Iodine-129 Dose in LLW Disposal Facility Performance Assessments

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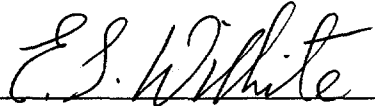# IODINE-129 DOSE IN LLW DISPOSAL FACILITY PERFORMANCE ASSESSMENTS (U)

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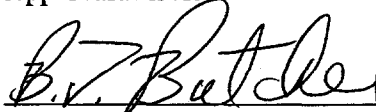
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SUMMARY

Iodine-129 has the lowest Performance Assessment derived inventory limit in SRS disposal facilities. Because iodine is concentrated in the body to one organ, the thyroid, it has been thought that dilution with stable iodine would reduce the dose effects of [129]I.

Examination of the dose model used to establish the Dose Conversion Factor for [129]I shows that, at the levels considered in performance assessments of low-level waste disposal facilities, the calculated [129]I dose already accounts for ingestion of stable iodine. At higher than normal iodine ingestion rates, the uptake of iodine by the thyroid and the mass of the thyroid itself decrease, which effectively cancels out the isotopic dilution effect.

INTRODUCTION

Disposal of low-level radioactive waste (LLW) at DOE facilities must meet the requirements of DOE Order 5820.2A[1]. DOE 5820.2A specifies performance objectives and requires that a radiological performance assessment (PA) be prepared to provide reasonable assurance that the performance objectives will not be exceeded. The PA projects migration of radionuclides from the disposed waste through the environment to points of potential uptake by hypothetical future persons, calculates the dose that could be received by these persons under certain exposure scenarios, and compares the calculated doses with performance objectives. A PA for the SRS Low Level Waste Disposal Facility (LLWDF) has been prepared[2]. The results of the PA are used, along with other information (e.g., Safety Analysis Report) to establish limits on the amount of radionuclides that can be disposed in conformance with performance objectives[3].

Iodine-129 is a fission product with a half-life of about 16 million years. It is very mobile in the environment due to its low retention in sub-surface soil. Iodine-129 has the lowest PA-derived limit on acceptable radionuclide inventory in the LLWDF[2]. Iodine-129 has long been considered a potentially problematic radionuclide in LLW disposal[4, 5, 6, 7, 8, 9, 10].

Due to the long half-life of [129]I, the specific activity (i.e., the radioactivity associated with a given mass of the radionuclide) is low. This, in association with the limited uptake of ingested iodine by the body, has led some to believe that potential doses from [129]I are inherently limited and that [129]I should not be considered a problematic radionuclide in LLW disposal[11, 12, 13, 14, 15, 16, 17, 18, 19, 20].

DISCUSSION

Iodine Radiochemistry

Iodine has 34 isotopes[21] only one of which, [127]I, is stable. Fourteen of the isotopes, including [127]I, are produced in fission[21]. Some properties of the iodine fission product isotopes are listed in Table 1.

Table 1. Iodine Fission Product Isotopes[21]		
Mass Number	Half-life	Fission yield, %
127	stable	0.126
129	1.57×10^7 years	0.75
131	8.04 days	2.89
132	2.3 hours*	4.31
133	20.8 hours*	6.69
134	52.6 minutes*	6.87
135	6.57 hours	6.54
136	1.39 minutes*	6.32
137	24.5 seconds	6.19
138	6.5 seconds	6.71
139	2.30 seconds	6.4
140	0.86 seconds	6.21
141	0.45 seconds	5.8
142	≈ 0.2 seconds	5.84

* Isotope has two isomeric states, the longest half-life is listed

Of the iodine fission products, only [129]I and ^{131}I are potentially significant contributors to radiological exposure of the public from nuclear operations; the half-lives of the other isotopes are too short for them to provide appreciable exposure. Iodine-131 is important in nuclear facility operations[22] and [129]I is potentially significant in waste disposal. The mass of fission product iodine is comprised primarily of [127]I and [129]I; every gram of [129]I produced in fission will be accompanied by 0.16 grams of [127]I.

