

Evaluation of the terrestrial impact of carbone 14 and tritium released by PWR

M. Fournier, F. Siclet¹ and G. Gontier²

IPSN/DPRE/SERNAT/LMRE, bâtiment 501, Bois des Rames, 91400 Orsay cedex, France

¹ EDF-R&D Laboratoire National d'Hydraulique et Environnement,
6 quai Watier, 78400 Chatou, France

² IPSN/DPRE/SERNAT/LERCM, CEA Cadarache, bâtiment 153,
13108 Saint-Paul-lez-Durance, France

Abstract. Within the scope of a joint IPSN/EDF research program, it is possible to present a statement of the environment out of industrial influence for two areas, one in the Nogent-sur-Seine region, the other around Dampierre, where seasonal sampling was monitored over two years. In Dampierre, the same sampling was implemented within areas exposed to atmospheric releases of the nuclear plant in order to assess the facility impact. The matrices selected for this specific survey include perennial self-sown vegetation (ivy leaves, meadow grass, blackberries), caducous tree leaves (durmast-oak), honey, game, cultivated fruits (apple), pasture grass, milk and cattle meat. The available quantity of results evidences a discrete marking effect by the facilities both in radiocarbon and tritium, but does not allow proposing a modeling.

1. INTRODUCTION

The radiocarbon and tritium activity in the terrestrial ecosystem has been measured from 1994 with various matrices (bryophyta, meadow grass, milk,...), within the scope of yearly and ten-year monitoring of the French nuclear plant. The available quantity of results evidences a discrete marking effect by the facilities both in radiocarbon and tritium, but does not allow proposing a modeling. Interpretation problems are due to the lack of zero point or baseline for both radio-elements, to the fact that they also are from natural origin and are involved in the water cycle and carbon cycle and, with regard to radiocarbon, to the contributions of nuclear weapon atmospheric tests. Within the scope of a joint IPSN/EDF research program, we present a statement of the environment out of industrial influence for two areas, one in the Nogent-sur-Seine region, the other around Dampierre. The site selected in the Nogent-sur-Seine region is considered as a reference site for this survey. In Dampierre, the sampling has undergone a seasonal monitoring over two years, and the same sampling was implemented within areas exposed to atmospheric releases of the nuclear plant in order to assess the facility impact. The matrices selected for this specific survey include perennial self-sown vegetation (ivy leaves, meadow grass, blackberries), caducous tree leaves (durmast-oak), honey, game, cultivated fruits (apple), pasture grass, milk and cattle meat.

2. RADIOCARBON ABUNDANCE

The results of the various sampled matrices are expressed in Becquerel per kilogram of carbon, noted as Bq.kg⁻¹ C. In order to incorporate the isotopic fractionation, and obtain comparable values for radiocarbon activity, the same results are expressed in Δ ‰ [1]. The measured activity is corrected by the isotopic fractionation, using the following equation {1}:

$$A_n = A \left(1 - \left(2 + \delta_{\text{PDB}}^{13\text{C}/12\text{C}} \right) / 1000 \right) \quad \{1\}$$

where A is the measured activity, expressed in Becquerel; $\delta_{\text{PDB}}^{13\text{C}/12\text{C}}$ is the measured value of the isotopic ratio $^{13}\text{C}/^{12}\text{C}$ of the matrix considered in the Pee Dee Belemnite scale; A_n represents the normal activity. The expression as $\Delta \text{‰}$ is obtained with the following equation {2}:

$$\Delta^{14}\text{C} = \left(\left(\frac{A_n e^{\lambda(1950-p)}}{A_0} \right) - 1 \right) \times 1000 \quad \{2\}$$

where $\lambda = 1 / 8267 \text{ years}^{-1}$; p is the sampling year; A_0 is the reference activity, expressed in Becquerel. The value of A_0 is $226 \text{ Bq.kg}^{-1} \text{ C}$.

The values obtained for spot samples of atmospheric carbon dioxide through sparging cannot be used in the current status of data. The problems met during implementation and operation of the cleaners might explain the varying results. Also, the values of the isotopic ratio $^{13}\text{C}/^{12}\text{C}$ measured on the carbon dioxide generated by the combustion of ground matrices are more negative than the expected values, in accordance with the bibliography [2, 3]. The increase in atmospheric carbon dioxide by combustion of fossil products with a low value of isotopic ratio $^{13}\text{C}/^{12}\text{C}$ may have a relationship with this observation.

