Radioprotection 50(2), 123-128 (2015)
© EDP Sciences 2015
DOI: 10.1051/radiopro/2014039

Available online at:
www.radioprotection.org

Article

# $^{210}$Pb and $^{210}$Po activities in French foodstuffs

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Received 15 July 2014 – Accepted 24 November 2014

**Abstract –** The data on $^{210}$Pb and $^{210}$Po activities in French foodstuffs acquired during the last 15 years are numerous enough to derive reference values which will be usable to assess the dose to the French population due to the intake of these two natural radionuclides. The means and ranges are close to those observed in various countries and are most often higher than the reference values proposed by UNSCEAR. Mussels and oysters exhibit the highest $^{210}$Po activities of all kinds of foodstuffs, with a mean value of 27 Bq.kg$^{-1}$ fresh weight , followed by crustaceans and small fish species (anchovies, mullets, sardines, etc.) with $^{210}$Po activities in the order of 3 to 10 Bq.k$^{-1}$ fresh weight; cephalopods and other fish species presenting activities around 1 to 3 Bq.kg$^{-1}$ fresh, close to the UNSCEAR reference value. Below these highest $^{210}$Po activities are those of $^{210}$Po and $^{210}$Pb in terrestrial kinds of foodstuffs, by decreasing order: meats (around 1 Bq.kg$^{-1}$ fresh), cereals (0.4 Bq.kg$^{-1}$), leafy vegetables (0.3 Bq.kg$^{-1}$), other vegetables and fruits (0.1 Bq.kg$^{-1}$), and milk (from 0.03 to 0.1 Bq.L$^{-1}$).

**Keywords:** polonium-210 / lead-210 / foodstuffs

## 1 Introduction

The study carried out by Picat *et al.* (2002) showed that the French data were at that time insufficient to have a good idea of activity levels of natural radionuclides in French foodstuffs. For this reason, no more accurate internal dose assessments such as those performed by UNSCEAR on the global scale could be made for France. However, since the mid-90s, $^{210}$Pb is regularly provided in the results of gamma-spectrometry analysis. The main source of $^{210}$Pb and $^{210}$Po in the terrestrial environment is $^{222}$Rn gas, which diffuses into the atmosphere from rocks and soil. Attached to aerosol particles, these two nuclides are transported back to soils, plants and aquatic environments by dry and wet deposition. The resulting contamination is higher than that due to root transfer, which is very low. However, the main transfer pathway for plants is the foliar deposit of re-suspended soil particles (Coppin and Roussel-Debet, 2004). This process explains partly why, despite the fact that $^{210}$Po activities in air are in the order of 10 times lower than those of $^{210}$Pb (Daish *et al.*, 2005), the $^{210}$Po/$^{210}$Pb activity ratio in plant food is centered on 1 (Smith-Briggs *et al.*, 1986; Pietrzack-Flis *et al.*, 1997; McDonald *et al.*, 1999; UNSCEAR, 2000). The transfer of polonium and lead to animal products results mainly from their ingestion. Despite the fact that the transfer factors given by the IAEA (2012) were based on a very limited set of data, the transfer intensity of these two nuclides is quite similar. It results in an activity ratio also centered on unity. So, the numerous results of $^{210}$Pb provide a good indication of $^{210}$Po levels in terrestrial foodstuffs.

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In the marine environment, $^{210}$Po and $^{210}$Pb come from soil washout, the outgassing of radon in water, and the deposit on the water surface of radon-chain radionuclides produced on continental soils. Unlike in the terrestrial environment, their activities in marine organisms are very different. The $^{210}$Po/$^{210}$Pb activity ratio observed in seawater is around 1, often slightly less than 1 (Fowler, 2011). This ratio becomes much greater than 1 in organisms. Despite the fact that both lead and polonium have a high propensity to bind to particles, the strong assimilation of polonium by phytoplankton and its trophic transfer to zooplankton leads to a $^{210}$Po/$^{210}$Pb activity ratio of 2 to 10 in phytoplankton or algae, and 2 to 100 or more in zooplankton. Therefore, $^{210}$Po activities decrease with trophic level: consumers of plankton (filter organisms such as shellfish and small fish such as anchovies) have the highest activities, while large fish such as tuna have the lowest activities. Due to the significant quantities of $^{210}$Po in seafood, $^{210}$Po analyses have been carried out since 2002 on mussels, oysters and fish from the Channel and the Mediterranean coast. The aim of the present study is to obtain mean values of activities of these two nuclides, which have a good representativeness for the various kinds of foodstuffs produced on the territory and consumed by the population and suitable for internal dose assessments.