Iodine Metabolism

Iodine is an essential element in the body[23]. Ingested or inhaled iodine is accumulated by the thyroid gland that produces the hormones thyroxin and triiodothyronine. These hormones are important for regulating the metabolic rate of the body. The amount of iodine required by the body is about 70 μg per day.

Iodine can be toxic[23]. Exposure to iodine vapor irritates the lungs and eyes. The Occupational Safety and Health Act has established a limit on iodine concentration in air in the workplace of 0.1 ppm (1 mg/m³)[24]. Ingestion of iodine in amounts significantly greater than the normal dietary intake can lead to irritation of the gastrointestinal tract; ingestion of gram quantities can be fatal[23]. The state of Maine has set a guideline for iodide in drinking water of 340 μg/L[25].

Iodine-129 Dosimetry

To determine the radiological dose from assimilated iodine radionuclides, the metabolic processes must be simulated. The iodine metabolic model adopted by the International Commission on Radiological Protection is shown in Figure 1[26]. The model assumes an iodine intake of 200 µg/day. It assumes that the thyroid takes up 30% of the ingested iodine. It also assumes that the iodine in the thyroid has a biological half-life of 120 days. Data pertinent for [129]I dose calculations are shown in Table 2.

Figure 1 ICRP Iodine Metabolic Model for Adults

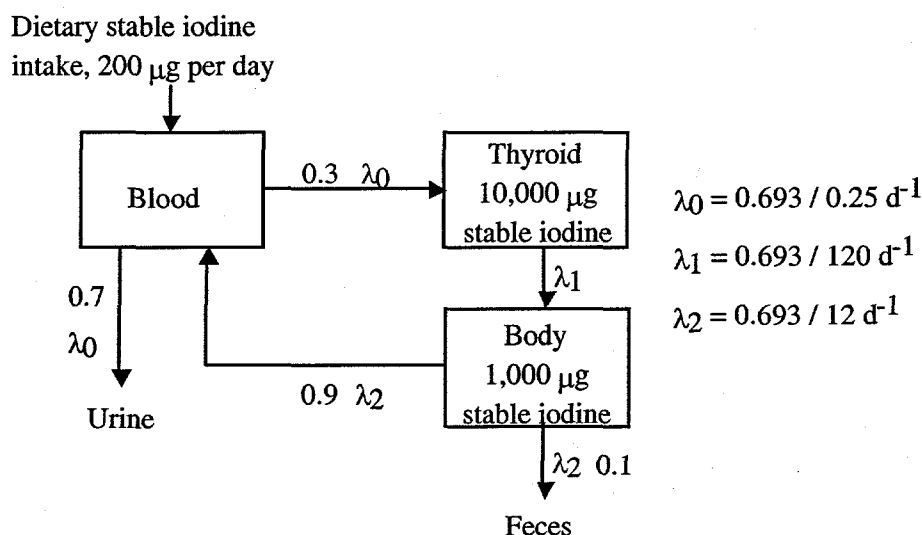Dietary stable iodine intake, 200 µg per day

Blood

$0.3 \quad \lambda_0$

Thyroid 10,000 µg stable iodine

$\lambda_0 = 0.693 / 0.25 \ d^{-1}$

$0.7 \quad \lambda_0$

Urine

λ_1

$0.9 \quad \lambda_2$

Body 1,000 µg stable iodine

$\lambda_1 = 0.693 / 120 \ d^{-1}$

$\lambda_2 = 0.693 / 12 \ d^{-1}$

$\lambda_2 \quad 0.1$

Feces

Table 2. Standard Parameters for [129]I Dose Calculation	
Parameter	Value
Radioactive half-life	1.57×10^7 years[27]
Specific Activity	1.77×10^{-4} Ci/g[28] or 5,650 g/Ci
Specific Effective Energy	0.068 Mev per disintegration[27]
Mass of thyroid	20 grams[29]
Fractional uptake of ingested iodine by thyroid	0.3^{30}
Mass of iodine in thyroid	10 mg[26]
Thyroid dose weighting factor	0.03^{26}
Dose conversion factor	0.28 Rem/µCi intake[31, 32]

