Elsevier/North-Holland Publishers; 1978:37–96.
- Hunt, G. J.; Allington, D. J. Absorption of environmental polonium-210 by the human gut. *J. Radiol. Prot.* 13:119–126; 1993.
- International Commission on Radiological Protection. Limits for intakes of radionuclides by workers. ICRP Publication 30; Oxford, U.K. 1979.
- International Commission on Radiological Protection. Recommendations of the ICRP. ICRP Publication 60. *Annals of the ICRP* 21(1/3); Oxford, U.K. 1991.
- Jaworowski, Z. Radioactive lead in the environment and in the human body. *Atomic Energy Rev.* 7:3–45; 1969.
- Kametani, K.; Ikebuchi, H.; Matsumura, T.; Kawakami, H.  $^{226}\text{Ra}$ ;  $^{210}\text{Pb}$  concentrations in foodstuffs. *Radioisotopes* 30:681–683; 1981.
- Kendall, G. M.; Harrison, J. D.; Fell, T. P. Report of the Nuclear Energy Agency expert group on gut transfer factors: implications for dose per unit intake. *Radiat. Protect. Dosim.* 25:59–65; 1988.
- Khandekar, R. N. Polonium-210 in Bombay diet. *Health Phys.* 33:148–150; 1977.
- Ladinskaya, L. A.; Parfenov, Y. D.; Popov, D. K.; Fedorova, A. V.  $^{210}\text{Pb}$ ;  $^{210}\text{Po}$  content in air, water, foodstuffs, and the human body. *Arch. Environ. Health* 27:254–258; 1973.
- Magno, P. J.; Groulx, P. R.; Apidianakis, J. C. Lead-210 in air and total diets in the United States during 1966. *Health Phys.* 18:383–388; 1970.
- Moroz, B. B.; Parfenov, Y. D. Metabolism and biological effects of polonium-210. *Atomic Energy Rev.* 10:175–232; 1972.
- Mussalo-Rauhamaa, H.; Jaakkola, T. Plutonium-239,  $^{240}\text{Pu}$ ;  $^{210}\text{Po}$  contents of tobacco and cigarette smoke. *Health Phys.* 49:296–301; 1985.
- Parfenov, Y. D. Polonium-210 in the environment and in the human organism. *Atomic Energy Rev.* 12:75–143; 1974.
- Phipps, A. W.; Kendall, G. M.; Stather, J. W.; Fell, T. P. Committed equivalent organ doses and committed effective doses from intakes of radionuclides. Chilton, Didcot, Oxfordshire, U.K.: National Radiological Protection Board; NRPB-R245; 1991.
- Santos, P. L.; Gouvea, R. C.; Dutra, I. R.; Gouvea, V. A. Accumulation of  $^{210}\text{Po}$  in foodstuffs cultivated in farms around the Brazilian mining and milling facilities on Poco de Caldas Plateau. *J. Environ. Radioact.* 11:141–149; 1990.
- Simon, S. L.; Ibrahim, S. A. The plant/soil concentration ratio for calcium, radium, lead and polonium: evidence for non-linearity with reference to substrate concentration. *J. Environ. Radioact.* 5:123–142; 1987.
- Smith-Briggs, J. L.; Bradley, E. J. Measurement of natural radionuclides in U. K. diet. *Sci. Tot. Environ.* 35:431–440; 1984.
- Smith-Briggs, J. L.; Bradley, E. J.; Potter, M. D. The ratio of lead-210 to polonium-210 in U. K. diet. *Sci. Tot. Environ.* 54:127–133; 1986.
- Spencer, H.; Hotzman, R. B.; Kramer, L.; Ilcewicz, F. H. Metabolic balances of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  at natural levels. *Radiat. Res.* 69:166–184; 1977.
- Takata, N.; Watanabe, H.; Ichikawa, R. Lead-210 content in foodstuffs and its dietary intake in Japan. *J. Radiat. Res.* 9:29–34; 1968.
- Takizawa, Y.; Zhao, L.; Yamamoto, M.; Abe, T.; Ueno, K. Determination of  $^{210}\text{Pb}$  and  $^{210}\text{Po}$  in human tissues of Japanese. *J. Radioanalyt. Nucl. Chem.* 138:145–152; 1990.
- Thomas, P. A. Dosimetry of  $^{210}\text{Po}$  in humans, caribou, and wolves in northern Canada. *Health Phys.* 66:678–690; 1994.
- Torvik, E.; Pfizer, E.; Kereiakes, J. G.; Blanchard, R. Long term effective half-lives for lead-210 and polonium-210 in selected organs of the male rat. *Health Phys.* 26:81–87; 1974.
- United Nations Scientific Committee on the Effects of Atomic Radiation. Sources and effects of ionizing radiation. New York: United Nations; 1977.
- United Nations Scientific Committee on the Effects of Atomic Radiation. Ionizing radiation: sources and effects. New York: United Nations; 1982.
- United Nations Scientific Committee on the Effects of Atomic Radiation. Sources, effects and risks of ionizing radiation. New York: United Nations; 1988.



- United Nations Scientific Committee on the Effects of Atomic Radiation. Sources and effects of ionizing radiation. New York: United Nations; 1993.
- Vasconcellos, L. M. H.; Amaral, E. C. S.; Vianna, M. E.; Penna Franca, E. Uptake of  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$  by food crops cultivated in a region of high natural radioactivity in Brazil. *J. Environ. Radioact.* 5:287-302; 1987.
- Watson, A. P. Polonium-210; lead-210 in food and tobacco

products: transfer parameters and normal exposure and dose. *Nucl. Safety* 26:179-191; 1985.

- Yamamoto, M.; Abe, T.; Kuwabara, J.; Komura, K.; Ueno, K.; Takizawa, Y. Polonium-210; lead-210 in marine organisms: intake levels for Japanese. *J. Radioanal. Nucl. Chem.* 178:81-90; 1994.

