Radioprotection 50(2), 111-115 (2015)

DOI: 10.1051/radiopro/2014035

Available online at:
www.radioprotection.org

ARTICLE

^{226}Ra and ^{228}Ra activities in French foodstuffs

Ph. Renaud*, S. Roussel-Debet, L. Pourcelot and R. Gurriaran

IRSN, BP3, 13108 Saint-Paul-Lez-Durance, France.

Received 15 July 2014 – Accepted 20 November 2014

Abstract – The activities of ^{228}Ac and ^{214}Bi in French foodstuffs acquired during the last 15 years allow one to obtain reference values of ^{228}Ra and ^{226}Ra activities which are usable to assess the dose to the French population due to the intake of these two natural radionuclides. As expected, because of similar ^{238}U and ^{232}Th activities in French soils (around 40 Bq.kg^{-1}), the ^{226}Ra/^{228}Ra activity ratio in French terrestrial foodstuffs is close to 1. Most kinds of foodstuffs present similar mean activities: from 0.1 to 0.2 Bq.kg^{-1} fresh for cereals, leafy vegetables, root vegetables, eggs and fishes (marine and freshwater). The activity in fruits is lower, around 0.05 Bq.kg^{-1}, similar to that of meat calculated on the basis of the numerous grass measurement results. The lowest activities are noticeable for milk: around 0.015 Bq.L^{-1}. All these values are in good agreement with the data from various countries, but most often 2 to 3 times higher than the reference values proposed by UNSCEAR. This study also shows that due to the large variability of foodstuff activities reported by a large amount of data, it is not possible to distinguish the activities of samples from regions with a gap of only a factor of 2 between their soil activities.

Keywords: radium-226 / radium-228 / foodstuffs

1 Introduction

Behind lead-210 and polonium-210, radium-228 and radium-226 belong to the main contributors to internal doses linked to the intake of natural radionuclides (UNSCEAR, 2000). The study carried out by Picat *et al.* (2002) showed that the French data were at this time insufficient to know activity levels of natural radionuclides in French foodstuffs, particularly as regards radium. For this reason, no more accurate dose assessments such as those performed by UNSCEAR on the global scale could be made for France. The French Nuclear Protection and Safety Institute regularly samples food produced over the entire metropolitan territory for the purposes of radiological environmental monitoring or research activities. The analyses are focused on artificial radionuclides but since the mid-90s, ^{228}Ac and ^{214}Bi, decay products of ^{228}Ra and ^{226}Ra, respectively, are regularly provided in the results of gamma spectrometry. The aim of the present study is to obtain mean values of radium activities which may have a good representativeness for the various kinds of foodstuffs produced over the whole territory and consumed by the population. The variability of radium activities in foodstuffs is compared with that of soils inside the French metropolitan territory (outside the influence of former mining facilities).

* philippe.renaud@irsn.fr

2 Materials and methods

Most of the available results (namely, leafy vegetables, cereals and pasture grass used to assess milk and meat activities) are from 8 to 32 French Districts (of a total number of 90 metropolitan Districts). Fishes and mussels are from various locations along the coasts of the Channel, the Atlantic and the Mediterranean coast. Concerning root vegetables, fruits, berries, eggs and milk, the number of results above the Detection Limits is small (2 to 8 results for each) and only gives an indication to situate French radium levels in the scarce literature data. All samples are directly bought from farmers. Plants are washed and the cuticles of cereal grains are already removed. The samples are dried and then incinerated at 480 °C by a controlled increase in the temperature to prevent inflammation. Ashes are placed in radon-tight counting containers from 17 to 60 mL (geometries). The measurements of ^{228}Ac and ^{214}Bi are then performed by the metrology laboratory of IRSN-Orsay. The materials and the methods used are detailed in de Vismes Ott *et al.* (2013). The counting time is always started after a minimal delay of 30 days to allow equilibrium between ^{226}Ra and ^{214}Bi. For leafy vegetables, cereals and grass, the results are numerous enough to compare averages and ranges of activities for samples from Districts with low uranium-thorium activity in soil (limestone regions) with those with higher activity (granitic regions). All activities are given in Bq.kg^{-1} of fresh weight, except for soils, which are in Bq.kg^{-1} dry matter.

