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ARTICLE

Internal doses of French adult population linked to the intake of radionuclides from the decay-chains of uranium and thorium by foodstuffs ingestion

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Abstract – This study provides the first dose assessment to the French adult population due to the intake of radionuclides from the decay chains of uranium and thorium by foodstuff ingestion (water consumption excepted). This dose varies widely with the consumption of seafood, from less than 200 μSv.y^{-1} for people who do not consume shellfish or crustaceans at all, to more than 2,000 μSv.y^{-1} for the biggest consumers (about 150 kg.y^{-1} according to specific dietary surveys carried out along the French seaside). For moderate consumers of seafood (around 4.6 kg.y^{-1}), who probably represent a major part of the population, this internal dose would be around 330 μSv.y^{-1}. This variable consumption of seafood overshadows all the other causes of variability of these internal dose estimates.

Keywords: ingestion doses / radionuclides of uranium and thorium decay-chains

1 Introduction

Potassium 40 and the radionuclides of the ^{238}U and ^{232}Th decay chains are the main contributors to the internal dose due to the intake of natural radionuclides by the ingestion of foodstuffs. The potassium content being strictly regulated by the organism and the amount of ^{40}K being well known and stable, the dose due to this nuclide is thus around 170 μSv.y^{-1} for an adult (UNSCEAR, 2000). However, many studies show that the dose due to the intake of U-Th chain nuclides depends on the foodstuff activities, and above all on dietary habits, notably the consumption of seafood (Rollo *et al.*, 1992; Yamamoto *et al.*, 1994; Pietrzack-Flis *et al.*, 1997a, 1997b, 2001; Desideri *et al.*, 2011). Due to their high dose conversion factors and their high activities in foodstuffs, ^{210}Po, ^{210}Pb, ^{228}Ra and ^{226}Ra could contribute more than 98% to this dose (UNSCEAR, 2000).

The study carried out by Picat *et al.* (2002) showed that the French data were at this time insufficient to have a good idea of the activity levels of natural radionuclides in French foodstuffs, particularly as regards these four nuclides. For this reason, no more accurate dose assessments such as those performed by UNSCEAR on the global scale could be made for France. Since the mid-90s, ^{210}Pb, ^{228}Ac and ^{214}Bi (decay products of ^{228}Ra and ^{226}Ra, respectively) are quite regularly provided in the results of gamma spectrometry performed by the Institute for Radioprotection and Nuclear Safety (IRSN) for its missions of radiological monitoring in the environment and research studies. Moreover, due to the high amount of ^{210}Po in seafood, some ^{210}Po analyses have been carried out since 2002 on mussels, oysters and fishes from the Channel and the Mediterranean coast. On this basis, reference values have been established by Renaud *et al.* (2015a, 2015b) concerning the activity of these four nuclides in French foodstuffs.

The aim of this study is to estimate the internal doses to the French population using these reference values and the consumption rates established from French dietary surveys.

2 Material and method

Table 1 presents the reference values obtained by Renaud *et al.* (2015a, 2015b) for the different categories of foodstuffs proposed by UNSCEAR, to which 3 supplementary foodstuffs have been added (small fishes, crustaceans and shellfish). In France, the mean value of ^{238}U activity in soil is equal to ^{232}Th one (around 40 Bq.kg^{-1} dry), and most French soils present activities between 20 (in limestone regions) and 50 Bq.kg^{-1} dry (on granitic or metamorphic rocks). Therefore, previous studies show that it is not possible to distinguish activities of samples from regions with a gap of only a factor of 2 between their soil activities due to the strong variability of transfer to plants and animal products. This is in agreement with Blanco Rodriguez *et al.* (2002), having shown that the linearity of the relation between soil activities and plant activities in real agricultural conditions (unlike experimental conditions) is effective for a concentration range of at least 2 orders of magnitude. Furthermore, temporal records measured for the same crop on

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Table 1. Reference values of ^{226}Ra, ^{228}Ra, ^{210}Po and ^{210}Pb activities (mBq.kg^{-1} fresh matter) from Renaud *et al.* (2015a, 2015b).

	^{226}Ra	^{210}Pb	^{210}Po	^{228}Ra
Milk products	11	27	30	10
Meats	50	100	100	43
Cereals	180	410	410	180
Leafy-vegetables	90	340	340	90
Root-vegetables and fruits	51	120	120	57
Fishes	160	550	2000	160
Crustaceans and shellfish	400	550	22500	130
Large fishes/cephalop.	160	550	2000	160
Small fishes	160	550	10 000	160
Crustaceans	400	550	18 000	130
Shellfish	400	1 000	27 000	130

the same site show that variability at the local level is as significant as the variability between Districts with various U-Th contents in soils. It follows that the average activity of a kind of foodstuff established from the data acquired over the whole territory is representative enough to assess the average intake to a person consuming this kind of food throughout the year, even if this product comes exclusively from a specific place.

