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Article

Evaluation of ^{14}C doses since the end of the 1950s in metropolitan France

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ABSTRACT **The dosimetric consequences of ^{14}C dissemination resulting from the radioactive fall-out of atmospheric nuclear-weapons testing conducted in the 50s have been estimated. Owing to the lack of ^{14}C measurements in food consumed in France over the past 60 years, this evaluation is based on the standard modelling of the isotopic equilibrium: it is assumed that the specific activity ($^{14}C/C$) of living plants is in equilibrium with that of CO_2 in the air. It is also considered that this isotopic ratio remains constant in animals and their production. The specific activities of ^{14}C in the biosphere were drawn from reference publications. Since the 1950s, the effective dose for adults has risen from 12.1 µSv y^{-1} to a maximum of about 22.3 µSv y^{-1} in 1964, before falling to 12.9 µSv y^{-1}, which is slightly higher than the initial value. The excess dose due to atmospheric testing reached about 10 µSv y^{-1} in the sixties. The generation of 1940 was exposed to the highest dose: 190 µSv in 60 years due to nuclear-weapons testing, which remains low in relation to natural exposure.**

Keywords: ^{14}C / atmospheric testing / effective doses

RÉSUMÉ **Évaluation des doses dues au ^{14}C, depuis la fin des années 1950, en France métropolitaine.**

L'estimation des conséquences dosimétriques, en France, de la dissémination du ^{14}C provenant des retombées des essais aériens d'armes depuis les années 50 est réalisée. En l'absence de chroniques de mesures du ^{14}C dans les aliments consommées en France durant les 60 dernières années, cette évaluation repose sur la modélisation usuelle de l'équilibre isotopique : on suppose que l'activité spécifique ($^{14}C/C$) des végétaux vivants est en équilibre avec celle du CO_2 de l'air. On considère aussi que ce rapport isotopique reste constant dans les animaux et leurs productions. Les valeurs de l'activité spécifique du ^{14}C dans la biosphère sont issues de la bibliographie. Pour l'adulte, la dose efficace est passée depuis les années 1950 de 12,1 µSv an^{-1} à un maximum de l'ordre de 22,3 µSv an^{-1} en 1964 pour redescendre ensuite à un niveau de 12,9 µSv an^{-1}, très légèrement supérieur à sa valeur initiale. La contribution des retombées des essais nucléaires à cette dose a donc atteint durant les années 60 une dizaine de µSv an^{-1}. La génération 1940 a reçu la dose la plus élevée : 190 µSv en 60 ans imputables aux tirs, ce qui reste très faible par rapport à l'exposition naturelle.

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Introduction

Carbon has two stable isotopes: carbon-12 and carbon-13, which have respective abundances of 98.93% and 1.07%. ^{14}C is a β-emitter with a half-life of 5 730 ± 40 years and is only found in infinitesimal quantities. ^{14}C does not cause external irradiation. Internal irradiation of the human body is mainly due to the ingestion of carbon found in food. ^{14}C is naturally produced in the upper atmosphere from nitrogen-14 at the rate of about 1.54 PBq y^{-1} for an estimated worldwide inventory of 12 700 PBq. Another 213 PBq resulting from atmospheric nuclear tests is to be added to this natural radiocarbon. Furthermore, nuclear power plants and reprocessing plants released a total of about 2.8 PBq of ^{14}C between the 1950s and 1997 (UNSCEAR, 2000). The radiological consequences of such fall-out in France have not been evaluated, whereas several series of measurements have been performed in Europe for about ten years now, making it possible to determine the variations in ^{14}C found in plants, particularly by measuring tree growth rings.

1. Modelling carbon-14 transfers and exposure

1.1. Modelling principles

The evaluation of ^{14}C transfers between two biosphere compartments is based on the consensual hypothesis that:

– atmospheric radiocarbon is found as $^{14}CO_2$, with most releases assumed to be in this form and with the oxidation of other carbon compounds producing $^{14}CO_2$ over a period of several years (UNSCEAR, 2000);
– the specific activity of carbon in plants (becquerels of ^{14}C per unit mass of carbon, Bq kg^{-1}) is the same as that in the atmosphere (incorporation by photosynthesis);
– this isotopic equilibrium is retained in all compartments of the food chain.

All explicit or aggregated models (*e.g.*: Le Dizès, 2004; IAEA, 2001, 1985, 1982; Sheppard *et al*., 1994; OMS, 1987) rely on this basic assumption, expressed as:

$$\frac{^{14}C_{organism}}{C_{organism}} = \frac{^{14}C_{medium}}{C_{medium}} \quad (1)$$

where $^{14}C_{organism}$ is the ^{14}C concentration in the organism (Bq kg^{-1}), $C_{organism}$ is the C concentration in the organism (kg C kg^{-1}), $^{14}C_{medium}$ is the ^{14}C concentration in the medium (Bq kg^{-1} or Bq m^{-3}), C_{medium} is the C concentration in the medium (kg C kg^{-1} or kg C m^{-3}).