<u>Limiting ^{129}I Dose</u>

Because of the relatively large iodine mass associated with ^{129}I and the restricted uptake of ingested iodine by the thyroid, the dose from ingested ^{129}I is inherently limited. The limiting dose would be received when every atom of iodine in the body was an atom of ^{129}I. Because of the limited uptake of ingested iodine and the low specific activity of ^{129}I, this situation would only happen as a consequence of prolonged intake of pure ^{129}I. Assuming that the daily intake of 200 µg of iodine was all ^{129}I, the dose would be calculated as follows:

$$200 \text{ µg } ^{129}\text{I} * 10^{-6} \text{ g/µg} * 1.77 \text{x} 10^{-4} \text{ Ci/g} * 10^6 \text{ µCi/Ci} = 0.035 \text{ µCi } ^{129}\text{I ingested/day}$$

$$0.035 \text{ µCi } ^{129}\text{I/day} * 365 \text{ day/year} * 0.28 \text{ rem/µCi} = 3.6 \text{ rem/year}$$

While 3.6 rem/year is not a life-threatening dose, it is about 1,000 times greater than the groundwater protection performance objective[2], which must be met for LLW disposal.

<u>Other Studies</u>

Iodine-129, Limits to Radiologic Dose

An earlier study[11] considered the limitation of ^{129}I dose due to the reduction of iodine uptake by the thyroid as the mass of ingested iodine increased. This effect is illustrated by calculating the dose from ingesting ^{129}I and ^{131}I, assuming a constant ingestion of ^{127}I of 200 µg/day. Figure 2 shows the dose, in mrem per nCi ingested as a function of the amount of the radionuclide ingested. The calculated dose is adjusted[33] to account for the suppression of iodine uptake by the thyroid when total iodine intake exceeds 200 µg/day. Iodine-131, which has a half-life of only 8 days (specific activity of $1.2 \text{x} 10^5$ Ci/g) does not contribute enough mass to change the daily iodine intake.

The dose per nCi of ^{129}I shown in Figure 2 remains constant up to about 70 nCi. Ingestion of 70 nCi of ^{129}I would result in a dose of 20 mrem. The performance objectives for LLW disposal[1] are 4 mrem/year (groundwater protection), 25 mrem/year (all-pathways to member of public), and 100 mrem/year (inadvertent intruder). In the LLWDF PA[2], the groundwater protection performance objective of 4 mrem/year was limiting for ^{129}I. Since 4 mrem/year is less than the 20 mrem inflection point derived in this study, the reduction of calculated dose to account for the mass of ingested ^{129}I would not significantly affect the dose calculated in the PA.

However, this study did not take into account the concomitant reduction in thyroid mass associated with increased iodine ingestion[33].

