$^{129}I/^{127}I$ RATIO MEASUREMENTS IN BOVINE THYROIDS FROM THE NORTH COTENTIN AREA.

C. Fréchou[1], D. Calmet[1], X. Bertho[2], A. Gaudry[2]

[1] IPSN/DPRE/SERNAT – CE Saclay, Bât 130 – 91191 Gif sur Yvette Cedex - France
* E-mail : carole.frechou@ipsn.fr
[2] CEA/DSM/DRECAM/LPS, CEN Saclay, Bât 647, 91191 Gif sur Yvette Cedex – France

1. INTRODUCTION

Since 1945, the explosions of nuclear devices in the atmosphere and the increasing effluent discharge from the nuclear spent fuel reprocessing plant contributed to significantly enhance the production of ^{129}I in the environment [1]. Its activity has increased with time in various components of the atmosphere, hydrosphere and geosphere compartments [2]. While the pre-nuclear $^{129}I/^{127}I$ ratio is estimated between 10^{-12} and 10^{-9}, nowadays, it reaches levels near 10^{-5} in the vicinity of european nuclear fuel reprocessing plants [3, 4]. Thus, the ^{129}I activity monitoring in the environment of fuel reprocessing plants requires particular radioecological studies to assess the corresponding impact on human population [5].

Recently, it was shown that direct γ-X spectrometry is an efficient and rapid method to measure ^{129}I. It can be used for the radiological surveillance of the terrestrial environment of the spent nuclear fuel reprocessing plant, as well as to qualify the long-distance impact on the marine environment [6].

Iodine is accumulated in the mammalian thyroid where hormones are metabolised. Thus, thyroids can be used as sentinel organs to monitore the activity level of ^{129}I [7]. The routine measurement of the ^{129}I activities in mammalian thyroids living near a fuel reprocessing plant can be particularly efficient to characterize the spatio-temporal influence of the ^{129}I gazeous effluent discharges. It can also demonstrate a potential exposure, through inhalation or ingestion pathways, of the human population to ^{129}I discharges in the environment.

This study presents the ^{129}I activity and $^{129}I/^{127}I$ ratio measured in bovine thyroids collected in 1980 and 1999 in the North Cotentin area under the influence of the low level radioactive gaseous effluents authorized to be discharged by the La Hague reprocessing plant.

2. MATERIALS AND METHODS

In February 1980 and March-April 1999, thyroids were sampled on cows from herds located respectively in four and fifteen sites in the North-West Cotentin, in the nearfield environment of the nuclear fuel reprocessing plant of La Hague (Figure 1). The thyroid dried masses obtained after blending varied between 10 and 20 grams. Samples were aliquoted and prepared separately for their ^{129}I activity measurement by direct γ-X spectrometry with self-absorption correction [6, 8] and Instrumental Neutron Activation Analysis (INAA) for ^{127}I with the ORPHEE reactor in Saclay [9].

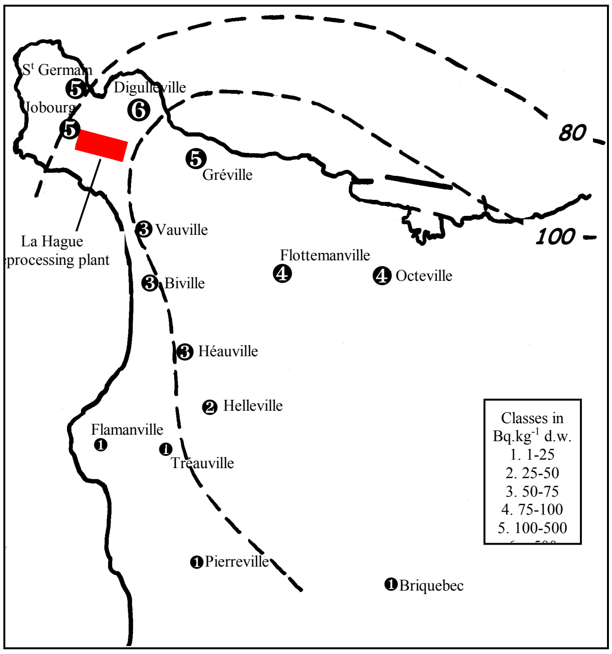

Figure 1 : Location of the herds sampled in the North Cotentin region. Dotted lines figure the rain amplitudes (cm.yr^{-1}). Each circled number represents one of the six classes of measured ^{129}I activities.

