

A TECHNIQUE FOR ESTIMATING THYROID DOSES  
FROM THE INTAKE OF IODINE-129  
FOR THE  
PROCEEDINGS OF THE THIRD INTERNATIONAL CONGRESS OF IRPA

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Abstract

Recent concern for the environmental buildup of long-lived radionuclides has suggested that dose estimation techniques should be developed for such nuclides. Iodine-129 is a long-lived radionuclide which has recently been found in low concentrations in milk in the environs around a nuclear fuel reprocessing facility. Thus, there was a need to develop a method for estimating the public health impact, in terms of thyroid dose, resulting from the consumption of this milk.

In developing the technique various approaches were considered including specific activity and dietary modeling. It was concluded the most acceptable approach was that using standard man information. The method developed here indicates that for the same level of intake thyroid dose would be only slightly higher from iodine-129 as compared to iodine-131.

Introduction

The projected proliferation of nuclear power reactors implies significantly greater production of radioactive waste products. Currently, the environmental buildup of reactor-produced long-lived radionuclides is causing concern over the health hazards to the world population. One isotope of iodine,  $^{129}\text{I}$  (with a 17 million year half-life), represents an essentially permanent contaminant when released to the biosphere. A normally operating reactor will release very little iodine; however, when the spent reactor fuel is dissolved at a chemical reprocessing plant a portion of the iodine contained in the fuel may be released to the environment.

The critical organ for radioiodine uptake by the general population is the thyroid<sup>1</sup> and the critical pathway considered is ingestion of cow's milk. This pathway is well defined for iodine-131, with its 8-day half life, and should act similarly for iodine-129 when the difference in half lives is considered.

Methods available for thyroid dose calculations include analysis of specific activity, of direct foliar contamination, and dietary and standard man modeling. The first two methods have been discussed by Russell<sup>2</sup> and Bryant<sup>3</sup>. Specific activity analysis may be a very good method for evaluating the impact from the long-term environmental build-up of iodine-129; however, standard man modeling is probably better suited for arriving at action levels. This analysis utilized standard man data and suggests guidance for monitoring operations.

#### Iodine-129 Releases from Reprocessing Plants

The reactor-produced radioactive fission products, including iodine-129, are contained, for the most part, by the fuel cladding. Dissolving the spent reactor fuel at a reprocessing plant removes this cladding barrier and reduces the fission products to a more easily dispersible form. Iodine is particularly susceptible to volatilization and must be removed from the offgas streams. Present technology, using caustic scrubbers in conjunction with silver zeolite, is expected to remove at least 99.8% of the iodine<sup>4</sup>. A typical reprocessing plant will handle 5 metric tons (MT) of fuel a day which contains about  $4 \times 10^{-2}$  Ci <sup>129</sup>I/MT<sup>5</sup>. Expected releases could therefore average  $4 \times 10^{-4}$  Ci <sup>129</sup>I/day.

The first commercial fuel reprocessing plant in the U.S. was operated by Nuclear Fuel Services, Inc. (NFS) at West Valley, New York from 1966 to 1972, when it was shut down for expansion. The State of New York, Department of Environmental Conservation has maintained an extensive environmental sampling program throughout the state. Their milk sampling program produced positive iodine-129 results, ranging from 0.4 to 2.1 pCi/l, in the vicinity of NFS during the first quarter of 1972<sup>6</sup>. These results were higher than previously measured and indicated a need for the more effective iodine control. NFS is currently installing additional cleanup systems, including silver zeolite, to reduce their iodine emissions.

#### Hazard Calculation for Iodine-129 Using Standard Man Data

When considering the public health hazards from iodine-129 the following characteristics are important:

1. The extremely long half-life of  $1.7 \times 10^7$  years, which makes it essentially a permanent environmental contaminant;
2. The physiological concentration of iodine by the human thyroid;
3. The food chain concentration of iodine in cow's milk; and
4. The low energy of the beta and gamma emitters, which make detection and measurement difficult.

The techniques used for calculating thyroid dose from iodine-129 are an adaption of the methods developed by the International Commission on Radiological Protection (ICRP) in their Publication 2 on "Permissible Dose for Internal Radiation"<sup>1</sup>.

Although standard man data are used where applicable, it is modified by considering the 6-months-old infant as the critical individual<sup>3</sup>. In addition, the philosophy and methods promulgated by the former Federal Radiation Council (FRC), whose responsibilities were transferred to the U.S. Environmental Protection Agency (EPA), are utilized for these calculations.