3 Results

The averages of 1,800 analyses of ^{228}Ac and 1,200 analyses of ^{234}Pa in soil samples from 45 French Districts (of a total number of 90) distributed over the whole metropolitan territory are 42 Bq.kg^{-1} (ranging from 2 to 239 Bq.kg^{-1}) and 43 Bq.kg^{-1} (ranging from 4 to 400 Bq.kg^{-1}), respectively. Each set of data follows a normal distribution with median values close to the averages (40 Bq.kg^{-1} and 38 Bq.kg^{-1}, respectively) and standard deviations of 21 and 26 Bq.kg^{-1}, respectively. These parameters are necessarily influenced by the representativeness of the analyzed soils considering the different kinds of French soils. Among the 45 Districts from which data on soils are available, two sets of Districts were selected: a set of 17 Districts situated mainly on granitic rocks and with a mean ^{238}U activity of 52 ± 22 Bq.kg^{-1}, and a second set of 12 Districts mainly on limestone rocks and with a mean activity of 24 ± 21 Bq.kg^{-1}. These two sets will allow studying the potential influence of this gap in soil activities on the activities of foodstuffs from them.

In order to carry out a further comparison of results on foodstuff activities, similar ranges of U-Th activities in soils can be found in Germany and the United Kingdom (BfS, 2003; Jones *et al.*, 2009), and slightly lower activities in Italy (around 26 Bq.kg^{-1} according to De Bortoli and Gaglione, 1972), while levels in Romania and Poland (UNSCEAR, 2000), and especially in central Poland (7 to 24 Bq.kg^{-1}, Pietrzack-Flis *et al.*, 2001), are close to the lowest activities which can be found in France. Considering the highest values observed in France, Picat *et al.* (2002) indicate an average of 98 Bq.kg^{-1} for French soils located around former uranium mines but outside their influence.

Among 724 samples of leafy vegetables, 216 exhibited ^{228}Ac activities above the Detection Limit (DL). It was also the case for 54 samples out of 123 for ^{214}Bi activities. These similar activities of ^{226}Ra and ^{228}Ra were expected, due to the fact that ^{238}U and ^{232}Th present similar activities in French soils, and that the soil-to-plant transfer is the same for the two isotopes. Santos *et al.* (2002) explain that the higher ^{228}Ra/^{226}Ra activity ratio (about 3) in Brazilian leafy vegetables is the same in soils. The French average value for ^{226}Ra is situated in the middle of the literature values (Table 1), ranging from 0.05 to 0.2 Bq.kg^{-1}. In the case of ^{228}Ra, the French average is very close to those given by Asefi *et al.* (2005) and Choi *et al.* (2008). The reference values proposed by UNSCEAR (2000) (0.04 and 0.05 Bq.kg^{-1} for ^{228}Ra and ^{226}Ra, respectively) are two times lower than the French average and most of the others reported. It is not possible to distinguish ^{228}Ac activities in samples from low U-Th-background Districts, with an average value of 0.086 ± 0.02 Bq.kg^{-1}, from those of samples from higher U-Th background Districts, with a mean value of 0.097 ± 0.03 Bq.kg^{-1}. The high variability of the leafy vegetable activities probably masks the gap resulting from the difference of only a factor of 2 between the soil activities from these two kinds of regions. Blanco-Rodriguez *et al.* (2002) showed that the linearity of the relationship 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. When the gap between soil concentrations is too small, other variables affecting the soil-to-plant transfer are more significant (Simon and Ibrahim, 1987, 1988; Petterson *et al.*, 1988). In the same way and probably for the same reasons, one can note that the average values given in the various publications are quite close.

UNSCEAR proposes a reference value for other vegetables and fruits two times lower than that of leafy vegetables. The small set of French data on fruits seems to confirm this, but it is not the case for root vegetables, which present practically the same average activity as leafy vegetables. More generally, observing the bibliographic data, it appears that the root vegetable activities are not widely different from those of leafy vegetables. In the case of fruits, although BfS (2003) gives the same average (0.2 Bq.kg^{-1}) and similar ranges for the three categories, the fact that fruits may have activities in the order of 2 times lower seems to be true, notably in the case of Korean data. The data on berries are even rarer. However, the French data confirm observations from Poland and Germany, where activities in berries seem to be clearly higher than those of fruits of trees. Finally, according to Santos *et al.* (2002), tropical fruits such as banana, papaya and pineapple consumed in France do not present any difference in their radium contents compared with oranges or apples. With the exception of berries, grains of cereals and legumes present the highest radium activities. The mean value of French wheat grains (0.18 Bq.kg^{-1}) is in the middle of the averages from the literature data (from 0.1 to 0.3 Bq.kg^{-1}) and the ranges are similar. Note that the reference values proposed by UNSCEAR are 2 to 3 times lower.