Such modelling disregards isotopic discrimination that occurs during photosynthesis, which over-evaluates by about 5%, except when specific activity measurements of ^{14}C have been standardised thanks to the simultaneous measurement of ^{13}C (see Appendix), which has become the rule over the past ten years or so.

1.2. Specific activity of ^{14}C in the terrestrial environment

1.2.1. Values from literature

^{14}C measurements in the atmospheric compartment are both numerous and widely published. In Europe for example, Nydal and Löyseth (1983) complied a set of atmospheric measurements (1962–1980) recorded by their laboratory from 7 different stations including Lindesnes in Norway (altitude of 50 m) and Santiago de Compostela in Spain (altitude of 360 m). Research by Levin *et al*. (1985) also summarises several series of measurements (1959–1983) recorded in the troposphere, particularly at the Vermunt station in Austria (altitude of 1800 m), with the exhaustive records being available on the internet (Levin *et al*., 1994). Furthermore, it is widely accepted that the specific activity of carbon in plants directly reflects that of atmospheric carbon (*e.g*.: Daillant *et al*., 2004; McGee *et al*., 2004; Isogai *et al*., 2002; Muraki *et al*., 2001; Mc Namara and Mc Cartney, 1998). The main causes of discrepancy in the specific activities of plants and air were examined in detail by Hua *et al*. (1999) based on a state-of-the-art review of measurement results from the 1945–1997 period recorded in both the atmosphere and tree growth rings. According to the authors, the main causes of abnormalities are as follows:

– trees use carbohydrates that date prior to the year of measurement,
– samples taken from the lower part of the canopy (effect of CO_2 releases from the ground),
– bias introduced during sampling and treatment prior to measurements.

Apart from these very few exceptions, the series of ^{14}C measurements reveal remarkable agreement between plants and the air. Various references therefore make it possible to estimate variations in the specific activity in relation to time in different areas around the world and especially in France. Values in publications are either given as a specific activity, or as a percent of modern carbon (PMC) or in terms of DELTA ^{14}C ($\Delta^{14}C$). Conversions were performed according to formulas recalled in the appendix in order to express all data in the form of a specific activity. A certain number of these values are represented in Figure 1 based on measurements reported in the following research papers:

– Roussel-Debet *et al*. (2006) – leaves of trees or shrubs and agricultural plants, 1994–2003, France;

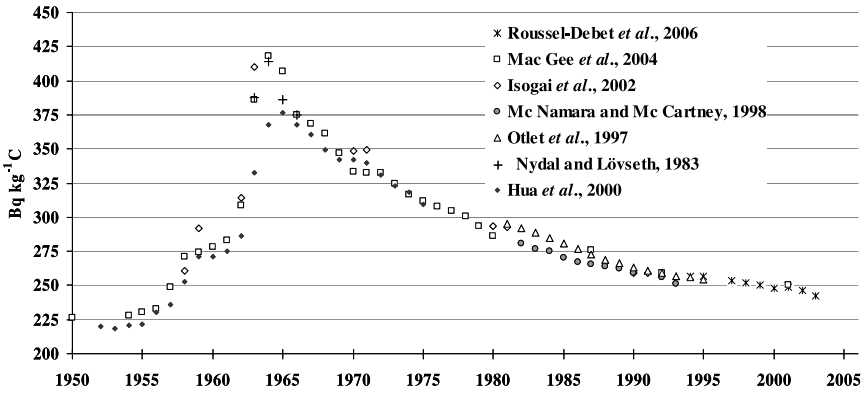

Figure 1 – ***Specific activity of ^{14}C in the biosphere recorded from 1950 to today. All data corresponds to the northern hemisphere, excepting that of Hua et al. (2000) (mean values recorded in Tasmania and Thailand).***

Activité spécifique du ^{14}C dans la biosphère de 1950 à nos jours. Les données figurées sont relatives à l'hémisphère nord, sauf celles de Hua et al. (2000) (moyenne Tasmanie et Thaïlande).

- McGee *et al.* (2004) – tree rings from a specimen of *Pinus Sylvestris*, 1954–2001, Ireland;
- Isogai *et al.* (2002) – rings from a sycamore tree, 1958–1991, England;
- Mc Namara and Mc Cartney (1998) – tree rings from a larch tree, 1950–1993, England;
- Otlet *et al*. (1997) – foodstuffs, 1981–1995, England;
- Hua *et al.* (2000) – tree rings from pine species, 1952–1975, Thailand and Tasmania.

For comparative reasons, the results of atmospheric carbon-14 measurements recorded by Nydal and Lövseth (1983) between 1963 and 1966 in Norway and Spain are plotted in this figure. It can be observed that they match the bundle of curves for plants. Furthermore, the presentation of yearly data must not mask the seasonal variability in the activity of ^{14}C, which is plotted in Figure 2 for illustrative purposes. This is especially due to seasonal movements that influence both interactions between the stratosphere and troposphere and air mass transfers, as well as seasonal variations in CO_2 capture and release phenomena by the biosphere (Hua *et al*., 1999). The latitudinal variability – plotted in Figure 3 – was maximal during the years of 1962–63 when the magnitude and number of the atmospheric nuclear detonations were particularly high in the northern hemisphere. Lastly, it is worth pointing out that measurement uncertainties are small, being equivalent to about 1 to 1.5%.