The basic equation<sup>1</sup> for determining the radionuclide concentration in the medium of interest, M, in this case  $\mu\text{Ci }^{129}\text{I}/\text{cm}^3$  milk, is:

$$M = \frac{q f_2 \lambda}{(1 - e^{-\lambda t}) S} \quad (1)$$

where:  $q$  = burden of iodine-129 in the whole body ( $\mu\text{Ci}$ )

$f_2$  = fraction of iodine-129 in thyroid to that in the whole body

$\lambda$  = effective decay constant =  $0.693/\tau_{\text{eff}}$

$\tau_{\text{eff}}$  = effective half-life (days)

$t$  = period of exposure

$S$  = product of the average rate of intake ( $\text{cm}^3/\text{day}$ ) of milk and the fraction of the iodine-129 arriving in the critical organ ( $f_w$ ).

The above equation may be solved more conveniently by combining it with the following equation<sup>1</sup> for the maximum permissible body burden,  $q(\mu\text{Ci})$ , based on a maximum permissible dose rate of  $R$  rem/week:

$$q = \frac{2.8 \times 10^{-3} m R}{f_2 \epsilon} \quad (2)$$

where:  $m$  = mass of thyroid (grams)

$R$  = dose rate (rem/week)

$f_2$  = defined above

$\epsilon$  = effective absorbed energy per disintegration of a radionuclide in the organ of reference (MeV).

The combined equation is as follows:

$$M = \frac{2.8 \times 10^{-3} m R \lambda}{\epsilon (1 - e^{-\lambda t}) S} \quad (3)$$

The assumptions used in solving this equation come from a number of references and are listed below:

1. Mass of the 6-months-old infant thyroid;  $m = 1.8 \text{ grams}^3$
2. Radiation Protection Guide (RPG) for the thyroid, averaged for the general population, of  $0.5 \text{ rem/year}^2$  yields;  $R = .0096 \text{ rem/week}$

3. A biological half-life for iodine in the infant thyroid of 23 days and, since the radiological half-life of iodine-129 is  $1.7 \times 10^7$  years, the effective half-life would be;  $\tau_{\text{eff}} \approx \tau_{\text{bio}} = 23 \text{ days}$ <sup>5</sup>
4. When the period of exposure,  $t$ , is long compared to  $\tau_{\text{eff}}$ , then  $(1 - e^{-\lambda t}) \approx 1$ . This is true after about 120 days
5. Milk intake of one liter per day =  $1000 \text{ cm}^3/\text{day}$ <sup>7</sup>
6. Fraction of iodine-129 ingested reaching the infants thyroid;  $f_w = 0.35$ <sup>3</sup>
7. Fraction of iodine-129 in the thyroid to that in the total body;  $f_2 = 0.2$ <sup>1</sup>
8. The effective energy of iodine-129 in the thyroid has been given as 0.068 MeV by ICRP<sup>1</sup>. For this paper the value has been recalculated using more recent data. Effective energy is assumed to be the sum of the average beta as energy, that portion of the gamma which is converted to directly ionizing particles, and the fraction of emitted x-rays which are absorbed. The average beta energy is 0.40 MeV<sup>8</sup>. The iodine-129 gamma ray (0.038 MeV) is very highly converted, i.e.,  $e_k/\gamma = 22$  and  $K/L \approx 10$ <sup>9</sup>. Therefore, only 4% of the gamma energy remains as unconverted photons. However, the fluorescent yield from the converted electron is large and much of the K x-ray energy escapes from the gland. Summing the net kinetic energy of the converted electrons, the energy given to auger and L electrons and that portion of the K x-rays that is absorbed in the gland yields 17 KeV per disintegration<sup>10,11</sup>. Therefore;  $\epsilon = 0.040 + 0.017 = 0.057$ .

Performing the calculation yields a value of  $0.072 \text{ pCi/cm}^3$  or  $72 \text{ pCi/l}$  for the concentration of iodine-129 in milk which, upon consumption of one liter per day, will deliver an annual thyroid dose of 0.5 rem to the 6-months-old infant.

#### Summary and Conclusions

Using the values calculated above, ranges similar to those defined by the FRC are proposed for iodine-129 concentration in milk. For this purpose 72 picocuries per day was rounded off to 70 picocuries per day. The corresponding ranges of transient rates of daily intake would be:

RANGE I      0 to    7 picocuries per day

RANGE II     7 to    70 picocuries per day

RANGE III    70 to 700 picocuries per day

By applying this reasoning to the New York State data one sees that  $2.1 \text{ pCi/l}$  falls within Range I. Under these conditions it would be appropriate to conduct routine surveillance to assure that levels would not reach Range II without knowledge of the authorities concerned<sup>7</sup>.

From this example it can be seen how this method of calculation might be used to provide guidance for monitoring operations. The problem of the long-term environmental buildup of this radio-nuclide appears to require considerable investigative efforts before any definitive guidance can be developed.

#### References

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