Only 8 and 2 results on French milk are available for ^{214}Pb (^{226}Ra) and ^{228}Ac (^{228}Ra), respectively (Table 2). The mean activity of ^{226}Ra is in good agreement with the literature data. The reference value proposed by UNSCEAR is much lower for the two isotopes of radium (0.005 Bq.L^{-1}). Concerning meat, no French data is available because the activity of all samples was below the detection limit. An alternative approach is to estimate ^{228}Ra and ^{226}Ra activities in milk and meat on the basis of activities measured in grass using transfer factors to animal products. Among 540 analyses of cultivated grass, 340 allow quantifying ^{228}Ac activity. For ^{214}Bi, it is the case for 102 samples of a total amount of 138. The averages are 0.50 ± 0.04 Bq.kg^{-1} and 0.59 ± 0.08 Bq.kg^{-1} for ^{228}Ac and ^{214}Bi, respectively. The transfer factors for radium proposed by the IAEA (2010) are 3.8×10^{-4} Bq.L^{-1} per Bq.d$^{-1,}$ ranging from 9×10^{-5} to 1.4×10^{-3} for milk and a single value of 1.7×10^{-3} Bq.L^{-1} per Bq.d^{-1} for beef. Considering a consumption of 50 kg.d^{-1} of fresh grass, the mean and the ranges given in Table 2 are rather consistent with the measurement results for milk and give an indication of what the level of radium is in French beef. A recent study (Jeambrun *et al.*, 2012) provided some data on radium in eggs which are fairly consistent with the rare literature data available.

Data on radium in seafood are very scarce and even the bibliographic syntheses performed by Stewart *et al.* (2008) and Young *et al.* (2002) report very little data on the levels of ^{226}Ra, and none for ^{228}Ra. UNSCEAR has also not been able to provide a reference value for this last radionuclide in fish. The results published by Choi *et al.* (2008) show that the activities of the two isotopes of radium are similar in different species of fish and shellfish studied. They also provide the main elements

Table 1. French data and bibliographic data on ^{226}Ra and ^{228}Ra activities in vegetable foodstuffs: mean, (range) in Bq.kg^{-1} fresh and [*number of results*].