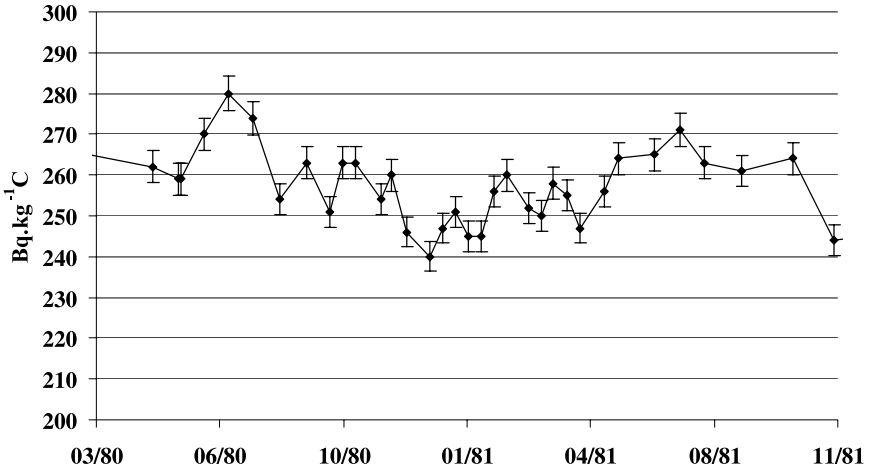

Figure 2 – ***Seasonal variability in the specific activity of tropospheric ^{14}C recorded in Vermunt, Austria (from Levin et al., 1994).***

Exemple de variabilité saisonnière de l'activité spécifique du ^{14}C troposphérique observée à Vermunt, Autriche (d'après Levin et al., 1994).

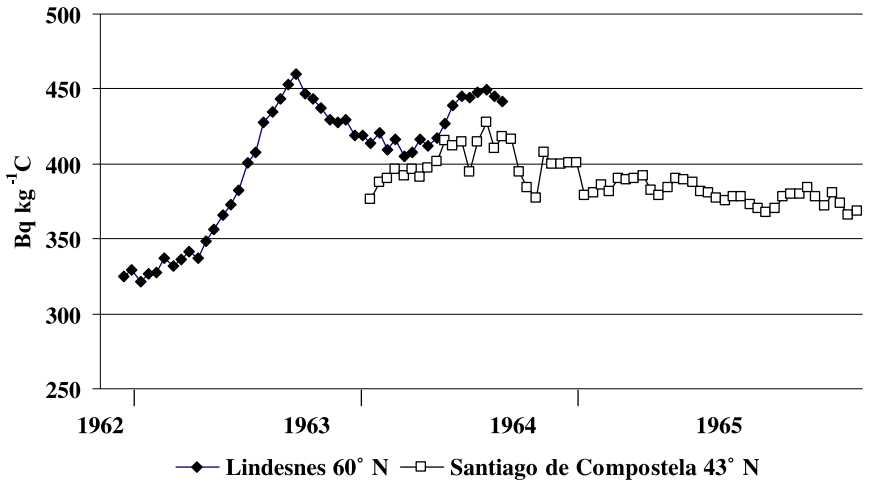

Figure 3 – ***Effect of latitude on the specific activity of atmospheric ^{14}C (from Nydal and Lövseth, 1983).***

Illustration de l'influence de la latitude sur l'activité spécifique du ^{14}C atmosphérique (d'après Nydal et Lövseth, 1983).

In the northern hemisphere, the specific activity of ^{14}C increases irregularly from the reference value A_0 = 226 Bq kg^{-1} in 1950, the base year for "modern" carbon (Stuiver and Polach, 1977) up to a maximum value of about 420 Bq kg^{-1} in 1964. After this date, the specific activity of ^{14}C decreases until reaching about 240 Bq kg^{-1} in 2003, which is still higher than its initial value. This increase of about a factor of 2 in the early 60's and the decrease subsequent to the 1963-Treaty, which banned nuclear tests in the atmosphere has been illustrated by Jean-Baptiste and Paterne (2003). Variations are similar in the southern hemisphere, however the initial level (1950s) and maximum value of about 376 Bq kg^{-1} in 1965 are slightly lower than those recorded in the northern hemisphere.