## 2 Materials and methods

For vegetable foodstuffs, the samples come from 8 to 32 French Districts (of a total number of 90 metropolitan Districts) and are representative of the main sites of foodstuff

**Table 1.** $^{210}$Pb and $^{210}$Po in vegetal foodstuffs: mean [range] in Bq.kg$^{-1}$ fresh weight; (number of samples for this study).

| Country | | Leafy-vegetables | Root-vegetables and fruits | Cereals (grain) | References |
|---|---|---|---|---|---|
| France | $^{210}$Pb | 0.34 [0.08–4.2] (501) | 0.12$^{i}$ [0.05–0.98] (59) | 0.41 [0.15–18.4]$^{[w]}$(74) | This study (501) |
| Germany | $^{210}$Pb | 0.3$^{ii}$ [0.004–1.3] | 0.2 [0.02–2.3] | 1.4 [0.04–10.2]$^{[U]}$ | BfS, 2003 |
| | $^{210}$Po | 0.2$^{ii}$ [0.004–1.1] | 0.1 [0.02–1.1] | 0.3 [0.2–1.94]$^{[U]}$ | |
| UK | $^{210}$Pb | 0.34 [0.034–2.5] | | | RIFE, 1996–2010 |
| | $^{210}$Po | 0.13 [0.008–0.9] | | | |
| | $^{210}$Pb | | 0.16 [0.1–0.29] | 0.16-0.26 [0.08–0.69]$^{[U]}$ | McDonald |
| | $^{210}$Po | | 0.12 [0.02–0.4] | 0.23-0.34 [0.01–0.63]$^{[U]}$ | *et al.*, 1999 |
| Finland | $^{210}$Pb | | | 0.29 [0.17–0.47]$^{[w]}$<br>0.36 [0.11–0.52]$^{[O]}$<br>0.36 [0.08–0.56]$^{[B]}$ | Turtiainen *et al.*, 2011 |
| India | $^{210}$Po | 0.34$^{iii}$ [0.1–0.6] | | 0.037 [0.032–0.045]$^{[W]}$<br>0.5 [0.3–0.7]$^{[Rz]}$ | Kannan *et al.*, 2001 |
| Turkey | $^{210}$Po | [0.18–9.4] | | | Ekdal *et al.*, 2006 |
| Egypt | $^{210}$Po | [0.045–3] | [0.14–0.53] | [<0.01–0.4]$^{[W]}$ | Din, 2011 |
| Poland | $^{210}$Pb | [0.03–0.05] | 0.0296 | 0.11$^{[W]}$ ; 0.16$^{[R]}$ | Pietrzack-Flis |
| | $^{210}$Po | [0.04–0.075] | 0.0549 | 0.092$^{[W]}$ ; 0.14$^{[R]}$ | *et al.*, 1997 |
| S-Africa | $^{210}$Po | | | [0.28–9.6]$^{[R]}$ [1.4–5.6]$^{[O]}$ | Louw *et al.*, 2009 |
| Europe | $^{210}$Pb | 0.08$^{iv}$ [0.004–7.4] | 0.03$^{iv}$ [0.018–4.9] | 0.05$^{iv}$ [0.05–4.0] | UNSCEAR, 2000 |
| | $^{210}$Po | 0.10$^{iv}$ [0.004–4.1] | 0.04$^{iv}$ [0.012–5.2] | 0.06$^{iv}$ [0.02–1.9] | |

$^{i}$Fruits of trees only; $^{ii}$value given for cabbage; $^{iii}$spinach ; $^{iv}$UNSCEAR reference value;
$^{[W]}$Wheat; $^{[R]}$Rye; $^{[O]}$Oats; $^{[B]}$Barley; $^{[Rz]}$Rize; $^{[U]}$Unspecified

production. Fishes and mussels come from various locations along the coasts of the Channel, the Atlantic and the Mediterranean coast. All samples are directly bought from the producer. They are washed and the cuticles of cereal grains are already removed. Samples are dried at a temperature of 80 °C, then burned at 480 °C with a slow increase in order to avoid ignition. They are then packed in order from 17 to 60 mL. The measurements of $^{210}$Pb were performed by the metrology laboratory of IRSN-Orsay under conditions described by Bouisset *et al.* (1999). The analyses of $^{210}$Po on seafood were performed at the IRSN-STEME laboratory (Le Vésinet, France) by alpha spectrometry after acid digestion of around 0.2 to 1.1 g of dried matter followed by deposition onto a stainless steel disc. The method is described in Charmasson *et al.* (2011).