	^{228}Ra (Bq.kg^{-1} fresh matter)	**^{226}Ra (Bq.kg^{-1} fresh matter)**	**References**
Leafy vegetables			
France	0.09 (0.020–1.97) *[216]*	0.09 (0.018–1.22) *[54]*	This study
Germany (cabbage)		0.2 (0.01- 0.68)	BfS, 2003
UK		0.05 (0.002–0.440) *[60]*	RIFE, 1996–2010
Poland		(0.010–0.137)	Piertrzack-Flis, 2001
Iran	0.10 (0.03–0.20) *[12]*	0.08 (<0.12–0.12) *[9]*	Asefi *et al.*, 2005
Korea (spinach)	0.15 *[21]*	0.13 *[21]*	Choi *et al.*, 2008
Brazil	0.22 (<0.022–2.37) *[26]*	0.075 (<0.01–0.81) *[26]*	Santos *et al.*, 2002
UNSCEAR reference values and European ranges	0.04	0.05 (0.002–1.15)	UNSCEAR, 2000
Root vegetables			
France	0.12 (0.046–0.44) *[6]*	No Data available	This study
Germany		0.2 (0.02–1.3)	BfS, 2003
UK		0.032 (0.0015–0.15) *[51]*	RIFE, 1996–2010
Poland		(0.01–0.059)	Pietrzack-Flis *et al.*, 2001
Iran	0.052 (<0.018–0.096) *[20]*	0.031 (0.013–0.062) *[23]*	Asefi *et al.*, 2005
Brazil	0.27 (0.009–0.93) *[27]*	0.071 (0.0066–0.653) *[27]*	Santos *et al.*, 2002
Korea	0.024 *[33]*	0.018 *[33]*	Choi *et al.*, 2008
Fruit vegetables			
Italy (mix leaf.& fruits veg.		0.036 (0.027–0.044)	De Bortoli and Gaglione, 1972
Poland		(0.0088–0.059)	Pietrzack-Flis *et al.*, 2001
Iran	0.045 (0.039–0.051) *[3]*		Asefi *et al.*, 2005
Brazil	0.11 (0.008–0.928*) [9]*	0.084 (<0.0034–0.57) *[9]*	Santos *et al.*, 2002
Fruits (from trees)			
France	0.042 (0.018–0.074) *[7]*	0.057 (0.074–0.31) *[5]*	This study
Germany		0.2 (0.005–2.29)	BfS, 2003
Italy		0.014	De Bortoli and Gaglione, 1972
Poland		(0.012–0.15)	Pietrzack-Flis *et al.*, 2001
Brazil	0.058 (0.01–0.31) *[9]*	0.028 (0.01–0.1) *[9]*	Santos *et al.*, 2002
Korea	0.0096 *[33]*	0.0090 *[33]*	Choi *et al.*, 2008
UNSCEAR reference values and European ranges	0.02 (–)	0.03 (0.009–9.4)	UNSCEAR, 2000
Berries			
France	0.32 (0.1–0.73) *[7]*	No Data available	This study
Germany		2.2 (0.03–5.38)	BfS, 2003
Poland		(0.022–0.047)	Pietrzak-Flis *et al.*, 2001
Cereals			
France	0.18 (0.047–5.55) *[137]*	0.18 (0.056–1.5) *[49]*	This study
Germany		0.3 (0.04–1.54)	BfS, 2003
Italy		0.14	De Bortoli & Gaglione, 1972
Poland (flour)		(0.048–0.062)	Pietrzack-Flis *et al.*, 2001
Iran	0.10 (0.071–0.71) *[9]*	0.19 (0.04–0.45) *[9]*	Asefi *et al.*, 2005
Korea	0.17 *[38]*	0.12 *[38]*	Choi *et al.*, 2008
UNSCEAR reference values	0.06	0.08 (0.0007–5.2)	UNSCEAR, 2000

Table 2. French data and bibliographic data on ^{226}Ra and ^{228}Ra activities in animal products: mean, (range) in Bq.kg^{-1} fresh and [*number of results*].

	^{228}Ra (Bq.kg^{-1})	**^{226}Ra (Bq.kg^{-1})**	**References**
Milk			
France	(0.037–0.047) *[2]*	0.017 (0.007–0.059) *[8]*	This study
France	0.010 (0.0023–0.035)	0.011 (0.0026–0.041)	This study (estimated value)
Germany		0.025 (0.001–0.13)	BfS, 2003
Italy		0.007	De Bortoli and Gaglione, 1972
Poland		0.010 ± 0.0013	Pietrzack-Flis *et al.*, 1997
Korea	0.041 ± 0.014	0.022 ± 0.07 *[26]*	Choi *et al.*, 2008
Brazil	0.030 (0.004–0.050) *[20]*	0.015 (0.0022–0.027) *[20]*	Santos *et al.*, 2002
UNSCEAR reference values and European ranges	0.005	0.005 (0.0009–0.2)	UNSCEAR, 2000
Meat			
France	0.043	0.050	This study (estimated value)
Germany		0.1 (0.03–7.8)	BfS, 2003
Poland		(0.0098–0.0196)	Pietrzack-Flis *et al.*, 2001
Iran		0.08 (0.064–0.10) *[3]*	Asefi *et al.*, 2005
UNSCEAR reference values and European ranges	0.010	0.015 (0.002–0.22)	UNSCEAR, 2000
Eggs			
France	0.087 (0.07–0.11) *[3]*	0.15 (0.013– 0.017) *[5]*	Jeambrun *et al.*, 2012
Italy		0.24	De Bortoli and Gaglione, 1972
Poland		0.0996 ± 0.0053	Pietrzack-Flis *et al.*, 2001
Iran	0.078 (0.072–0.084) *[2]*	0.063 (0.040–0.10) *[2]*	Asefi *et al.*, 2005
Freshwater fish			
France	0.093 (0.037–0.14) *[8]*	0.13 (0.015–0.48) *[8]*	This study
Italy (from lakes)		0.08 (0.052–0.12)	De Bortoli and Gaglione, 1972
Fish (marine)			
France	0.16 (0.05–0.7) *[130]*	No data available	This study
Europe		0.1 (0.1–7.4)	UNSCEAR, 2000
Germany		1.5 (0.05–7.8)	BfS, 2003
UK		0.02 (<0.001–0.037)	Young *et al.*, 2002
UK		(0.007–0.19)	Stewart *et al.*, 2008
Poland		(0.0126–0.0196)	Pietrzack-Flis *et al.*, 2001
Korea	0.092 *[28]*	0.16 *[28]*	Choi *et al.*, 2008
Shellfish			
France	0.13 (0.017–0.58) *[154]*	0.4 (0.14–0.52) *[60]*	This study
UK		(0.094–1.3)	Young *et al.*, 2002
Korea	0.22 *[25]*	0.18 *[25]*	Choi *et al.*, 2008

