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^{14}C SOURCES AND DISTRIBUTION IN THE VICINITY OF LA HAGUE NUCLEAR REPROCESSING PLANT: PART I—TERRESTRIAL ENVIRONMENT

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ABSTRACT. COGEMA-La Hague nuclear reprocessing plant in the Cotentin Peninsula (northwest France) releases in the atmosphere about 19 TBq.yr^{-1} of radiocarbon. Three experiments in a terrestrial environment with sampling of a bio-indicator like furze were performed in 1997, 1998, and 1999, and additional air samples in the chimney plume were measured. Results presented here establish the ^{14}C distribution in the La Hague environment and suggest that a part of the ^{14}C content in the vegetation near the coast results from a $^{14}CO_2$ degassing of seawater supplied with the liquid waste from the nuclear plant.

INTRODUCTION

The COGEMA-La Hague nuclear reprocessing plant is located in the northwest part of the Cotentin Peninsula near Cherbourg, France. This nuclear plant releases radioelements in the atmosphere and in the English Channel. Radiocarbon is released to the environment as CO_2 through a 100-m-high chimney, and as liquid waste a few km off the shore, west of the reprocessing plant. Atmospheric ^{14}C releases are estimated to be 19 TBq.yr^{-1} (COGEMA data 2000).

The carbon dioxide is assimilated by plants through photosynthetic processes. Consequently, the ^{14}C activity of vegetation constitutes an integrated record of emissions from COGEMA-La Hague during the vegetative period (spring to fall). The aim of this study is to present the distribution of ^{14}C in the vicinity of the La Hague reprocessing plant.

MATERIALS AND METHODS

During 3 yr (1997–1999), the spring growth of furze was collected by the Laboratoire d'Etudes Radioécologiques de la Façade Atlantique (LERFA) and the Groupe d'Etudes Atomiques (GEA) at 70 stations (Figure 1). Twenty stations were chosen each year for ^{14}C analyses. The samples were prepared using classical methods (Delibrias 1985). The ^{14}C activity was measured by ß-counting in CO_2 proportional gas counters at the Laboratoire de Sciences du Climat et de l'Environnement (LSCE) at Gif/Yvette for years 1997 and 1998 and by benzene liquid scintillation at the University of Georgia (USA) for 1999 samples.

In the COGEMA-La Hague reprocessing plant, atmospheric releases of CO_2 through the chimney are not continuous and occur only during the opening of the nuclear fuel casing, about 10 to 15 times a day. Each release lasts for 30 to 40 min. CO_2 air samples were collected in the COGEMA plant downwind of the chimney plume at Digulleville (Eperons Mount), 1000 m north of the chimney, and at sea, southwest of the plant during the TE-SEA cruise (June 2000). Collection of air samples was done at ground level or 2 m above sea level. The chimney plume is detected by continuously measuring the ^{85}Kr activity (Maro et al. 2001), another gas released by COGEMA-La Hague.

CO_2 in a few liters of air was trapped by bubbling air in sodium hydroxide prepared a few hr before sampling to avoid contamination by sodium carbonate generally present within sodium hydroxide

