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To the paper

Iodine-129 in thyroid glands: A sensitive biological marker of fission product exposure

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Iodine-129 may be no radiation hazard but it is a useful marker. Animal thyroids concentrate the isotope to 4 orders of magnitude greater than the intake. This results in a potential biological and physical indicator of radioactive contamination. Since 1943, $^{129}\text{I}/^{127}\text{I}$ ratio in animal thyroids from the Northern Hemisphere has increased 2 to 5 orders of magnitude. Since 1985, thyroids of deer, living near a nuclear reprocessing facility have contained ^{129}I which is 3 to 7 orders of magnitude greater than pre-nuclear levels. Limited measurements of ^{129}I in thyroids from the Southern Hemisphere have shown little increase. An appendix is presented to show that ^{129}I may be helpful to evaluate past radiation hazard from fission products.

Introduction

The low energy beta, gamma and X-ray emissions from ^{129}I with its long half-life ($1.6 \cdot 10^7$ y) and low specific activity (5.7 mg/ μCi), result in its being a minimal biological hazard.¹ The isotope undergoes intense biological concentration into the thyroid of grazing animals. Bovine thyroids can accumulate 20 to 30% of the iodine, from approximately 30 to 60 days of grazing, into their 10 to 15 g thyroid and it is retained for a long biological half-life. The thyroid may contain 10^4 to 10^5 times higher concentration of iodine than the diet.

The fission yield of ^{129}I from ^{235}U and ^{239}Pu is 0.9% and 1.6% of the atoms undergoing fission.

Iodine-129 is rare in nature having decayed to immeasurable levels since creation but small quantities are produced by natural fission and by spallation of xenon in the upper atmosphere of the earth. Influx of the isotope into the biosphere is expressed as the ratio ^{129}I to non radioactive ^{127}I .

Measurement

Direct measurement of ^{129}I in thyroid glands has been conducted by X-ray spectrometry in a germanium well when ^{129}I exceeded 10^{-2} Bq/g.

Accurate measurements from lower concentrations were obtained by preserving the glands in paraformaldehyde powder and sending them to the laboratory of HANDL et al. in Hannover, Germany, where neutron activation analyses² were conducted and the data are reported as mass ratio of ^{129}I to ^{127}I . This has proven satisfactory for isotope ratios as low as 10^{-10} . More recently, HANDL³ has applied Accelerator Mass Spectrometry (AMS) to determine $^{129}\text{I}/^{127}\text{I}$ less than 10^{-10} .

Environmental findings (Table 1)

The most pristine biological material tested for $^{129}\text{I}/^{127}\text{I}$ are human thyroids⁵ from pre-1936 and powdered hog thyroids³ from 1943. Their $^{129}\text{I}/^{127}\text{I}$ were (<4 to 7) $\cdot 10^{-11}$ respectively. These values may be greater than those found in geologic brine,⁴ (0.5 to 1) $\cdot 10^{-11}$. BRAUER et al.⁵ showed that during 1964–1969 the $^{129}\text{I}/^{127}\text{I}$ ratios in thyroids of grazing animals ranged from 10^{-9} to 10^{-5} throughout the USA. The ratios were partly dependent upon the distance of the grazing area from nuclear reprocessing operations.

In Europe, during 1981 the $^{129}\text{I}/^{127}\text{I}$ in 22 bovine thyroids was measured by HANDL et al.⁶ Their results showed the background ratio in Germany at that time was $4.7 \cdot 10^{-9}$.

Within 2 months after the Chernobyl accident, the $^{129}\text{I}/^{127}\text{I}$ ratio in Austria and Southern Germany was $3 \pm 0.6 \cdot 10^{-7}$; it had increased 50–100 times greater than before the accident.² Five to six years after the accident, HANDL⁷ measured the isotope ratio in soil, 75 to 420 km from Chernobyl. He found 88% of 17 soil samples showed $^{129}\text{I}/^{127}\text{I}$ ratios which averaged $(4 \pm 3) \cdot 10^{-7}$. This was similar to the ratio in fresh bovine thyroids, 5 years earlier. The remaining soil samples and those closer to Chernobyl had ratios 5 to 8 times greater.

By 1992–3, the mean $^{129}\text{I}/^{127}\text{I}$ in 9 bovine thyroids from northern Germany⁷ averaged $(4 \pm 1.8) \cdot 10^{-8}$.

The variability of $^{129}\text{I}/^{127}\text{I}$ was illustrated by 2 bovine thyroids sampled in Ulm, Germany, 18 days before the Chernobyl accident;² the $^{129}\text{I}/^{127}\text{I}$ was 5.1 and $6.0 \cdot 10^{-8}$, which was more than 10 times greater than in 1981. In addition, unexpectedly high levels of ^{129}I were found 1–2 months after the Chernobyl accident, when 7 of 9 thyroids from Austria and Germany had $^{129}\text{I}/^{131}\text{I}$ ratios² which were higher than predicted from the expected composition of the reactor release.⁸ It is suggested that unexpected increases in ^{129}I may have originated from unidentified sources.