of comparison with the data of this study. The number of measured ^{228}Ac activities in samples of fish and mussels from the French Mediterranean coast, which are relatively large (131 and 154, respectively), are thus particularly interesting. The range of measured ^{228}Ac values, from 0.056 to 0.7 Bq.kg^{-1}, and the average of 0.16 Bq.kg$^{-1,}$ are rather close to those observed in Korean samples. The activities of ^{214}Bi and ^{228}Ac measured in mussels from the French Mediterranean coast are also in fairly good agreement with the Korean data.

4 Conclusion

The data on ^{228}Ac and ^{214}Bi activities in French foodstuffs acquired during the last 15 years allow one to obtain reference values of ^{228}Ra and ^{226}Ra activities usable to assess the dose to the French population due to the intake of these two natural radionuclides.

Similar activities of ^{226}Ra and ^{228}Ra expected in terrestrial foodstuffs are due to the equilibrium in soils with their respective fathers ^{238}U and ^{232}Th, which present similar activities in French soils. Despite the absence of equilibrium and the gap between thorium and uranium activities in water, the data provided by this study seem to show that activities of the two radium isotopes in fishes and mussels are not so different. French data as well as those from the literature show that mean values are similar whatever the kinds of foodstuffs or the country (mainly between 0.05 and 0.1 Bq.kg^{-1} fresh), except in milk, which presents significantly lower activity (in the order of 0.015 Bq.L^{-1}) and berries, which exhibit the highest

(0.3 Bq.kg^{-1}). This study confirms 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, as is the case for most of the agricultural land in France, due to the strong variability of transfer of radium to plants and animal products. Therefore, the mean values obtained in the present study are representative of the entire French metropolitan territory.

References

Asefi M., Fathivand A.A., Amidi A., Najafi A. (2005) Determination of ^{226}Ra and ^{228}Ra concentrations in foodstuffs consumed by inhabitants of Tehran city of Iran, *Iran. J. Radiat. Res.* **3** (3), 149-151.

Blanco-Rodriguez P., Vera-Tomé F., Lozano J.C. (2002) About the assumption of linearity in the soil-to-plant transfer factors for uranium and thorium isotopes and ^{226}Ra, *Sci. Total Environ.* **284**, 167-175.

BfS (Bundesamt für Strahlenschutz) (2003) Umweltpolitiek. Umweltradioaktivität und Strahlenbelastung. Jahresbericht 2002. Bonn: Das Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit.

Choi M.S., Lin X.J., Lee S.A., Kim W., Kang H.D., Doh S.H., Kim D.S., Lee D.M. (2008) Daily Intakes of naturally occurring radioisotopes in typical Korean foods, *J. Environ. Radioact.* **99**, 1319-1323.

De Bortoli M., Gaglione P. (1972) Radium-226 in the environmental materials and foods, *Health Phys.* **22**, 43-48.

De Vismes Ott A., Gurriaran R., Cagnat X., Masson O. (2013) Fission products activity ratios measured at trace level over France during the Fukushima accident, *J. Environ. Radioact.* **125**, 6-16.