2.1 Site in the Nogent-sur-Seine region

The radiocarbon results for 1999 and 2000 are reported in $\text{en Bq.kg}^{-1} \text{ C}$ and in $\Delta \text{‰}$ in table 1.

Table 1: Results of samples on the Pierrellez, Sancy-les-Provins site.

$\delta_{\text{PDB}}^{13\text{C}/12\text{C}}$	Type	Sample date	Activity [Bq.kg ⁻¹ C]	$\pm 1 \sigma$	$\Delta \text{‰}$	$\pm 1 \sigma$	Activity ³ H [Bq.l ⁻¹]	$\pm 1 \sigma$	Activity ³ H [Bq.kg ⁻¹ sec]	$\pm 1 \sigma$
	Beef meat	03/01/1999					<1.22	DT		
-18.2	Beef meat	03/01/1999	258.31	6.58	120.76	28.55	2.66	0.14	1.48	0.08
	Oak leaves	09/01/1999					<1.21	DT		
-26.87	Oak leaves	09/01/1999	249.94	6.30	103.51	27.82	5.95	0.2	2.09	0.08
	Blackberries	09/01/1999					1.44	0.14		
-25.19	Blackberries	09/01/1999	251.20	6.36	105.36	27.99	6.82	0.4	3.06	0.18
	Beet leaves	09/01/1999					<1.22	DT		
-26.74	Beet leaves	09/01/1999	251.01	6.36	107.94	28.07	2.56	0.13	0.62	0.03
	Milk	09/01/1999					1.53	0.13		
-16.32	Milk	09/01/1999	256.62	6.50	109.18	28.09	4.35	0.19	2.79	0.13
	Meadow	09/01/1999					1.97	0.13		
-27.17	Meadow	09/01/1999	248.50	6.29	97.80	27.79	1.7	0.12	0.68	0.05
	Ivy leaves	09/01/1999					1.47	0.11		
-30.95	Ivy leaves	09/01/1999	250.80	6.34	116.30	28.22	2.92	0.13	1.36	0.07
-24.9	Blackberries	09/27/2000	251.20	6.29	104.71	27.52				
-27.5	Oak leaves	10/05/2000	252.57	6.34	116.52	27.89				
-28.3	Beet leaves	10/05/2000	245.46	6.15	86.81	27.10				
-17.7	Milk	10/05/2000	252.09	6.32	92.66	27.26				
-27.8	Alfalfa grass pasture	10/05/2000	246.08	6.17	88.48	27.16				
-30	Ivy leaves	10/05/2000	245.64	6.16	91.28	27.23				

The activity value obtained for beef meat, higher than the value obtained for the pasture grass sampled in 1999, may be due to the fact that the beef lived for several years with a varied food. The consistency of samples in ^{14}C , expressed in delta per thousand, shows that the type of matrix has little influence on the result. The average value in $\Delta \text{‰}$ (107 ± 28) obtained with grass and ivy leaves in 1999 is slightly higher than the value obtained in 2000 (90 ± 27). This decrease in ^{14}C activity might be related to an increase in atmospheric carbon dioxide on the sampling location. The overall average value in $\Delta \text{‰}$ is 103 ± 28 .

2.2 Dampierre site out of influence

The Dampierre site was studied with various sampling points located in areas out of influence. The radiocarbon results for 1999 and 2000 are reported in $\text{Bq.kg}^{-1} \text{C}$ and in $\Delta \text{‰}$ in tables 2 and 5. Excluding the atmospheric CO_2 values, the overall average value in $\Delta \text{‰}$ is 103 ± 28 .

Table 2: Results of samples on the Benne and Germigny-des-Prés sites.