Concerning the isotopes of uranium and thorium, the reference values proposed by UNSCEAR are still used for dose assessments, knowing that they potentially contribute less than 2% to the total ingestion dose (UNSCEAR, 2000).

The French dietary habits on the scale of the whole territory are known through 3 national surveys (Bertrand, 1993; Volatier, 2000; Lafay, 2009). The quantities consumed provided in the reports are not directly usable for dose assessment as they are not given for the food categories of Table 1. Most often, it is necessary to sum the rations: lettuce, spinach and cabbage for the leafy vegetable category; and pork, veal, beef and mutton for the meat category. When quantities are those of transformed products or cooked meals, it is necessary to estimate the contribution of the various categories. For example, a sandwich can be considered as composed of 50% cereals and 50% meat, thus neglecting possible components such as vegetables or butter. In the same way, pastries can be seen as a mix of 70% cereals, 20% milk (butter) and 10% neglected others. For the survey reported by Bertrand (1993) this work was done by Mourlon (2012). The same methodology was applied for the two other surveys. In addition to these national surveys, six local surveys were performed in order to find out local dietary habits around some nuclear facilities: Tricastin, Chinon, Marcoule, Gravelines, Blayais and Nord-Cotentin (La Hague) (Dufour and Volatier, 1998; Parache, 2009, 2010, 2011, 2012). These studies specifically acquired for internal dose assessments are much better suited to this study and require only minor adaptations.

Table 2 presents the mean values of the quantity consumed for each foodstuff categories on the basis of the results of these different surveys. The minimum and maximum values, and the names of the surveys corresponding to the min and max values are also given for information. For milk products, meats and fishes, the gaps between min and max values are less than a factor of 2. For cereals and vegetables, it is about 2; in the case of the category "Root vegetables and fruits", the gap of a factor of 3 is linked to a very high consumption of fruits from the Tricastin survey. All the previous studies do not provide enough detail of the consumption of seafood considering its high ^{210}Po activity and thus potentially high contribution to the dose. However, it appears that the total quantity of seafood consumed by people living close to the sea (Gravelines, Blayais and Nord-Cotentin surveys) is about 34 to 44 kg.y^{-1}, whereas it is only 15 to 25 kg.y^{-1} as seen from the national surveys (INSEE, INCA1 and INCA2) and in sites located away from the sea (Marcoule, Chinon and Tricastin). These consumption rates are consistent with the value of 36.8 kg.y^{-1} given by the French Institute for Agricultural Food and Seafood (FranceAgrimer, 2012). The survey named CALIPSO focused specifically on seafood consumption among 1200 persons living in seaside sites (Le Havre, Lorient, La Rochelle and Toulon) and qualified as high seafood consumers. This survey is also interesting because it details the consumption rates for most fish, crustacean and shellfish species, allowing one to obtain separate consumed quantities for the three seafood categories of Table 1 (Table 3).

From CALIPSO and AGRIMER data we can assume that fishes represent around 70% of the total quantity of seafood consumed. The surveys around the Gravelines, Blayais and Nord-Cotentin sites and the conclusions of the CALIPSO project show that the quantities consumed by the biggest seafood consumers (percentiles 95) are around 150 kg.y^{-1} (from 135 to 161 kg.y^{-1}), corresponding to a daily rate of 40 g.d^{-1}.

The last element considered in this study is the dose conversion factors published in ICRP Publication 119 (2012).

3 Results

Due to the variability of the consumption of shellfish and crustaceans, and their significance in terms of the internal dose, it seems more appropriate to distinguish 4 cases.

- The dose received by a large majority of the population who consume about 4.6 kg.y^{-1} of seafood is about 319 μSv.y^{-1}. As expected, this dose is practically entirely due to ^{210}Po, ^{210}Pb, ^{228}Ra and ^{226}Ra.
- People who do not consume any shellfish or crustaceans would receive a dose of 193 μSv.y^{-1}. This dose is 1.5 times above the UNSCEAR assessment (127 μSv.y^{-1} without water consumption) based on a smaller consumption of "fish products" (15 kg.y^{-1}) without clearly considering shellfish and crustaceans.
- For the people who regularly consume shellfish and crustaceans the surveys carried out in seaside sites (from Table 2 and the mean values of Table 3) give similar results: around 730 μSv.y^{-1}.
- The range of doses received by the big consumers of seafood (about 135 to 161 kg.y^{-1}) as characterized by the percentile 95 of the CALIPSO survey (adults and seniors) and Nord-Cotentin reaches 2,000 to 2,600 μSv.y^{-1}.