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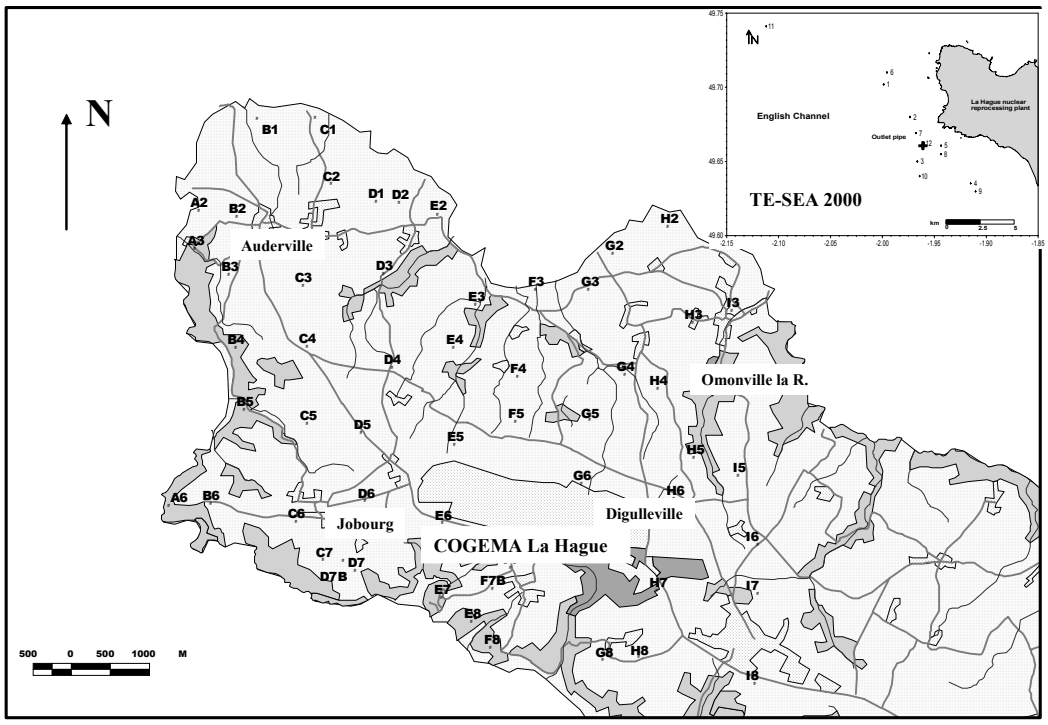

Figure 1 Location of the sampling points in the Cotentin Peninsula and during the TE-SEA cruise (June 2000)

tablets. During 30 min, bigger samples were also collected using a compressor that filled a 50-L bottle. Later, air in the bottle was bubbled in sodium hydroxide.

In the laboratory, barium hydroxide was added to the sodium hydroxide solution to get a precipitate of barium carbonate. This carbonate was then rinsed with degassed distilled water and dried at 50 °C before reaction with orthophosphoric acid under vacuum to evolve CO_2. Water used to prepare the solution was previously degassed. Precipitation, filtration of precipitate, and rinsing were performed under a controlled nitrogen atmosphere. The blank was determined following this procedure on the same solution used for sampling. Very active samples were mixed meticulously with dead carbonate before acid reaction and measurement by ß-counting. Samples were analyzed for ^{14}C activity using accelerator mass spectrometry (AMS) at the Gif AMS facilities or by ß-counting.

Results are expressed in Bq.kg^{-1} C (100 pMC = 226 Bq.kg^{-1} C). The standard deviation varies between 0.5 and 2% for Gif measurements and between 1.3 and 4.4% for the University of Georgia.

RESULTS AND DISCUSSION

^{14}C Activity in the Influence of the Chimney Plume

Results are reported in Table 1. Four samples near the chimney show ^{14}C activity ranging from 4700 to 9800 Bq.kg^{-1} C, with a mean value of 7200 Bq.kg^{-1} C. At sea, 4 to 6 km southwest of the plant, ^{14}C activity ranges between 426 to 908 Bq.kg^{-1} C. Outside the plume influence, the activity ranges between 257 to 291 Bq.kg^{-1} C, with a mean value of 271 Bq.kg^{-1} C, identical to that found at station 11 far to the northwest of the reprocessing plant (Fontugne et al. 2002). This value is slightly higher

than can be expected for unpolluted air. These results show a rapid dilution of the plume, the activity decreasing by a factor of 10 in a few km.

Table 1 ^{14}C activity (Bq.kg^{-1} C) in the chimney plume.

Sample reference	^{14}C activity (Bq/kg C)
Digullville (near COGEME-La Hague reprocessing plant)	
B1	4767 ± 71
B2	7821 ± 67
B3	6518 ± 23
B4	9783 ± 61
TE-SEA cruise	
6	428.9 ± 2.5
7	468.7 ± 2.7
8	435.1 ± 2.7
9	908.1 ± 3.2
10	426 ± 2.7
11[a]	271.1 ± 2.7

[a]Value for sea air samples outside the influence of the chimney plume.