Cesium-137 is a readily measured fission product and it is sometimes used as an indicator of potential presence of ^{129}I . Chemically and biologically Cs resembles potassium and ^{137}Cs is incorporated, but not necessarily concentrated, by living organisms. We measured ^{129}I , ^{131}I and ^{137}Cs in available thyroids after the Chernobyl accident. Concentrations of the isotopes in the thyroids

depended upon the composition of the original mixed fission products as well as biological and environmental factors. In an appendix to this report, data are shown to relate the measured and expected concentrations of the three isotopes in representative thyroids.

It is emphasized that continuous increases in environmental ^{129}I and ^{137}Cs will limit the usefulness of these measurements as indicators of new increments, unless background levels are established periodically.

There are few measurements of $^{129}\text{I}/^{127}\text{I}$ in animal thyroids from the Southern Hemisphere but they show lower ratios than in the Northern Hemisphere. In 1985–1986, HANDL et al.⁷ found $^{129}\text{I}/^{127}\text{I}$ in bovine thyroids from Southern Chile ranged $(1.5 \text{ to } 2.2) \cdot 10^{-9}$ and in ovine thyroids from Australia the ratios were only $(6 \text{ and } 5) \cdot 10^{-10}$ in animals sampled in 1957 and in 1989 respectively.

Monitoring of ^{129}I and ^{137}Cs within a nuclear reprocessing area

During the past 12 years in the USA we have utilized germanium X-ray spectrometry to monitor ^{129}I in thyroids of wild deer from a 310 sq. mi. area surrounding the nuclear reprocessing facility at the Savannah River Site (SRS) in South Carolina⁹ (Fig. 1). Each year public hunts are conducted to control the deer population. Thyroid and muscle samples were preserved in paraformaldehyde and sent to the laboratory for isotope measurements.

Table 1. Iodine-129 in thyroid glands

	Year	Area	$^{129}\text{I}/^{127}\text{I}$	Reference
pre	1936	USA (h)	$<4 \cdot 10^{-11}$	4
	1943	USA (p)	$7 \cdot 10^{-11}$	1
	1969–75	USA (b)	$1 \cdot 10^{-9} \text{ to } 10^{-5}$	4
	1981	Germany (b)	$4.7 \cdot 10^{-9}$	3
	1986	S. Germany (b)	$5.8 \cdot 10^{-8}$	2
After Chernobyl accident (Apr. '86)				
	June-86	Germany & Austria (b, d)	$2.9 \cdot 10^{-7}$	2
	1992–93	Germany (b)	$4.0 \cdot 10^{-8}$	3
Southern hemisphere				
	1957	Australia (s)	$6.0 \cdot 10^{-10}$	3
	1985–86	S. Chile (b, h)	$(1.0\text{--}4.2) \cdot 10^{-9}$	3
	1989	Australia (s)	$5.0 \cdot 10^{-10}$	3

(h) = human; (p) = porcine; (b) = bovine; (s) = sheep.

(1) Handl, J., unpublished (personal communication 1998).

(2) VanMiddlesworth, L., Handl, J., Health Phys., 73 (1997).

(3) Handl, J., Radiochimica Acta, 72 (1996).

(4) Brauer, F. P., Soldat, J. K., Tenny, H., Strebin, R. S., Vienna, IAEA (1974).

Fig. 1. Savannah River Site wildlife compartments; central compartments.
Redrawn from Savannah River Environmental Report, 1984, DPSPU 85-30-1

Fig. 2. ^{129}I Bq g $^{-1}$ in Savannah River Site deer thyroids, 1995–1997

Fig. 3. Cesium-137 by year and by subdivision of Savannah River Site

The ^{129}I in the deer thyroids has ranged between 10^{-3} to 10^2 Bq/g thyroid (Fig. 2). If we assume that the deer have normal concentrations of chemical iodine in

their thyroids ($0.84 \text{ mg } ^{127}\text{I/g thyroid}$) then, the $^{129}\text{I}/^{127}\text{I}$ ratio would range between 10^{-7} to 10^{-2} .

The Savannah River Co. reported a special study^{11,12} and summary¹³ of ¹³⁷Cs at SRS. These reports suggested that if deer from the Site contained >5 pCi ¹³⁷Cs/g muscle (>0.19 Bq/g muscle) then, they contained ¹³⁷Cs in excess of world-wide fallout. (This limit would be equivalent to approximately >0.13 Bq ¹³⁷Cs/g thyroid).

The measurement of ^{129}I , and possibly ^{137}Cs , in animal thyroids (with other information) may prove to be a sensitive integrator of environmental contamination and may be helpful in evaluating radiation dosimetry associated with past exposure to mixed fission products.