$\delta_{\text{PDB}}^{13}\text{C}/^{12}\text{C}$	Type	Sample date	Activity [$\text{Bq.kg}^{-1}\text{C}$]	$\pm 1 \sigma$	$\Delta \text{‰}$	$\pm 1 \sigma$
-10.90	CO_2	03/26/1999	254.76	6.52	88.99	27.87
-22.10	CO_2	03/13/2000	284.86	7.19	245.73	28.05
-20.10	CO_2	06/13/2000	250.52	6.44	91.15	27.80
	CO_2	12/05/2000	253.34	6.37		

2.3 Dampierre site under influence

The Dampierre site was studied with various sampling points located in areas under influence, and most samples were performed 2 km south-south west from the site.

The radiocarbon results for 1999 and 2000 are reported in $\text{Bq.kg}^{-1} \text{C}$ and in $\Delta \text{‰}$ in tables 3 and 6.

Excluding the atmospheric CO_2 values, the overall average value in $\Delta \text{‰}$ is 125 ± 28 .

Table 3: Results of samples on the Lion-en-Sullias and La Tabarderie sites.

$\delta_{\text{PDB}}^{13}\text{C}/^{12}\text{C}$	Type	Sample date	Activity [$\text{Bq.kg}^{-1} \text{C}$]	$\pm 1 \sigma$	$\Delta \text{‰}$	$\pm 1 \sigma$
-14.12	CO_2	03/26/1999	270.63	6.91	164.50	27.65
-21.10	CO_2	03/13/2000	252.60	6.39	102.43	28.89
	CO_2	06/13/2000	233.13	5.96		
-25.00	Miel	08/28/2000	253.97	6.42	117.11	28.11
	CO_2	09/26/2000	271.15	6.80		

2.4 Impact

The value in $\Delta \text{‰}$, slightly higher than the value in the area out of influence, allows assessing the impact of a plant operating four 900 MWe sections: the concentration added to the terrestrial ecosystem close to the facility varies from less than 2 to a maximum of 6Bq.kg^{-1} of carbon.

3. TRITIUM ABUNDANCE

The matrices are analyzed for the tritium content of free water and tritium content of hydrogen bound to the matrix. The tritium content of hydrogen bound to the matrix is measured on the water generated by combustion. The results are expressed in Bq.L^{-1} . With regard to the tritium content of hydrogen bound to the matrix, the results also are expressed in Bq.kg^{-1} of dry matter. The values at detection threshold are identified as DT.

3.1 Site of the Nogent-sur-Seine region

The results of the tritium activity for matrices sampled in 1999 are reported in tables 1 and 4. The high discrepancies observed between free tritium and bound tritium activities for oak leaves and blackberries may be due to high evaporation conditions in drained environment. The overall average value for free tritium is 1.4 Bq.L^{-1} and the overall average value for bound tritium is 3.6 Bq.L^{-1} .

Table 4: Tritium activity for samples on the Pierrelez, Sancy-les-Provins site.

Tritium type	Sample date	Type	Activity [Bq.l ⁻¹]	± 1σ	Activity [Bq.kg ⁻¹ sec]	± 1σ
Free tritium	09/01/1999	Apples	<1.21	DT		
Bound tritium	09/01/1999	Apples	1.98	0.12	0.97	0.06

3.2 Dampierre site out of influence

Due to the availability of the various matrices, the Dampierre site was studied with various sampling points located in areas out of influence. The results of tritium activity for matrices sampled in 1999 and 2000 are reported in table 5. The first series of high values, obtained in 1999 for free tritium and bound tritium, correspond to an area very close to the river (Loire): the overall average value for bound tritium is 4.8 Bq.L^{-1} . Another area out of influence was selected. In this area, the overall average value for free tritium is 1.85 Bq.L^{-1} and overall average value for bound tritium is 3.72 Bq.L^{-1} .

Table 5: Activity of samples on sites out of influence: Benne, Germigny-des-Prés and Saint-Martin d'Abbat.