1.2.2. Values used for dose evaluations

The data used to recreate yearly doses in France are arithmetic means per year of values plotted in Figure 1; the results obtained by Hua *et al.* (2000) for the southern hemisphere were obviously not taken into account. These values are reported in Table I. The reference value of 226 Bq kg^{-1} in 1950 was used as a basis to calculate "excess" activity due to nuclear tests. This represents an approximation that is probably slightly under-evaluated owing to the constant increase of CO_2 in the atmosphere, which is naturally depleted in ^{14}C because of our use of fossil fuels (see Prentice *et al.*, 2001). Moreover, the selected values take into account the production of ^{14}C from nuclear-related plants, which represent about 1% of the activity emitted by nuclear-weapons testing over the past 50 years. Owing to the complexity of the carbon cycle on a global scale, it is not possible to obtain, in a simpler way, a better evaluation of the excess activity due to nuclear-weapons test fall-out.

1.3. Means of exposure – calculation method

Carbon-14 does not cause external irradiation (low-energy β-emitter). In terms of internal irradiation, carbon is mainly found in the human body due to ingestion (ICRP, 2003). Consequently, evaluations only take into account transfer paths related to the food chain. The dose related to inhalation will nevertheless be estimated for comparative purposes. Given the physical half-life of carbon-14, it is unnecessary to consider neither its radioactive decay nor a storage delay for foodstuffs. Additionally, the daughter element is stable, which means that irradiation *via* daughter products is not to be taken into account. For atmospheric carbon-14, the dose calculation models assume the conservation of the isotopic ratio – from the atmosphere to the human body – either in an aggregated manner (total ingestion of carbon) or *via* a step-by-step evaluation of the specific activity of carbon-14 in foodstuffs, followed by ingestion. The specific activity of the

TABLE I

^{14}C specific activity considered for dose evaluation (Bq kg^{-1}).
Activité spécifique du ^{14}C prise en compte pour les évaluations (Bq kg^{-1}).

Année	Total* Bq kg^{-1}	Excess ** Bq kg^{-1}	Année	Total Bq kg^{-1}	Excess Bq kg^{-1}	Année	Total Bq kg^{-1}	Excess Bq kg^{-1}
1950	226	0	1970	341	115	1987	275	49
1954	228	2	1971	341	115	1988	269	43
1955	231	5	1972	332	106	1989	266	40
1956	233	7	1973	324	98	1990	261	35
1957	249	23	1974	316	90	1991	260	34
1958	266	40	1975	312	86	1992	259	33
1959	283	57	1976	308	82	1993	258	32
1960	278	52	1977	305	79	1994	257	31
1961	283	57	1978	301	75	1995	256	30
1962	311	85	1979	294	68	1996	255	29
1963	398	172	1980	290	64	1997	254	28
1964	418	192	1981	294	68	1998	252	26
1965	407	181	1982	292	66	1999	251	25
1966	375	149	1983	289	63	2000	248	22
1967	368	142	1984	285	59	2001	249	23
1968	362	136	1985	281	55	2002	247	21
1969	347	121	1986	277	51	2003	242	16

** Total: due to natural and man made ^{14}C; **Excess: due to man made ^{14}C (mainly produced in atmospheric testing).*

atmosphere is considered as constant per calendar year, seeing that measurements allowing for a more refined time step are not available. In addition, the specific activity of the atmosphere will be considered as homogeneous on a European level.

1.3.1. Calculation equations and parameters for ingested doses

The most detailed type of model refers to quantities of carbon ingested by humans via different products composing their diet (*e.g.*: Le Dizès, 2004; Simmonds *et al*., 1995; Zach and Scheppard, 1992; US-NRC, 1977). This formula requires evaluating the specific activity of carbon-14 in each product, which makes it possible to better integrate specificities of food rations. The effective dose by ingestion is expressed according to equation (2), whereas the specific activities of

TABLE II

Other parameters values.
Valeurs des autres paramètres.

Age class		0–1 year	1–2 years	2–7 years	7–12 years	adult
Effective dose factor $FD_{ing,a}$ (Sv Bq^{-1})		1.4×10^{-9}	1.6×10^{-9}	9.9×10^{-10}	8.0×10^{-10}	5.7×10^{-10}
Foods	$f_{c,i}$ (kg **C** kg^{-1})	Mean annual ingestion rate (kg y^{-1})				
Leaves	0.050		3	26	29	40
Fruits	0.050		40	73	84	113
Roots	0.095		13	13	15	26
Potatoes	0.360		18	22	33	29
Cereals	0.950		9	51	73	73
*Milk products**	0.067	292	146	204	175	146
*Meat***	0.265		11	51	69	84
Sugar	0.438		15	28	31	33
Vegetal oil	0.750		4	7	7	8
Total ingested carbon (kg C y^{-1})		20	31	71	85	92

** Milk equivalent, ** beef equivalent.*

plant or animal products are calculated according to equation (3):

$$D_a = FD_{ing,a} \times \sum_i R_{i,a} \times {}^{14}C_i \quad (2)$$

$${}^{14}C_i = f_{c,i} \times A_{air} \quad (3)$$

where: D_a is the age-dependent effective dose rate from intakes by ingestion (Sv y^{-1}), $FD_{ing,a}$ is the effective dose coefficient for ingestion for age group a (Sv Bq^{-1}), $R_{i,a}$ is the annual food intake (kg $_{FW}$ y^{-1}), ${}^{14}C_i$ is the ${}^{14}C$ concentration in the product i (Bq $kg^{-1}{}_{FW}$), $f_{c,i}$ is the proportion of stable carbon in the product (kg C $kg^{-1}{}_{FW}$), A_{air} is the atmospheric concentration of stable carbon in the form of CO_2 (Bq kg^{-1} C), where $A_{air} = {}^{14}C_{air}/{}^{12}C_{air}$.