## 3 Results

All results are given in Bq.kg$^{-1}$ of fresh mass except for soil, which is in Bq.kg$^{-1}$ of dry matter. The averages and ranges observed in French vegetable foodstuffs are close to those observed in various countries (Table 1). The reference values provided by UNSCEAR are most often much lower than these averages, although the ranges reported by this organism for Europe are in good agreement.

Although the uranium activity in soil is not known for each foodstuff sampling site, data of leafy vegetables and cereals are numerous enough to compare activities of samples from French districts with uranium activities in soil of

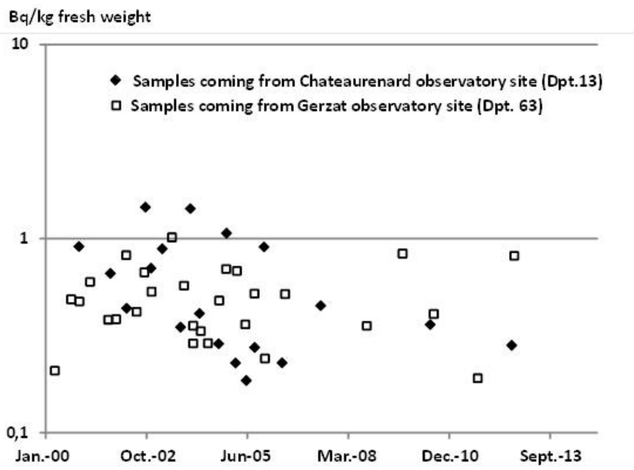


**Fig. 1.** $^{210}$Pb (Bq.kg$^{-1}$ fresh) activities in lettuces coming from 2 observatory sites having low (Châteaurenard ≅ 25 Bq.kg$^{-1}$ of dry soil) and higher (Gerzat ≅ 50 Bq.kg$^{-1}$) U-Th background, respectively.

around 24 Bq.kg$^{-1}$ with those of around 52 Bq.kg$^{-1}$ according to Renaud *et al.* (2015). The averages of $^{210}$Pb activities cannot be distinguished either for leafy vegetables (0.37 ± 0.11 Bq.kg$^{-1}$ against 0.41 ± 0.15 Bq.kg$^{-1}$) or for cereals (0.49 ± 0.2 Bq.kg$^{-1}$ against 0.51 ± 0.4 Bq.kg$^{-1}$). This is confirmed in Figure 1, which allows comparing lettuce samples from two sites: Châteaurenard (with mean soil activity of around 25 Bq.kg$^{-1}$) and Gerzat (with soil activity of around 50 Bq.kg$^{-1}$). As expected, the high variability of activities

**Table 2.** $^{210}Pb$ and $^{210}Po$ in milk and meat; mean (min–max) values in $Bq.kg^{-1}$ fresh for meat and $Bq.L^{-1}$ for milk.

| Country | | Milk | Meat | References |
|---|---|---|---|---|
| France | $^{210}Pb$ | 0.12 (0.024–0.42) | 0.34 (0.13–0.54) | This study |
| | $^{210}Pb$ | 0.027 (0.001–0.17) | 0.099 (0.028–0.23) | This study (estimated value) |
| | $^{210}Po$ | 0.030 (0.013–0.042) | | This study (estimated value) |
| Germany | $^{210}Pb$ | 0.04 (0.004–0.26) | 0.5 (0.1–1) | BfS, 2003 |
| | $^{210}Po$ | 0.024 (0.003–0.07) | 2 (0.2–4.0) | |
| India | $^{210}Po$ | 0.01 (0.008–0.012) | (0.04–0.10) | Kannan *et al.*, 2001 |
| Europe | $^{210}Pb$ | (0.011–0.28) | (0.015–3.7) | UNSCEAR, 2000 |
| | $^{210}Po$ | (0.002–0.22) | (0.037–67) | |

within a single site occults any inter-site differences that might have been expected as a result of soil activities. This variability at each site may be due mainly to foliar transfer due to the resuspension of soil particles, or to radon outgassing conditions during plant growth.