Sample date	Type	Activity ³ H Bq.l ⁻¹	± 1σ	Activity ³ H [Bq.kg ⁻¹ sec]	± 1σ	^δ PDB ¹³ C/ ¹² C	Activity [Bq.kg ⁻¹ C]	± 1σ	Δ ‰	± 1σ
03/26/1999	Ivy leaves	3.02	0.12				250.56	6.36		
03/26/1999	Ivy leaves	3.08	0.11	1.52	0.05					
03/26/1999	Alfalfa grass pasture	5.22	0.18				249.06	6.33		
03/26/1999	Alfalfa grass pasture	2.81	0.11	1.27	0.05					
07/08/1999	Ivy leaves	3.65	0.13				254.51	6.45		
07/08/1999	Ivy leaves	7.37	0.23	3.66	0.13					
07/08/1999	Alfalfa grass pasture	1.91	0.14				257.42	6.52		
07/08/1999	Alfalfa grass pasture	5.83	0.19	2.93	0.1					
10/19/1999	Meadow	2.7	0.13			-29.94	246.94	6.22	96.93	27.63
10/19/1999	Meadow	6.41	0.21	3.35	0.12					
10/19/1999	Ivy leaves	2.78	0.15			-29.56	249.80	6.31	108.80	28.01
10/19/1999	Ivy leaves	7.44	0.21	3.91	0.13					
08/28/2000	Honey	2.81	0.13			-25.70	249.00	6.29	96.78	27.67
08/28/2000	Honey	1.97	0.15	1.1	0.08					
08/13/2000	Water vapor	<1.23	DT							
08/13/2000	Alfalfa grass pasture	2.61	0.12				243.12	6.13		
08/13/2000	Alfalfa grass pasture	1.52	0.11	0.72	0.05					
08/13/2000	Ivy leaves	<1.22	DT			-31.56	247.88	6.26	104.64	28.11
08/13/2000	Ivy leaves	3.85	0.16	2.19	0.1					
05/13/2000	Ivy leaves	<1.13	DT				247.98	6.31		
05/13/2000	Ivy leaves	1.62	0.1	0.87	0.05					
09/26/2000	Alfalfa grass pasture	<1.19	DT			-29.20	245.43	6.15	88.62	27.88

09/26/2000	Ivy leaves	<1.19	DT			-30.40	249.20	6.24	107.98	27.54
10/20/1999	Durmast-oak leaves	1.65	0.15			-30.27	249.88	6.31	110.71	31.29
10/20/1999	Durmast-oak leaves	7.13	0.22	3.25	0.12					
10/20/1999	Blackberries	1.34	0.11			-27.44	251.37	6.36	111.08	27.76
10/20/1999	Blackberries	3.18	0.14	1.45	0.07					
10/20/1999	Vegetables	1.29	0.09			-29.78	248.53	6.30	103.64	27.72
10/20/1999	Vegetables	1.62	0.12	0.8	0.06					
10/20/1999	Milk	<1.16	DT			-29.76	248.41	6.26	103.07	27.57
10/20/1999	Milk	2.62	0.13	1.75	0.09					
10/20/1999	Alfalfa grass pasture	1.35	0.13			-29.92	247.05	6.23	97.37	27.14
10/20/1999	Alfalfa grass pasture	5.6	0.22	2.59	0.11					
26/10/1999	Chicken	1.28	0.11			-24.16	251.48	6.35	104.31	27.61
26/10/1999	Chicken	1.76	0.09	1.05	0.06					
09/26/2000	Blackberries	1.39	0.11			-27.50	249.66	6.26	103.65	27.91

3.3 Dampierre under influence

The Dampierre site was studied with various sampling points located in areas under influence. The overall average value for free tritium is 1.54 Bq.L^{-1} and the value for bound tritium is 3.69 Bq.L^{-1} . As regards bound tritium, it should be noted that this average value drops from 5.57 Bq.L^{-1} in 1999 to 2.74 Bq.L^{-1} in 2000.

3.4 Impact

No significant discrepancy is detected between samples out of influence and under influence. This non-marking might be due to the prevailing influence of supplies from the Loire river and the alluvial sheet loaded with tritium by the liquid discharges from plants on one side and by the oceanic rain water containing less tritium. It can be noted that tritium levels are globally higher in bound tritium than in free tritium, both within the area under influence and in the area out of influence.

4. CONCLUSION

The measurement of certain indicators such as meadows and ivy leaves allows detecting the radiocarbon impact of the plant. With regard to the reference site and the area out of influence, the average value of radiocarbon activity expressed in $\Delta \text{‰}$ is 103 ± 28 . With regard to the area under influence, the average value of radiocarbon activity expressed in $\Delta \text{‰}$ is 125 ± 28 . The impact of the plant discharges results in an increased radiocarbon concentration ranging from less than 2 to a maximum of 6 Bq.kg^{-1} of carbon, in the environment close to the plant. As for tritium, The edaphic, climatic and hydrologic natural conditions play a part in the distribution of these radionuclides and conceal the possible impact of plant discharges into the terrestrial ecosystem.