Dose factors and parameters related to the food ration and the carbon content in foodstuffs are reported in Table II. Effective dose factors $FD_{ing,a}$ were provided by Euratom (1996) from the dose factors of the International Commission on Radiological Protection (ICRP) for an integration time of 50 years for adults and 70 years for other age classes. The food ration $R_{i,a}$ developed by Vray and Renaud (2005) was used, but fresh and preserved products of the same food type were grouped into the same category, seeing that it is unnecessary to take into account radioactive decay for ${}^{14}C$. This ration is related to the gross weight of raw food (unpeeled, unprepared, etc.), which does not alter the specific activity of ${}^{14}C$ in

terms of concentration. Furthermore, considering the important consumption of "sugar" and "vegetable oil" categories with regard to the incorporation of carbon via foodstuffs, these products were added into the ration described by Vray and Renaud (2005). It is assumed that these products are contaminated with ^{14}C regardless of their origin (metropolitan or elsewhere), which is justified by the dispersion of ^{14}C on a global scale. The ration of these products was drawn from a bibliographic review by Roussel-Debet *et al*. (2002) for adults. The teenagers (12–17 years) are considered as adults for their food rations, which agrees reasonably with the investigation of Volatier (2000), who considers that individuals over 14 years have the same ingestion rate than adults; for younger age groups, the adult ration was distributed in proportion to the total mass of ingested food, excepting breast-fed children for which these two products were not taken into consideration. Lastly, the ration does not take into account the ingestion of both fresh-water and salt-water fish for which the contamination in atmospheric ^{14}C can be assumed to be negligible vis-à-vis that of farm products. The carbon fractions $f_{c,i}$ of foodstuffs were drawn from the bibliographic review by Garnier-Laplace *et al*. (1994). The combination of these last two parameters makes it possible to evaluate the total mass of carbon ingested by an individual The total daily ingestion of carbon for an adult is equal to 251 g d^{-1}. This value is less than the 300 g d^{-1} specified by default for "standard" adults (ICRP, 2003), but corresponds to the value of 255 g d^{-1} of carbon announced by Charles and Jones (2005) during the evaluation using the PC-CREAM computer model belonging to the Health Protection Agency (HPA) in Great Britain. Logically, the most carbon-rich products that are the most consumed contribute the most to the incorporation of carbon. For an adult for example, the total of sugar + cereals + meat represents 60% of the ingested carbon.

1.3.2. Simplified formula for ingested dose calculations

For comparative reasons, ingested doses for adults will also be evaluated using the simplified formula provided by the International Atomic Energy Agency, IAEA (2001):

$$D_a = g_{ing} \times A \times f \tag{4}$$

where: D_a is the effective dose rate (Sv y^{-1}), g_{ing} is the dose rate factor that relates the annual dose rate to the specific activity of ^{14}C in people. The dose rate factor recommended for screening is 5.6×10^{-5} (Sv y^{-1} per Bq g^{-1}), A is the specific activity to which food products are chronically exposed (Bq g^{-1}), equal to $1 \times 10^{-3} \times A_{air}$, f is the fraction of local dietary carbon assumed to be unity (dimensionless).

1.3.3. Estimation of inhaled doses

This evaluation only concerns adults, with the aim of estimating the order of magnitude of inhaled doses, in principle assumed negligible in relation to ingested doses. The inhaled effective dose is expressed according to equation (5), whereas the activity concentration of ^{14}C in the atmosphere can be estimated using equation (6):

$$D_{inh} = FD_{inh} \times C_{air} \times T_{inh} \quad (5)$$

$$C_{air} = f_{air} \times C_{CO_2} \times A_{air} \quad (6)$$

where: D_{inh} is the annual effective dose from inhalation (Sv y^{-1}), FD_{inh} is the inhalation dose coefficient, $FD_{inh} = 6{,}2 \times 10^{-12}$ Sv Bq^{-1} (Euratom, 1996), C_{air} is the ^{14}C concentration in air (Bq m^{-3} air), T_{inh} is the inhalation rate (m^3 y^{-1}), 8 400 m^3 y^{-1} for adult (IAEA, 2001), f_{air} is the volumetric CO_2 concentration in the atmosphere (m^3 CO_2 m^{-3} air), C_{CO_2} is the concentration of carbon in the CO_2 (kg C m^{-3} CO_2), A_{air} is the ^{14}C specific activity of the atmosphere (Bq kg^{-1} C).