The dose assessments provided in the literature are sometimes partial and can concern only some kinds of foodstuffs and some radionuclides. Table 4 allows comparing all these evaluations by giving the foodstuffs or the nuclides considered,

Table 2. Mean, min and max values of the quantity consumed for each foodstuff categories from the different surveys, and names of the surveys corresponding to the min and max values in kg.y^{-1}.

Foodstuffs cat.	Mean	Min	Survey	Max	Survey
Milk products	118	102	INCA2	155	Nord-Cotentin
Meats	85	71	INCA2	98	Blayais
Cereals	82	66	Marcoule	123	Nord-Cotentin
Leafy vegetables	31	22	INCA1	40	Tricastin
Root veg. & fruits	214	128	INCA1	337	Tricastin
Fishes 1**	15	6	INSEE	19	Tricastin
Fishes 2**	20	15	Gravelines	23	Nord-Cotentin
Shellfish and crustaceans 1*	4.6	2	INCA2	6	Tricastin
Shellfish and crustaceans 2**	19	19	Nord-Cotentin	20	Gravelines

* National value and continental sites: INCA1, INSEE, Marcoule, Tricastin and Chinon surveys.
** for seaside sites: Gravelines, Blayais and Nord-Cotentin surveys.

and some comments in order to compare with the corresponding partial internal dose from our study. This table shows that the variability of doses is mainly linked to the consumption rates considered, and notably to seafood consumption rates (Table 4). This appears when comparing the results obtained by Pietrzack-Flis *et al.* (2001) with those of Kannan *et al.* (2001), or when comparing Pollard *et al.* (1998) with Rollo *et al.* (1992) in the case of the United Kingdom. Desideri *et al.* (2011) following Ryan *et al.* (1999) propose, as in this study, considering the populations with radically different behaviors regarding the consumption of seafood separately. One can note that among the highest dose assessments, Kannan *et al.* (2001) give a value of 3,364 μSv.y^{-1} linked to the consumption of 25 kg.y^{-1} of crabs.

Behind seafood, cereals exhibit the highest activities for the four main nuclides. Considering their lowest (66 kg.y^{-1}) and highest consumption rates (123 kg.y^{-1}) observed from the various surveys and without any compensation by the consumption of other kinds of foodstuffs, the associated dose variability would be only 17%. Even the absence of cereal consumption would reduce the average dose of 319 μSv.y^{-1} by only 26%.

Concerning water consumption, the 12,000 samples analyzed annually in France for the sanitary control of tap water put bounds on the highest corresponding doses but do not allow determining an average. Indeed, only the samples exceeding the French guideline values (0.1 Bq/L for alpha global activity and 1 Bq/L for beta global activity) are subject to further analysis to assess doses.

It should also be noted that, unlike other foodstuffs for which the mean values have a meaning, a person consumes his tap water or the bottled water of his convenience. Thus, the individual dose is directly related to the dispensing unit or to consumer choices rather than to the national average. In addition, the activities of U-Th decay products in water and their relative contributions to the dose are completely erratic and cannot be predicted without specific measurement results. Although doses are often linked to radium or uranium activities, they can also result from isolated high activities of ^{210}Po or ^{210}Pb due to physical and chemical characteristics of the water. In this context, we can only say that the dose received by French adults consuming 730 L.y^{-1} could exceed 100 μSv.y^{-1} only for less than 0.2% of the tap water consumers

Table 3. Mean value and percentile-95 of seafood consumption for adults and seniors from the CALIPSO survey in kg.y^{-1}.

Foodstuff cat.	Adults (17–64 y)		Seniors (>65 y)	
	Mean	P95	Mean	P95
Large fishes/cephalop.	31	69	36	71
Small fishes	4,2	19	6,5	30
Crustaceans	6.7	27	8.4	31
Shellfish	4.6	19	4.7	20
Total	46.5	161	55.5	152

(ASN-DGS-IRSN, 2011) or for people who have chosen one of the 4% of bottled waters whose consumption can lead to exceeding this dose (ASN-DGS-IRSN, 2013). For the wide majority of French adults, the dose by ingestion of water is probably between a few μSv.y^{-1} and a few tens of μSv.y^{-1}.

4 Conclusion

This study provides the first dose assessment to the French adult population due to the intake of radionuclides from the decay chains of uranium and thorium. This estimate was made possible by the acquisition during the last 15 years of data about the main radionuclide contributors to this dose: ^{210}Po, ^{210}Pb, ^{228}Ra and ^{226}Ra. For most adults this dose was evaluated at 319 μSv.y^{-1}, being 1.5 times higher than the UNSCEAR assessment (without water consumption), which has been considered until now as the reference for France. The gap is mainly due to more precise estimates of seafood activities, notably for ^{210}Po, and of their consumption. As shown in the literature, the variability of dietary habits concerning shellfish and crustaceans is the main cause of the variability for this dose. Some specific data show that people living near the French coasts consume more seafood, leading to a dose of around 730 μSv.y^{-1}. The literature studies based on similar consumptions of these products obtain similar results.