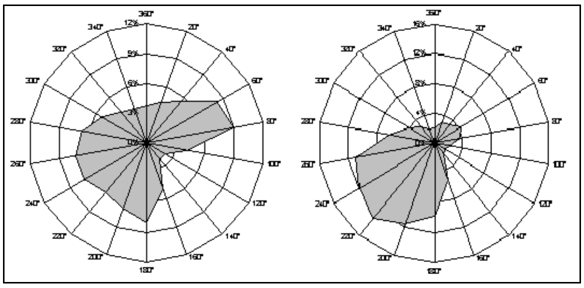

Figure 2 : Wind roses for dry an rainy weather conditions measured at the La Hague gazeous effluents exhaust stack.

3. RESULTS AND DISCUSSION

There are significant differences for thyroid ^{129}I activities between samples collected the same year at different locations, as well as between those sampled in 1980 and 1999 at the same sites (Table 1). They vary between 6.6 ± 0.8 and 234 ± 20 Bq.kg^{-1} d.w in 1980 samples and between 8 ± 1 and 2080 ± 160 Bq.kg^{-1} d.w in 1999 samples. Stable iodine concentrations are comprised between 1010 ± 4 and 1670 ± 8 mg.kg^{-1} d.w in 1980 and between 821 ± 45 and 2700 ± 124 mg.kg^{-1} d.w in 1999.

^{129}I/^{127}I ratios vary by a factor of 20 in 1980 samples from (1.00 ± 0.08)×10^{-6} to (21.4 ± 2.5)×10^{-6}, whereas they are spread over two orders of magnitude, between (0.91 ± 0.11)×10^{-6} and (251 ± 0.23)×10^{-6}, for 1999 samples. By comparison, the ratio was given at 0.13×10^{-6} in a thyroid sample collected in 1973 in the same area [10].

The Bravais-Pearson coefficient computed for stable iodine concentration and ^{129}I activities data of this study shows no significant linear correlation (r=0.073) and confirms the relative impact, with time and space, of an industrial source of ^{129}I.

The ^{129}I/^{127}I ratios measured in this study are close to those of roe and fallow deer thyroids collected in 1983 in the vicinity of the Karlsruhe reprocessing plant, 9 and 15×10^{-6} respectively [3]. They are substantially higher than those measured in the last ten years in animal thyroids coming from sites contaminated with ^{129}I global atmospheric fallout but not by industrial releases (10^{-11} to 10^{-9})[11] and than those of thyroids collected between 1978 and 1993 in western European countries (10^{-9} and 10^{-8})[12].

^{129}I/^{127}I ratio levels, and their variability clearly indicate a relative influence, in space and time, of the reprocessing plant gaseous effluent discharges.

Breeding's location	Slaughter date	^{129}I Bq.kg^{-1} d.w.	Stable I mg.kg^{-1} d.w.	$^{129}I/^{127}I$ .10^{-6}
La Hague	Oct. 1973	0.7-2.3*	870-2700*	0,13[a]
Héauville	Feb 1980	6,6 ± 0,8	1010 ± 4	1,00 ± 0,08
Flottemanville	Feb 1980	55 ± 5	1600 ± 9,6	5,26 ± 0,05
Flamanville	Jan. 1980	15,6 ± 1,8	1593 ± 6	1,50 ± 0,13
Digulleville	Jun. 1980	234 ± 20	1670 ± 8	21,4 ± 2,5
Pierreville	Mar. 1999	8 ± 1	1390 ± 71	0,91 ± 0,11
Tréauville	Mar. 1999	16 ± 2	1830 ± 88	1,34 ± 0,18
Briquebec	Mar. 1999	24 ± 3	1110 ± 56	3,31 ± 0,45
Flamanville	May 1999	18 ± 2	821 ± 45	3,37 ± 0,38
Vauville	Apr. 1999	64 ± 5	2700 ± 124	3,63 ± 0,33
Helleville	May 1999	26 ± 3	1080 ± 55	3,68 ± 0,52
Biville	Apr. 1999	55 ± 6	2250 ± 108	3,74 ± 0,44
Octeville	Mar. 1999	81 ± 6	1970 ± 93	6,29 ± 0,69
Héauville	Mar. 1999	63 ± 5	1390 ± 71	6,94 ± 0,65
Flottemanville	Apr. 1999	93 ± 11	1850 ± 89	7,69 ± 0,98
St Germain des Veaux	Mar. 1999	128 ± 10	2340 ± 110	8,37 ± 0,76
Jobourg	Apr. 1999	329 ± 25	2260 ± 106	22,28 ± 1,99
Gréville Hague	Mar. 1999	411 ± 45	1630 ± 81	38,59 ± 4,64
Digulleville	Mar. 1999	2080 ± 160	1270 ± 66	250,68 ± 23,56

Table 1 : ^{129}I in Bq.kg^{-1} dry weight measured by direct γ-X spectrometry, stable iodine concentration in mg.kg^{-1} dry weight measured by INAA and $^{129}I/^{127}I$ ratio calculated for each thyroid collected in the Cotentin area. * : Considering that the stable iodine content in our thyroids varies between 870 and 2700 ppm, ^{129}I in the 1973 sample can be estimated between 0.7 and 2.3 Bq.kg^{-1}.