The conventional values of f_{air} are measured on a continuous basis at the internationally-renowned Mauna Loa Observatory in Hawaii (Keeling and Whorf, 2001). Since the 1950s, the proportion of the CO_2 volume in the atmosphere (m^3 CO_2 m^{-3} air) has risen from 315×10^{-6} to more than 375×10^{-6} in 2003. The concentration of carbon in CO_2 (kg C m^{-3} CO_2) is estimated based in the molar mass of carbon 12 g mol^{-1}) and the molar volume of an ideal gas at atmospheric pressure (22.4 l mol^{-1}), which is equivalent to about 0.537 kg C m^{-3} CO_2. The concentration of carbon in air would therefore be about 0.2 g C m^{-3} air in 2003. The specific activity reported in Table I was used for A_{air}. The mean global activity of ^{14}C in air increased in relation to its initial value, *i.e.*: 0.07 Bq m^{-3} in 1950 up to a maximum of 0.13 Bq m^{-3} in 1964 before now dropping to value of about 0.09 Bq m^{-3}. The excess ^{14}C due to atmospheric nuclear-weapons testing and other man-made releases reached a maximum of 0.06 Bq m^{-3} in 1964.

2. Results and discussions

Figure 4 represents the total annual dose per age group related to ingested ^{14}C calculated according to equations (2, 3). The effective dose for adults rose from 12.1 µSv y^{-1} in the 1950s to a maximum of about 22.3 µSv y^{-1} in 1964, before dropping to a level of 12.9 µSv y^{-1}, which is slightly higher than its initial value. The "excess" dose reached about ten µSv y^{-1}. Values for children in the 2 to 7 and 7 to 12 age groups were are higher than those for adults, with breast-fed babies being two times less-exposed than adults, owing to their distinctly smaller ration.

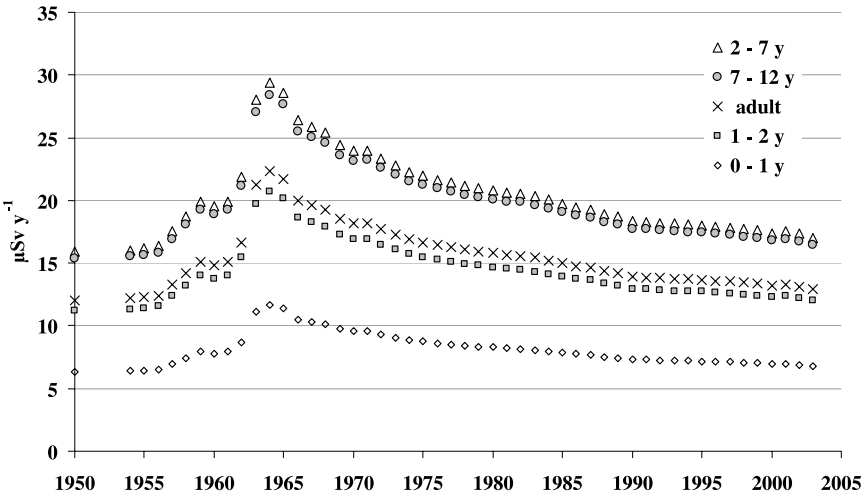

Figure 4 – ***Total annual effective dose by ingestion.***
Dose efficace annuelle totale par ingestion.

Comparison of adult doses thus calculated according to the IAEA simplified method (Eq. (4)) shows that this last method provides results which correspond remarkably with those obtained using the present evaluation (see Fig. 5). In terms of adult doses owing to ^{14}C background radiation, the results of our study: 12.1 µSv y^{-1} in 1950, and those provided by the United Nation Scientific Committee on the Effect of Atomic Radiation (UNSCEAR, 2000): 12 µSv y^{-1} are remarkably similar and do not require further comment. In terms of the "excess" dose related to nuclear-weapons testing, comparison of results obtained here with values estimated by UNSCEAR (2000) revealed a difference that reached a maximum of 2.6 µSv y^{-1} for adults in 1964, with the UNSCEAR doses being smaller. UNSCEAR data provides mean values for the entire globe, which are mainly intended for evaluating collective doses. UNSCEAR modelling involves a succession of specific activity calculations of ^{14}C in the atmosphere, before calculating the biosphere compartments and oceans based on release estimates due to nuclear-weapons testing. The model parameters are adjusted for coherence with available specific activity measurements. Other than the uncertainty related to the UNSCEAR model (18 compartments are taken into account), the difference between UNSCEAR results and those of this study can be partially explained by the specific activity values taken into account seeing that the specific activity considered in this study concerns the European zone and the UNSCEAR takes into account mean planetary values. The specific activity of carbon is higher in the