Among 700 gamma-spectrometry analyses on French milk samples, only 14 give a result above the Detection Limit (DL) for $^{210}Pb$. The range of measured values varies from 0.024 to 0.42 with an average of 0.12 $Bq.L^{-1}$. Concerning meat, among 97 analyses since 1994, only 2 allow quantifying $^{210}Pb$ activity: 0.13 and 0.54 $Bq.kg^{-1}$ fresh. Over the period 2000−2013, DL varies mainly between 0.08 and 1 $Bq.kg^{-1}$ fresh. Bibliographic data on $^{210}Pb$ and $^{210}Po$ in milk and meat are also rare. Table 2 shows the ranges of values provided by the European publications consulted by UNSCEAR, as well as more recent publications. The reference values proposed for milk by UNSCEAR in 2000, 0.015 $Bq.L^{-1}$ for both radionuclides, are low compared with these ranges. The values proposed by UNSCEAR in 1993, 0.04 and 0.05 $Bq.L^{-1}$ for $^{210}Pb$ and $^{210}Po$, respectively, were more in line with European data.

Another approach is to estimate $^{210}Pb$ and $^{210}Po$ activities in milk and meat on the basis of activities measured in fodder (grass and hay) on one hand, and on the other hand using the transfer factors to animal products recommended by the IAEA (2010). Among 574 analyses of cultivated grassland and forage, 548 (95%) allow quantifying $^{210}Pb$ activity. The average is 2.82 ± 0.21 $Bq.kg^{-1}$ fresh or 12.8 ± 1.1 $Bq.kg^{-1}$ dry. The calculated value for milk (0.031 $Bq.L^{-1}$ for $^{210}Po$ or $^{210}Pb$) is close to the average value from a German publication (0.024 and 0.04 $Bq.L^{-1}$ for $^{210}Po$ and $^{210}Pb$, respectively), and also close to the reference values proposed by UNSCEAR in 1993 (0.04 and 0.05 $Bq.L^{-1}$, respectively). In the case of beef, the calculated values for $^{210}Po$ are practically equal to the German average.

Despite their variability, the bibliographic data and the few French data available for seafood are in fairly good agreement (Table 3) and allow clearly distinguishing the different kinds of seafood by order of $^{210}Po$ increasing activity: the main fish and cephalopod species, the small fishes consuming plankton (anchovies and sardines notably), and the crustaceans and shellfish. The higher values measured by Rollo *et al.* (1992) are noted, however. The value of 3.5 $Bq.kg^{-1}$ proposed by UNSCEAR[1] could apply to a mixture of large and small fishes, but not to crustaceans and shellfish. It should be noted that the mean value reported for mussels is higher: 42 $Bq.kg^{-1}$ (RIFE, 2013), whereas it is lower for other shell species (18 $Bq.kg^{-1}$), and notably for whelks and winkles (6.5 and 13 $Bq.kg^{-1}$, respectively).

As shown in Table 4, $^{210}Pb$ activities in seafood are lower and less variable than those of $^{210}Po$ and maybe slightly underestimated by the UNSCEAR reference value: 0.2 $Bq.kg^{-1}$. It is noteworthy that the activities of shellfish are higher, around 1 $Bq.kg^{-1}$.

## 4 Conclusion

The data on $^{210}Pb$ activities in vegetable foodstuffs and seafood, and on $^{210}Po$ in seafood obtained during the last 15 years are now numerous enough to give mean values representative of the French metropolitan territory. For milk and meat, the few available results and the estimated values based on grass activities allow situating the French values within those observed in other countries.

Thus, this study allows deriving useful reference values to assess the dose to the French population due to the intakes of $^{210}Po$ and $^{210}Pb$ by foodstuff ingestion. This study shows that due to the large variability of activities, it is not possible to distinguish $^{210}Pb$ activities in leafy vegetable, cereal and grass samples from Districts with a gap of only a factor of 2 between their uranium contents in soils.