The range of doses received by the biggest consumers of seafood would reach 2,000 to 2,600 μSv.y^{-1}.

However, a population who does not consume shellfish or crustaceans at all probably exists, who would receive a dose lower than 200 μSv.y^{-1}. This variable behavior regarding seafood overshadows all the other cause of variability, notably

Table 4. Comparison of different dose assessments.

Dose (μSv.y^{-1})	Nuclides	Foodstuffs	Comments; rations in kg.y^{-1}	References	This study (μSv.y^{-1})
90	All	All	Fish: 1.9 No crustaceans, no shellfish	Pietrzack-Flis *et al.*, 2001	319
731	^{210}Po	All	Fish: 25; Crustaceans: 50	Kannan *et al.*, 2001	319 and 640*
123	^{210}Po	Vegetable foodstuffs	Vegetarian diet	Kannan *et al.*, 2001	98
65 to 118	^{210}Po and ^{210}Pb	Vegetable foodstuffs		McDonald *et al.*, 1999	98
96	^{210}Po	Seafood	Fish: 14.5; Crustaceans: 0.9 shellfish: 0.9	Desideri *et al.*, 2011	160
466	^{210}Po	Seafood	Fish: 73.5; Crustaceans: 3.6 Shellfish: 3.6	Desideri *et al.*, 2011	561
20	^{210}Po	Seafood	Fish: 8.5 Crustaceans and shells: 0.5	Pollard *et al.*, 1998	165
150 to 270	All	Seafood	Fish: 20 to 37 Shellfish: 4 to 6 Crustaceans: 4 to 8	Rollo *et al.*, 1992	176 and 582*
311	^{210}Po and ^{210}Pb	Seafood	Based on tonnage caught	Yamamoto *et al.*, 1994	173 and 575*
216	^{210}Po and ^{210}Pb	Seafood		Yamamoto *et al.*, 1994	173 and 575*
46 to 129**	^{210}Po	Seafood	Fish: 24; shellfish: 10; no crustaceans		165
240	^{210}Po	Fish	Fish: 25	Louw *et al.*, 2009	48
1000–1700	^{210}Po	Cereals	Cereals: 150 Activity from 6 to 10 Bq.kg^{-1}	Louw *et al.*, 2009	40

*considering rations of 20 kg.y^{-1} for fishes and 19 kg. y^{-1} of crustaceans and shellfish; mean values from the surveys on marine sites (Gravelines, Blayais and Nord-Cotentin).
**A correction factor of 0.6 has been applied to take into account a fishing-consumption delay as proposed by UNSCEAR. To be compared with other doses, the corrected values would be 77 and 214 μSv.y^{-1}.

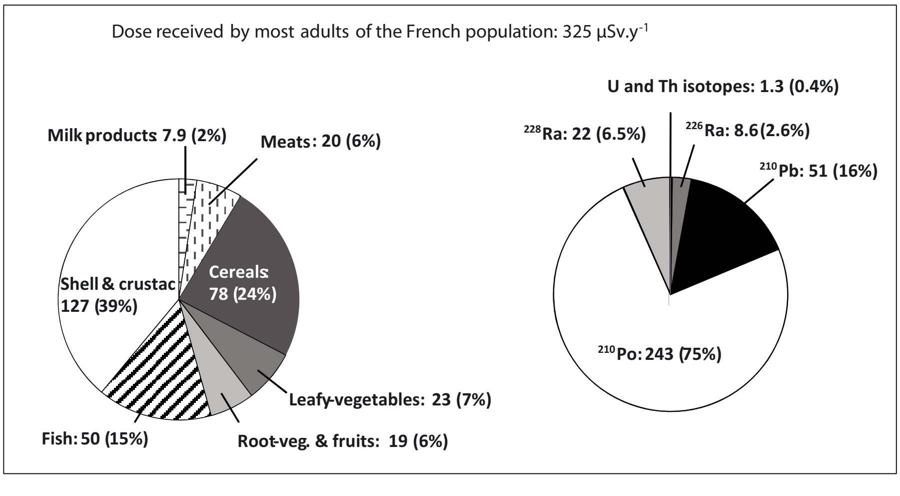

Fig. 1. Contributions of foodstuff categories and radionuclides to the dose received by most adults of the French population.

the slight gaps between the rations of other kinds of foodstuffs, even cereals, which exhibit the highest ^{210}Po activities behind seafood. Moreover, the studies carried out on polonium, lead and radium activities in French foodstuffs show that no distinctions can be made between sites regarding their content of uranium and thorium in soils.

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