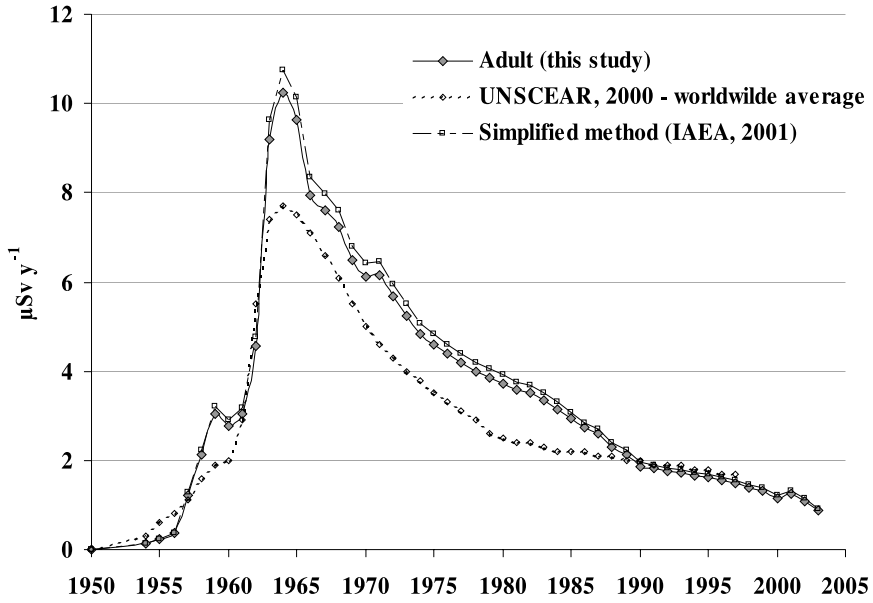

Figure 5 – ***Excess annual effective dose for adults owing to nuclear-weapons testing.***
Excédent de dose efficace annuelle due aux tirs pour l'adulte.

northern hemisphere than in the southern hemisphere: in 1964 for example, this difference reached 60 Bq kg^{-1} (comparison of values provided by Hua *et al*. (2000) with other values, see Fig. 1), which corresponds to a difference of about 3 µSv y^{-1} in terms of ingested doses for adults.

In addition, the estimation of inhaled doses (Eqs. (5, 6)) confirms that this way of exposure is negligible in comparison to ingestion, with a total dose less than 0.01 µSv y^{-1} in 1964 and about 0.005 µSv y^{-1} today. This is equivalent to 4/10000 times the ingested dose.

To estimate the accumulated effective dose, cohorts of generations born every 10 years between 1940 and 1980 were considered and the annual doses were added together taking into account the age of each cohort reached each year. Figure 6 shows the "excess" dose owing to nuclear-weapons testing (as well as illustrating other man-made releases) in relation to the year and generation. In terms of the total dose, the 1940 generation was exposed to the highest "excess" dose: almost 180 µSv after about sixty years, which remains low in relation to the "natural" dose of about 2 mSv per year. The following generations were exposed to smaller doses. The "excess" dose is nevertheless practically identical for generations born in

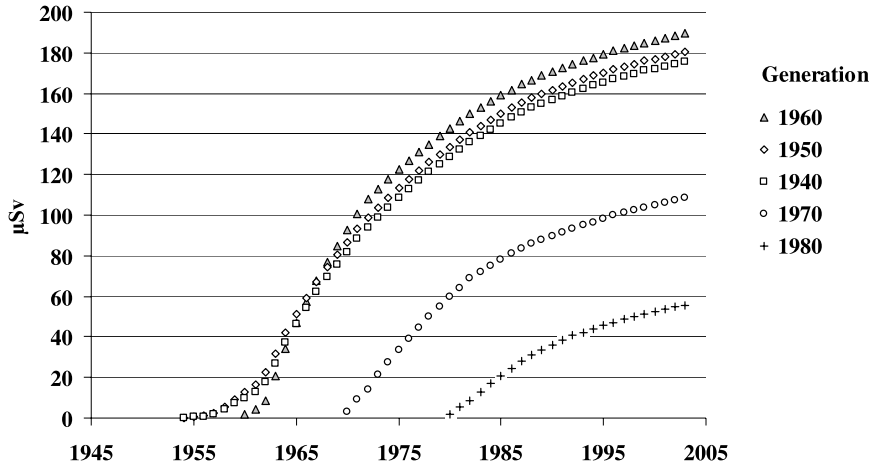

Figure 6 – ***Accumulated effective excess dose depending on the year of birth.***
Excédent de dose efficace cumulée, selon l'année de naissance.

1940, 1950 and 1960: 180 µSv in 2003. The younger generations are far behind with "excess" doses of about 50 to 100 µSv.

In fact, when comparing the modelling of specific activity "transfers" of carbon in compartments and ultimately in plants and animal productions with the modelling of specific activities based on plant measurements themselves, the latter is probably the most accurate method seeing that it immediately provides the specific activity in the terrestrial biosphere. Furthermore, coherence with background radiation values show that the results of estimates are similar for a source that is perfectly balanced on a global level (*i.e.*: natural production of ^{14}C). This tends to confirm the present study, particularly in terms of the food ration in question.