The means are most often higher (as far as 8 times higher in the case of cereals) than the reference values proposed by UNSCEAR. This is mainly due to the aim of UNSCEAR to propose reference values on a global scale.

The French data on mussels and oysters confirm that these products present the highest $^{210}Po$ activities of all kinds of foodstuffs, with a mean value of around 27 $Bq.kg^{-1}$ fresh. The activities of $^{210}Po$ measured in crustaceans from English and Italian coasts are probably quite representative of those of French coasts: with average values most often ranging from 3 to 10 $Bq.kg^{-1}$. $^{210}Po$ activities in fishes are lower, close to the UNSCEAR reference value, except for some small fish species (anchovies, mullets, sardines, mackerels), which have $^{210}Po$ activities on average around 5 times higher. Below these highest $^{210}Po$ activities in marine products are the activities of

[1] The UNSCEAR reference value for fish products is 2 $Bq.kg^{-1}$ fresh using a factor of 0.6 in order to take into account the $^{210}Po$ decay during the delay between fishing and consumption. So the effective value for fish products was 3.5 $Bq.kg^{-1}$.

**Table 3.** $^{210}$Po activities in seafood given in literature and measured by IRSN (Bq.kg$^{-1}$ fresh); mean [range] and (number of samples).

| | **Carvalho *et al.*, 1995, 2011** | **Cherry *et al.*, 1994** <br> ***Heyraud and Cherry*, 1979** | **Pollard *et al.*, 1998** <br> ***Rollo et al.*, *1992*** | **Young *et al.*, 2002** <br> **Study and *biblio*** | ***Desideri et al.*, *2011*** <br> **Melli *et al.*, 2008** | **Connan *et al.*, 2007** <br> ***This study*** |
|---|---|---|---|---|---|---|
| Anchovies | [12–47] | 36 [25–47] (3) | | | 27,5 [18–45] (4) | |
| Sardine | 66 | 5.2 [3.5–18] (5) | | | | |
| Mackerel | [3.5–19] | 8.3 | 1.9 [0.7–3.2] (3) | | 2.8 [1.8–4] (4) | |
| Mullet | | | | | 5.6 [5.2–6.6] (4) | 10 [5–18] (3) |
| Herring/scad | | | 1.7 [0.4–3.8] (4) | | 1.8 [0.6–2.4] (3) | |
| Small coastal fish | 0.5 | 2.9 [0.5–8.3] (16) | | | 0.9 [0.4–2.1] (8) | |
| Hake | 6.4; 7.8 | | | | 2.7 [1.6–4] (4) | |
| Plaice | | | 2.8 [1.3–6.4] (9) | 2.4 [0.9–4.4] (7) <br> *1.5 [0.8–6.4]* | | |
| Seabream | 2.4 | | | | 1.8 [1.2–2.2] (4) | 0.9*1* |
| Tuna/swordfish | 2.1-3 | *4.1 [3.4–4.7] (2)* | | | 5.9 [1.1–13] (4) | |
| Cod | | | 0.9 [0.4–2.0] (6) <br> *4.5 [2.3–8.4] (3)* | 0.6 [0.2–1.1] (5) <br> *0.9 [0.28–2]* | 1.6 [1–2.3] (4) | |
| Whiting | | | 0.9 [0.2–2.2] (7) | 0.8 [0.7–0.8] (2) <br> *1.2 [0.2–2.2]* | | |
| Sole/turbot/ray | | | 0.6 [0.1–1.5] (6) | | 0.8 [0.4–1.5] (4) | 0.4 |
| Small shark | [0.12–1.7] | | | | | |
| Crabs | | | *116 [27–319] (4)* | 18 [4.1–35] (19) <br> *13 [9–17] (2)* | | |
| Lobster | | | 2.5 [1.6–3.3] (2) <br> *42 [8.4–62.6] (3)* | 5.3 [2–10] (8) | 1.4 [0.7–2] (4) | |
| Shrimp | | | 5.3 [0.8–13] (8) | 9.5 [1.1–30] (18) | | |
| Nethrop/crawfish | | | | 2.7 [1–3.5] (3) | 3.6 [1.2–5.9] (4) | |
| Octop./squid/cuttle | [1.6–2.8] | *0.7 [0.4–1.0] (5)* | | | 1.9 [0.3–6.6] (8) | |
| Winkle | 283 | | *74 [7–371] (74)* | 16 [6–25] (8) <br> *13 [4–38] (38)* | | |
| Whelk | | | | 7.1 [3–11] (6) <br> *6.9 [3–11] (6)* | | |
| Limpet | 11.6 | | | 8.4 [5.9–15] (7) <br> *9.4 [5.9–15] (7)* | | |
| Mussel | | | 39 [30–75] (28) | 38 [19–2] (17) <br> *17 [5–57] (17)* | 18 [8–23] (4) <br> *22 [8–54] (180)* | 27 [17–52] (35) <br> *26 [10–56] (29)* |
| Clam, Cockle... | [45–152] | | | 24 [16–36] (4) <br> *20 [18–29] (3)* | 13 [12–16] (4) | |
| Oyster | 10 | | 30 [23–36] (2) | | | 29 [14–87] (40) |