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Table 6: Activity of samples on Dampierre sites under influence: Lion-en-Sullias and La Tabarderie.

Sample date	Type	Activity $\pm 1\sigma$ ^3H [Bq.L ⁻¹]	Activity ^3H [Bq.kg ⁻¹ sec]	$\pm 1\sigma$	$^6\text{PDB}^{13}\text{C}/^{12}\text{C}$	Activity $\pm 1\sigma$ [Bq.kg ⁻¹ C]	Δ	σ_{00}	$\pm 1\sigma$
03/26/1999	Ivy leaves	<1.37	DT			252.57	6.41		
03/26/1999	Ivy leaves	3.99	0.16	2.13	0.09				
03/26/1999	Alfalfa grass pasture	<1.37	DT			253.25	6.41		
03/26/1999	Alfalfa grass pasture	1.92	0.13	0.9	0.06				
07/08/1999	Ivy leaves	2.65	0.12			254.72	6.45		
07/08/1999	Ivy leaves	8.32	0.25	2.54	0.1				
07/08/1999	Alfalfa grass pasture	1.4	0.11			257.02	6.50		
07/08/1999	Alfalfa grass pasture	6.28	0.21	2.39	0.09				
07/08/1999	Ivy leaves	2.68	0.11			261.12	6.61		
07/08/1999	Ivy leaves	8.58	0.25	5	0.16				
07/08/1999	Alfalfa grass pasture	1.96	0.12			267.78	6.78		
07/08/1999	Alfalfa grass pasture	4.37	0.15	2.2	0.08				
08/28/1999	Honey	1.69	0.11						
08/28/1999	Honey	1.83	0.16	1.11	0.1				
09/30/1999	Blackberries	<1.16	DT			253.76	6.43		
09/30/1999	Blackberries	1.38	0.12	0.64	0.06				
10/20/1999	Milk	<1.16	DT			257.72	6.49	111.21	28.26
10/20/1999	Milk	<1.24	DT	<0.76	DT				
10/21/1999	Durmast-oak leaves	<1.16	DT			260.94	6.58	155.54	28.43
10/21/1999	Durmast-oak leaves	2.53	0.14	1.26	0.07				
10/21/1999	Vegetables	<1.22	DT			255.85	6.50	128.45	27.98
10/21/1999	Vegetables	1.72	0.11	0.78	0.05				
10/21/1999	Meadow	<1.23	DT			256.26	6.50	136.93	29.14
10/21/1999	Meadow	3.15	0.17	1.78	0.1				
10/21/1999	Ivy leaves	1.98	0.13			256.71	6.48	144.33	28.67
10/21/1999	Ivy leaves	9.19	0.26	5.26	0.17				
10/22/1999	Duck	2.48	0.16			256.70	6.50	119.30	28.84
10/22/1999	Duck	1.74	0.14	0.89	0.07				
03/13/2000	Water vapor	<1.21	DT						
03/13/2000	Alfalfa grass pasture	1.54	0.09			245.27	6.19	91.22	28.34
03/13/2000	Alfalfa grass pasture	1.81	0.16	49.14	4.36				
03/13/2000	Ivy leaves	<1.22	DT			256.83	6.49		
03/13/2000	Ivy leaves	4	0.17	2.23	0.1				
03/21/2000	Water	<1.29	DT						
06/13/2000	Meadow	<1.24	DT			253.09	6.41		
06/13/2000	Meadow	1.73	0.12	0.88	0.06				
06/13/2000	Ivy leaves	1.53	0.1			253.19	6.41		
06/13/2000	Ivy leaves	2.62	0.13	1.41	0.07				
09/26/2000	Blackberries	<1.19	DT			252.02	6.31	109.87	27.4
09/26/2000	Alfalfa grass pasture	<1.19	DT			256.23	6.42	133.60	28.26
09/26/2000	Ivy leaves	1.47	0.13			254.84	6.41	135.74	28.43

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