3. Conclusion

Exposure by ingestion of ^{14}C owing to atmospheric nuclear-weapons testing has been evaluated in France between 1961 and 1978. Calculations are based on the consensual hypothesis concerning the isotopic equilibrium of radiocarbon and stable carbon, a major compound found in organic material. The specific activity of carbon-14 in the biosphere – required for calculations – is estimated based on measurements recorded in Europe and available in recent scientific literature. Comparable results (adult effective doses) from this study and results published elsewhere prove to be coherent. Despite the extended persistence of ^{14}C in the

environment (insignificant physical decay and highly-specific "transfer" paths), the maximum effective dose owing to man-made ^{14}C releases remains relatively low: 10 μSv y^{-1} in 1964, in comparison to 270 μSv y^{-1} in 1961 for various fission products resulting from nuclear-weapons testing (Vray and Renaud, 2005). Excess ^{14}C in the 1960s – mostly due to nuclear-weapons testing – practically doubled the exposure related to natural ^{14}C. Exposure to ^{14}C has now dropped to a level that is slightly higher than its initial value in 1950, equivalent to about 12 μSv per year.

From a methodological viewpoint, it seems that simplified modelling is sufficient for such an evaluation, despite the very complex behaviour of carbon in the environment, on the condition that releases such as those resulting from nuclear-weapons testing can be compared to variable but chronic contamination. Estimating the specific activity of ingested carbon by assuming it equal to that measured in plants (this study) makes it possible to bypass sources of uncertainty related to compartment models preferably reserved for global-scale evaluations (UNSCEAR, 2000). An aggregate model (IAEA, 2001) that does not even provide details on the food ration but takes into account the global consumption of carbon (implying the use of an aggregated dose factor) proves to be just as appropriate. However, studying the consequences of local ^{14}C pollution or accidental ^{14}C releases requires mechanistic modelling such as that currently being developed by IRSN.

Appendix

Isotopic discrimination occurs during photosynthesis: the ratios $^{13}C/^{12}C$ and $^{14}C/^{12}C$ are smaller in plant tissue than in atmospheric CO_2 (with $\delta^{13}C$ approaching $-8^0/_{00}$). At equilibrium, the fractionation of ^{14}C is almost equal to two times the fractionation of ^{13}C expressed by $\delta^{13}C$ ($^0/_{00}$), which represents the isotopic ratio $^{13}C/^{12}C$ of the sample in relation to the absolute reference PDB (PeeDee Belemnite, Cretaceous, South Carolina, USA) for which $\delta^{13}C$ is equal to 0 by definition. This fractionation depends on the species according to their dominant photosynthetic cycle (*e.g.*: Waller and Mewis, 1979; Ehleringer *et al.*, 1997). For most C3 plants, *i.e.*: trees and most natural plants growing in temperate or cold zones, the isotopic fractionation varies between −20 and $-35^0/_{00}$, with a mean of $-27^0/_{00}$. C4 plants are less common: tropical grasses, certain halophytic plants, corn, sorghum, etc. Their $\delta^{13}C$ is on average equivalent to $-13^0/_{00}$ (−9 to $-17^0/_{00}$). For ^{14}C, this fractionation is taken into account by applying $\Delta^{14}C$ ($^0/_{00}$) or PCM (percent modern carbon, in percent). This represents the enhancement ($\Delta^{14}C > 0$) or decrease ($\Delta^{14}C < 0$) in the specific activity of ^{14}C, which is standardised in relation to that of the modern carbon reference after having corrected the isotopic discrimination $^{14}C/^{12}C$, itself inferred from the

measurement of $\delta^{13}C$ (Stuiver and Polach, 1977). Hence, the following expressions are currently used:

$$\Delta^{14}C = \left\{ \frac{A_{SN}\, e^{-\lambda(t - t_0)}}{A_0} - 1 \right\} \times 1000$$

(or $PMC = (A_{SN}/A_0) \times 100$), with

$$A_{SN} = A \times \left\{ 1 - \frac{2(\delta^{13}C_{PDB} + 25)}{1000} \right\},$$

where: A_{SN} (Bq kg^{-1} C) is the specific activity in the sample normalised by $\delta^{13}C$, A_0 (Bq kg^{-1} C) is the specific activity of modern carbon, $A_0 = 226$ Bq kg^{-1} C; A (Bq kg^{-1} C) is the non-corrected specific activity in the sample, λ (y^{-1}) is the conventional radioactive constant, $\lambda = (Ln2)/5\,568$, t (–) is the year of sampling and t_0 (–) is the reference year, $t_0 = 1950$, $\delta^{13}C$ ($^0/_{00}$) is the ratio of ^{13}C to ^{12}C in the sample by reference to PDB absolute standard.

These expressions are used for dating purposes, particularly in terms of tree-ring chronology; a derived application involves estimating the environmental contamination in relation to time. Taking into account fractionation guarantees a more accurate follow-up of data.

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