IAEA (2010) International Atomic Energy Agency. *Handbook of parameter values for the prediction of radionuclide transfer in terrestrial and freshwater environment*. Technical report No. 472. IAEA, Vienne, Austria.

Jeambrun M., Pourcelot L., Mercat C., Boulet B., Loyen J., Cagnat X., Gauthier-Lafaye F. (2012) Study on transfers of uranium, thorium and decay products from grain, water and soil to chicken meat and egg contents, *J. Environ. Monit.* **14**, 2170-2180.

Jones D.G., Copplestone D., Breward N., MacKenzie A.C., Scheib C., Beresford N.A., Barnett C.L., Wood D. (2009) Assessment of naturally occurring radionuclides around England and Wales: application of the G-BASE dataset to estimate doses to non-human species [electronic version]. British Geological Survey, Centre for Ecology and Hydrology, University of Liverpool. Great Britain Environment Agency. EDP Sciences, Les Ulis.

Petterson H.B.L., Hallstadius L., Hedvall R., Holm E. (1988) Radioecology in the vicinity of prospected uranium mining sites in the subarctic environment, *J. Environ. Radioact.* **6**, 25-40.

Picat P., Calmet D., Louvat D., Vray F., Lemaitre N., Linden G., Barbey P., Pigree G., Bourcier T., Levy F., Le Bar S., Baron Y., Delacroix D., Panaïva E., De Bruyne T., Hervé J.Y., Beguinel P., Cabanne N., De Paepe A., Santucci C., Prot T., Matrat J.L., Tillie J.L. (2002) Radioactivité d'origine naturelle dans l'environnement en France : niveaux non perturbés par l'homme, *Radioprotection* **37** (3), 283-327.

Pietrzack-Flis Z., Suplinska M.M., Rosiak L. (1997) The dietary intake of ^{238}U, ^{234}U, ^{230}Th, ^{232}Th, ^{228}Th and ^{226}Ra from food and drinking water by inhabitants of the Walbrzyck region, *J. Radioanal. Nucl. Chem.* **222** (1-2), 189-193.

Pietrzack-Flis Z., Rosiak L., Suplinska M.M., Chrzanowski E., Dembiska S. (2001) Daily intakes of ^{238}U, ^{234}U, ^{232}Th, ^{230}Th, ^{228}Th and ^{226}Ra in the adult population of central Poland, *Sci. Total Environ.* **273**, 163-169.

RIFE, Radioactivity in food and in the Environment, (1996–2010) United Kingdom Food Standards Agency, Northern Ireland Environmental Agency and Scottish Environmental Protection Agency. Reports from 1996 to 2000 and from 2008 to 2010.

Santos E.E., Lauria D.C. Amaral E.C.S., Rochedo E.R. (2002) Daily ingestion of ^{232}Th, ^{238}U, ^{226}Ra, ^{228}Ra and ^{210}Pb in vegetables by inhabitants of Rio de Janeiro city, *J. Environ. Radioact.* **62**, 75-86.

Simon S.L., Ibrahim S.A. (1987) The soil/plant concentration ratio for calcium, radium, lead and polonium: evidence for non-linearity with reference to substrate concentration, *J. Environ. Radioact.* **5**, 123-142.

Simon S.L., Ibrahim S.A. (1988) Biological uptake of radium by terrestrial plants: a review in *Radium in the Environment*, monography of the Atomic Energy Agency, Vienna, Austria.

Stewart G.M., Fowler S.W., Fisher N.S. (2008) The bioaccumulation of U- and T-series radionuclides in marine organisms. In: *Radioactivity in the Environment*, Vol. 13, Chap. 8. Elsevier.

UNSCEAR (2000) United Nations Scientific Committee on the Effects of Atomic Radiation. Source and effects of ionizing radiation. Report of the General Assembly, Vol. 1 with scientific annexes.

Young A.K., McCubbin D., Camplin W.C. (2002) Natural radionuclides in seafood. Center for Environment, Fisheries and Aquaculture Science, Lowesttoft Laboratory. Environment Report RL 17/02.

Cite this article as: Ph. Renaud, S. Roussel-Debet, L. Pourcelot, R. Gurriaran. ^{226}Ra and ^{228}Ra activities in French foodstuffs. Radioprotection 50(2), 111-115 (2015).