3.1 Spatial evolution

The ^{129}I activities in the 1999 thyroids show a variability of two orders of magnitude that can be explained by the geographical position of the grazing sites of the sampled cows, considering the prevailing wind directions, as well as their distance from the exhaust stack (Figure 1 and 2). Thyroids, collected from herds located between 6 and 30 km to the south of the La Hague reprocessing plant, present activity levels and isotopic ratios lower than 100 Bq.kg^{-1} d.w. and 10^{-5} respectively. Thyroids collected at a distance comprised between 1 and 6 km to the east and the west of the plant show activities and isotopic ratios lying between 80-500 Bq.kg^{-1} d.w. and 10^{-5}-10^{-4} respectively. The highest ratio, 2.5×10^{-4}, and the corresponding maximal activity of 2080 Bq.kg^{-1} d.w., was observed at Digulleville, 3 km at the North-West of the plant. Thyroids collected in 1980 from bovines grazing on the same locations show an identical spatial distribution of activities and ratios, with a maximum detected again at the same Digulleville site.

The wind roses (Figure2) show that Digulleville is situated under the dominant winds for both dry and rainy weather conditions. This can easily explain the results one order of magnitude higher in Digulleville compared to the thyroids collected on animals coming from other locations.

3.2. Temporal evolution

From the beginning of the La Hague reprocessing plant activities in 1966 up to 1999, there have been major fluctuations in the annual gaseous effluents discharges of ^{129}I (Figure 3) [5]. Nevertheless, there is a noticeable annual trend starting with a period of marked increase, wich peaked in 1996, then followed by a significant decrease, reaching back again the 1980 levels in 1999. The total gazeous activity discharged into the environment from 1966 to 1999 can be estimated at 493×10^9 Bq.

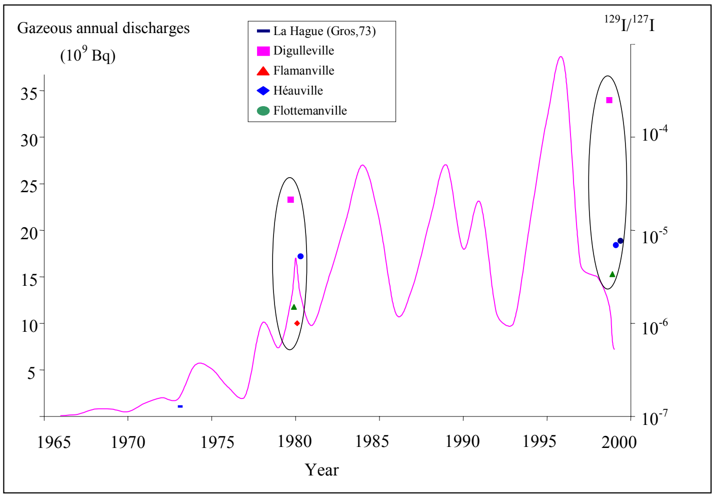

Figure 3 : $^{129}I/^{127}I$ values measured in the bovine thyroid samples collected in the Cotentin region at three different periods, compared to the ^{129}I gaseous effluents releases of the La Hague reprocessing plant.

The ratios at Digulleville show an increase of one order of magnitude between 1980 and 1999. The thyroids collected on the farthest sites Flamanville, Héauville and Flottemanville, show a lower increase of the ratio and the ^{129}I activity levels.

3.3. Transfer to the cow

Level differences observed in thyroid between the stations under the direct influence of the plant and the farthest ones, certainly reflect different dispersion and mixing levels, with the natural stable iodine, of the discharged ^{129}I before its inhalation and/or digestion after their deposition on soil and grass.

Iodine-129 uptake in the thyroid may occur by ingestion or inhalation pathways with fractional uptake coefficients of 0.3 and 0.23 respectively [13]. ^{129}I activity values measured in the grass samples collected in Digulleville area are comprised between 0.2 and 1 $Bq.kg^{-1}$ fresh weight [6]. Considering that a cow eats 50 kg of grass a day and that the residence time for iodine in the thyroid is 100 days, with a captation coefficient of 30% of the ingested iodine, the massic ^{129}I activities, taking into account the variability of the thyroid mass, in thyroid samples should be comprised between 3.10^3 and 60.10^3 $Bq.kg^{-1}$ dw. These estimated values are greater by a factor of 10 than the highest activity measured.