Table 2. Calculation of original ^{131}I from ^{129}I measured 10 years after the Chernobyl accident

Thyroid number	^{129}I in 1996 measurement (assume same as accident date)		Reactor equilibrium $^{129}\text{I}/^{131}\text{I}$ at 660 days	^{131}I (maximum) calculated to accident date	
	mBq/g	ng/g		ng/g	kBq/g
1A	0.98	0.14	16	0.0087	39.5
3A	2.2	0.34	16	0.0212	96
					(Mean) 68

Original ^{131}I from ^{131}I measurements 4–5 weeks after the Chernobyl accident			
Same as thyroids above	^{131}I in July 1986 measurement, ^{131}I as of sample dates		^{131}I calculated to accident date
			kBq/g
1A	26 May '86	2.78 kBq/g	*37
3A	2 June '86	3.45 kBq/g	*83
			(Mean) 60

* The published³ values were 14% larger.

Appendix I

Example of delayed measurement of ^{129}I , used to estimate original ^{131}I exposure (Table 2)

Some of our samples of bovine thyroids, after the Chernobyl accident in 1986, contained unexplained quantities of ^{129}I , and their ratios of $^{129}\text{I}/^{131}\text{I}$ exceeded the ratios within an operating nuclear reactor. We cannot explain the four highest ratios² of $^{129}\text{I}/^{127}\text{I}$.

In the following example (Table 2), we use two bovine thyroids from Bad Hall, Austria in which ^{131}I and ^{137}Cs were measured² within two weeks after the animals were killed. The initial radioactivities of ^{129}I , ^{131}I and ^{137}Cs were assumed to be the reactor equilibrium ratios ($\text{g}\cdot\text{g}^{-1}$) after 660 days of operation.⁸ That was the nearest time for which the authors had data approximating 2 years operating time of a nuclear reactor.¹⁰

The radioactivity was calculated² as of the accident date (April 26, 1986). The glands were stored for 10 years, after which ^{129}I was measured by neutron activation analysis.² The data² will be used here to illustrate the derivation of radiation exposure from early measurements and from delayed measurements.

After measurement of ^{129}I , an effort was made to derive ^{131}I from the ^{129}I and compare the result with the measured ^{131}I (10 years previously): The ^{129}I in thyroids, numbers 1A and 3A, showed a mean of 0.24 ng $^{129}\text{I}/\text{g}$ (wet wt of thyroid).² If the reactor had operated⁸ for 660 days, the mass ratio $^{129}\text{I}/^{131}\text{I}$ would have been 16.3. Therefore, the maximum ^{131}I incorporated with the ^{129}I , would have been the reactor equilibrium mixture at the time of the initial release. This would have been $0.24/16.3 = 0.0147$ ng $^{131}\text{I}/\text{g}$ thyroid. Since ^{131}I has 4500 kBq of $^{131}\text{I}/\text{ng}$, the thyroids would have

incorporated a mean of $(0.0147 \times 4500) = 66.2$ kBq $^{131}\text{I}/\text{g}$ thyroid, referred to the beginning of the accident. That would be 1.79 μCi $^{131}\text{I}/\text{g}$ thyroid, which would deliver approximately $1.1 \cdot 10^2$ cGy to the bovine thyroid, if present from the beginning of the accident.

In Table 2, ^{131}I was measured in the thyroids within six weeks after the accident; and when ^{131}I was calculated it was related to ^{131}I as of 26 April 1986, the beginning of the accident. In these two thyroids, the result was the mean of 42 and 95 kBq/g or (68 kBq/g thyroid). This average value (68 kBq/g) was considered similar to the 66 kBq/g estimated above.

Appendix II

Estimation of ^{131}I from measurement of ^{137}Cs

This effort must relate two elements which are physically and biochemically very different. It is necessary to establish empirical factors which may be applied to specific conditions. In this example, we use the same thyroids as in Appendix I. The composition of the initial intake was assumed to be the $^{131}\text{I}/^{137}\text{Cs}$ in a reactor after operating approximately two years¹⁰ (we have used 660 days). According to ORIGEN-2 calculation,⁸ the radioactive ratio $\text{Ci } ^{131}\text{I}/\text{Ci } ^{137}\text{Cs}$ in a PWR-U reactor would be 12.4.

The radioactive ratio of $^{131}\text{I}/^{137}\text{Cs}$ was measured² in the two thyroid glands 1A and 3A, cited in Appendix I. The measured mean ^{131}I was 68 kBq/g $^{131}\text{I}/\text{g}$ and the mean ^{137}Cs was 0.40 Bq/g, giving an empirical ratio, $^{131}\text{I}/^{137}\text{Cs} = 1.7 \cdot 10^5$. The largest part of the overwhelming preference for ^{131}I was caused by the biological concentration factor $1.4 \cdot 10^4$ favoring iodine over cesium. It is within the capacity of the thyroid to concentrate iodine in the gland $1.4 \cdot 10^4$ times greater than

in the natural diet depending, in part, upon the iodine content of the diet. Biological discrimination against ^{137}Cs may be relatively small.

Major biological differences between metabolism of ^{137}Cs and ^{131}I reduce the dependability of quantitative relations between the two isotopes.

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