^{14}C Activity in Furze

Results are reported in Table 2. ^{14}C activity varies between 250.5 and 587.1 Bq.kg^{-1} C. Reference samples were collected in Brittany, 300 km southwest from La Hague, and gave values between 252.6 and 254.3, in good agreement with ^{14}C activity in air and plants growing outside areas contaminated by industrial ^{14}C. Samples from La Hague clearly record the influence of the reprocessing plant, the activity varying between 1 to 2 times the reference activity. The ^{14}C distribution is similar for the 3 considered years (1997–1999) and clearly depends on the dominant wind direction, mainly a SW/ENE axis during the vegetative period. However, abnormally high values are found near the coast southwest of the plant at the extreme west of the peninsula, near Auderville and south of Jobourg. This excess of ^{14}C at the coast suggests a marine contribution through degassing from seawater of $^{14}CO_2$ originating from the liquid waste released at the pipe outlet south of the nuclear plant. The pCO_2 measurements in water indicate that the sea was a source of CO_2 for the atmosphere (Keir et al. 2001; Fontugne et al. 2002). Atmospheric ^{14}C values measured at sea outside of the chimney plume that are about 20 Bq.kg^{-1} C higher than atmospheric reference values (1997–1999) support the hypothesis of a marine contribution.

CONCLUSIONS

Measurements of ^{14}C activity in the chimney plume show a rapid dilution, from about 7200 Bq.kg^{-1} C near the chimney to values ranging between 400 and 900 Bq.kg^{-1} C at a distance of 4 to 6 km. Outside periods of release, the ^{14}C residual value is around 270 Bq.kg^{-1} C. Vegetation around the nuclear plant records these periods of release, concentration in bio-indicators ranging between 1 to 2 times the present atmospheric background. Higher ^{14}C concentrations are observed at the coast, suggesting a supplementary marine contribution though the degassing of the ^{14}C excess supplied by the liquid releases of the nuclear plant. This contribution has been estimated for year 2002 (Maro et al., these proceedings).

Table 2 ^{14}C activity (Bq.kg^{-1} C) in furze during years 1997, 1998, 1999.

1997		1998		1999	
A2	262.8 ± 2.6	A2	281.4 ± 1.4	A2	255.3 ± 8.0
A6	263.3 ± 2.6	A6	279.1 ± 1.7	A6	265.0 ± 7.8
B1	275.7 ± 2.8	B1	258.8 ± 1.8	B1	250.5 ± 3.3
C3	262.4 ± 2.6	C3	261.3 ± 1.0	C6	273.2 ± 12.0
C5	256.7 ± 2.6	C5	259.7 ± 1.6	D7	360.5 ± 6.2
C6	288.8 ± 2.9	C6	304.0 ± 2.4	E6	398.2 ± 9.3
C7	464.7 ± 4.6	C7	455.2 ± 4.6	E7	376.0 ± 12.7
E2	265.8 ± 2.7	D7	469.2 ± 2.3	F3	332.8 ± 8.8
E8	388.7 ± 3.9	E2	262.4 ± 0.8	F5	352.3 ± 7.0
F3	416.3 ± 4.2	E6	525.2 ± 3.7	G5	530 ± 8.8
F5	378.6 ± 3.8	E8	334.0 ± 2.3	G8	317.3 ± 8.5
G8	315.5 ± 3.2	F3	401.6 ± 1.2	H2	294.8 ± 11.5
H2	290.9 ± 2.9	F5	338.5 ± 2.0	H4	339.0 ± 11.2
I5	396.9 ± 4.0	G5	587.1 ± 4.7	I5	338.5 ± 11.0
I8	269.8 ± 2.7	G8	265.1 ± 1.6	I8	275.0 ± 8.0
ANSE CULERON	271.4 ± 2.7	H2	316.9 ± 1.6		
D7	461.5 ± 4.6	H4	272.1 ± 2.2	**Reference sample**	
E6	407.0 ± 4.1	I3	302.4 ± 1.5	ash leaves	243.7 ± 8.2
G5	365.0 ± 3.6	I5	353.2 ± 1.8		
H4	383.0 ± 3.8	I8	269.2 ± 1.1		
I3	299.2 ± 3.0				
		Reference samples			
		PEN MARC'H, furze	252.7 ± 1.3		
		LE CONQUET, oak	252.9 ± 1.0		
		A2-31/3/99	254.3 ± 2.5		

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