This difference can be explained by the environmental conditions in the North Cotentin area. This maritime region is under the ocean influence and stable iodine, through seaspray transport, is available in great quantities. Thyroid saturation is the result of an active process that occurs with a high plasma concentration of iodine [14]. Thus, stable iodine, in large quantities, prevents the transfer of radioactive iodine to the thyroid [15] and can explain that the ^{129}I activities measured in bovine are lower than predicted ones.

4. CONCLUSION

^{129}I levels as well as $^{129}I/^{127}I$ ratios measured in bovine thyroids coming from cows grazing in the North Cotentin area are significantly higher than natural levels demonstrating the influence of the La Hague reprocessing plant discharges. The maximum value observed in the vicinity of the plant is 4 orders of magnitude higher than the currently background level characteristic of natural environment not influenced by industrial releases of ^{129}I. It can be compared to a ratio of 1.5×10^{-3} that is considered to deliver an annual thyroid equivalent dose to man of 1 $mSv.y^{-1}$ [13]. The spatial distribution can be explained by predominant wind directions. Comparison of ^{129}I activity and $^{129}I/^{127}I$ ratios measured in 1980 and 1999, show an increase, more or less important considering the geographical position of the sites.

This study confirms the interest of bovine thyroid as the appropriate sentinel organ to monitore the environmental impact of ^{129}I gazeous release in the environment, using direct γ-X spectrometry technique under conventional measurement conditions of a surveillance laboratory.

Aknowledgements

The authors would like to thank Dr N. Parmentier for her encouragement and the provision of 1980 thyroid samples, Ms Dubois from the Veterinary Service of Cherbourg's slaughter for the sampling of the 1999 samples. We would like to also thank Mr D. Maro (IPSN) and Cogema for the ^{129}I gazeous effluent discharges data. ^{129}I γ-spectrometry measurements were performed at the Institut de Protection et de Sûreté Nucléaire (IPSN) by Mr Bouisset, Mr Cagnat, Mr Kerlau and Mr Mekhlouche.

References

1. NCRP, *Iodine-129 : evaluation of releases from nuclear power generation*. 1983, National Council on Radiation Protection and Measurements.
2. Michel R., et al. *Status and trends of Iodine-129 abundances in the European Environment*. in *IRPA 10*. 2000. Hiroshima: CD.
3. Robens E., Hauschild J., and Aumann D.C., *Iodine-129 in the environment of a nuclear fuel reprocessing plant : II. Iodine-129 and Iodine-127 content of soils, forage plants and deer thyroids.* J. of Environ. Radioac., 1988. **7**: p. 165-274.
4. Fréchou C., et al. *Détermination de l'iode 129 dans des matrices environnementales par activation neutronique et spectrométrie gamma-X directe*. in *Journées techniques CETAMA sur les Mesures et Analyses dans les Matrices Environnementales et Biologiques.* 1999. INSTN/Cadarache.
5. Groupe Radioécologie Nord Cotentin, *1- Inventaire des rejets radioactifs des installations nucléaires*. 1999.
6. Fréchou C., *Optimisation des protocoles de mesurage de ^{129}I et $^{129}I/^{127}I$. Etablissement d'une méthodologie applicable aux échantillons de l'environnement*. 2000, Report CEA-R- 5947. 150pp.
7. Van Middlesworth L., *^{129}I and ^{137}Cs fission products in thyroids of animals, 1984-1991.* Health Physics, 1993. **64**(1): p. 52-58.
8. Bouisset P., et al., *Direct gamma-X spectrometry measurement of ^{129}I in environmental samples using experimental self-absorption corrections.* Nucl. Instr. and Meth. in Phys. Res. A, 1999. **437**: p. 114-127.
9. Piccot D., et al., *Ko-LABSUE : a quasi non-dependent data format package for k0-quantification.* J. of Radioanal. and Nucl. Chem., 1997. **215**(2): p. 263-269.
10. Gros R., et al., *Dosage de l'iode 129 dans les thyroides*. 1975.
11. Hou X., Dahlgaard H., and Nielsen S.P., *Iodine-129 time series in Danish, Norwegian and Northwest Greenland coast and the Baltic Sea by seaweed.* Estuarine, Coastal and Shelf Science, 2000. **51**: p. 571-584.
12. Handl J., *Concentrations of ^{129}I in the biosphere.* Radiochimica Acta, 1996. **72**: p. 33-38.
13. UNSCEAR, *Sources and effects of ionizing radiation.* 1977.
14. Geoffroy B., *Pharmacocinétique de l'iode.* Radioprotection, 2000. **35**(2): p. 151-174.
15. Crout N.M.J., et al., *A model of radioiodine transfer to goat milk incorporating the influence of stable iodine.* Radiat. Environ. Biophys., 2000. **39**: p. 59-65.