**Table 4.** $^{210}$Pb activities in seafood given in literature (Bq.kg$^{-1}$ fresh); mean [range] and (number of samples).

| | **Carvalho *et al.*, 1995, 2011** | **Pollard *et al.*, 1998**<br>**Rollo *et al.*, 1992** | **Young *et al.*, 2002**<br>***Young et al.(biblio)*** | **Melli *et al.*, 2008, 2013**<br>***Heyraud and Cherry, 1979*** | **Connan *et al.*, 2007**<br>***This study*** |
|---|---|---|---|---|---|
| Anchoy/Sardine | [0.4–1] | | | | |
| Mackerel | 0.15 [0.1–0.7] (4) | 0.7 [0.2–1.5] (6) | | | |
| Hake | 0.16 [0.15–0.17] (2) | 0.8 [<0.2–2] (4) | | | |
| Plaice | | 0.6 [<0.1–1.4] (10) | 0.14 [0.04–0.55] (7) | | |
| Stream | 0.84 | | | | |
| Tuna | 0.32 | | | | |
| Cod | | | 0.05 [0.01–0.11] (5) | | |
| Miscellaneous | 0.33 | 0.3 [0.3–0.5) (4) | | | 0.4<br>*0.55 [0.19–1] (108)* |
| Crabs | | *0.5* | 0.24 [0.04–0.76] (19) | | |
| Lobster | | | 0.2 [0.02–0.79] (8) | | |
| Shrimp | | 0.2 [0.1–0.3] (6) | 0.27 [0.05–2.37] (18) | | |
| Nethrop | | | 0.12 [0.01–0.23] (2) | | |
| Octop./squid/cuttle | | | | *1.15 [0.2–3.2]* | |
| Winkle | 5.2 | *5.8 [2.6–10] (3)* | 1.5 [1.1–2.0] (4)<br>*0.9 [0.3–1.5] (8)* | | |
| Whelk | | | 0.4 [0.18–0.61] (4) | | |
| Limpet | 4.2 | | 2.2 [0.7–4.9] (7) | | |
| Mussel | 2.6 | 0.6 [0.2–0.8] (4)<br>*2.1 [1–4] (5)* | 1.8 [0.7–6.8] (17)<br>*1.6 [0.6–5.7] (12)* | 1.7 [0.3–3.6] (64) | 1.1 [0.7–1.7] (35)<br>*1.2 [0.19–17] (848)* |
| Clam, Cockle ... | [2.2–5.6] (3) | | | | |
| Oyster | 2.9 | | 0.9 [0.6–1.3] (8) | | 0.8 [0.4–0.9] (16) |

both $^{210}$Po and $^{210}$Pb in other terrestrial kinds of foodstuffs, by decreasing order: meats (around 1 Bq.kg$^{-1}$ fresh), cereals (0.4 Bq.kg$^{-1}$), leafy vegetables (0.3 Bq.kg$^{-1}$ fresh), other vegetables and fruits (0.1 Bq.kg$^{-1}$), and milk (from 0.03 to 0.1 Bq.L$^{-1}$).

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**Cite this article as:** Ph. Renaud, S. Roussel-Debet, L. Pourcelot, H. Thébault, J. Loyen, R. Gurriaran. $^{210}$Pb and $^{210}$Po activities in French foodstuffs. Radioprotection 50(2), 123-128 (2015).