Figure 2 Calculated Dose versus Amount of Radioactive Iodine Ingested
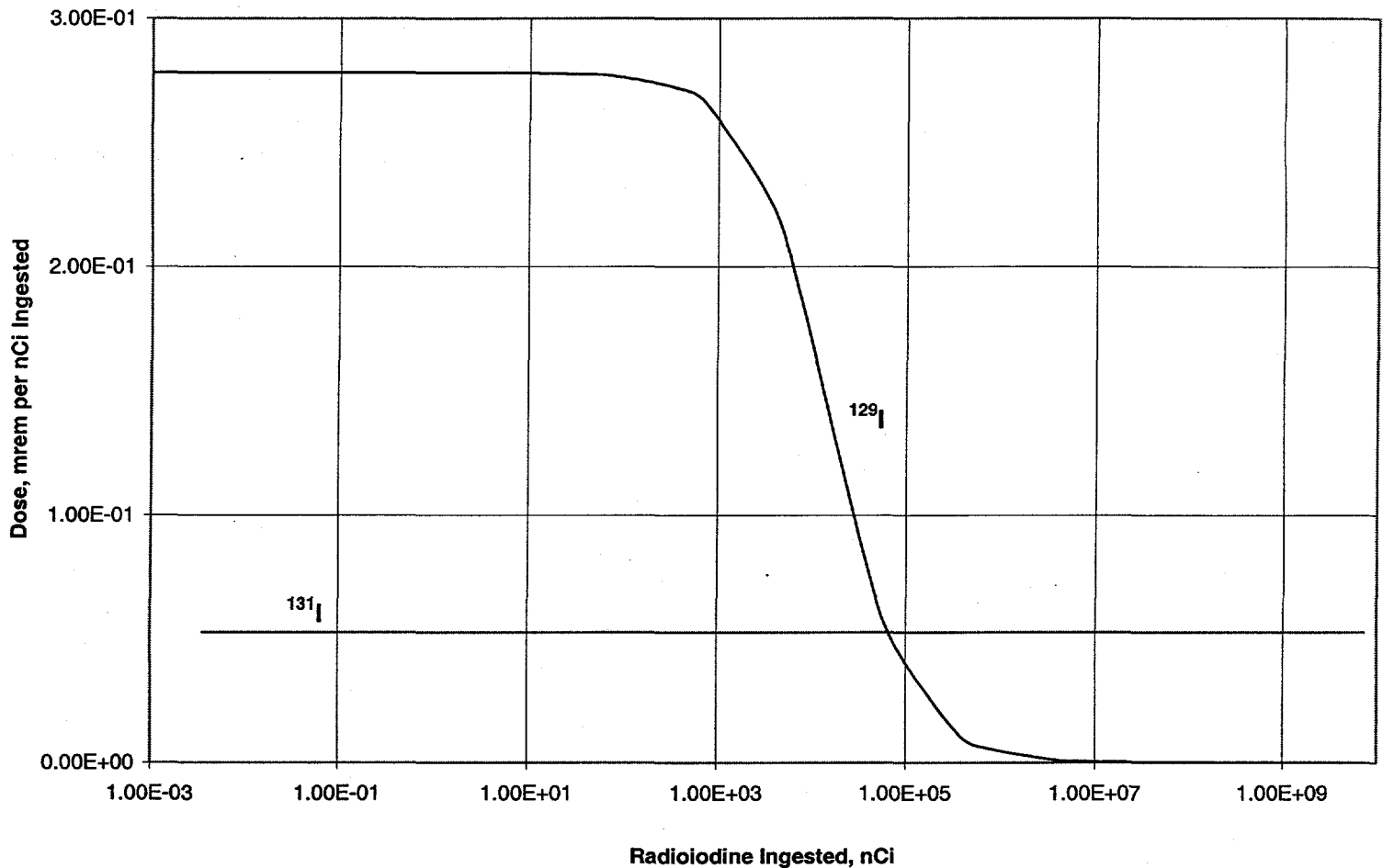
Isotopic Equilibrium Analysis

In a study of long-lived radionuclides in SRS LLW[34], Cohen et al. applied isotopic equilibrium analysis (IEA) to suggest that "I-129 from SRP wastes is not a significant problem". Following, with updated ICRP information, is the argument.

A study of ^{99}Tc and ^{129}I in groundwater near the SRS Old Burial Ground (OBG)[35] showed the following results (Data from well BG-109 were omitted because the ^{127}I value was less than the detection limit).

Table 3. Iodine Analyses of SRS Groundwater					
Well ID	PDQ-5	BGC-2C	BGC-3C	I-13	Geometric Mean
^{127}I, µg/L	1190	99	166	49	176
^{129}I, pCi/L	12.0	0.033	0.92	0.006	0.22
^{129}I, µg/L	6.8×10^{-2}	1.9×10^{-4}	5.2×10^{-3}	3.4×10^{-5}	0.0012
^{129}I/^{127}I ratio	5.7×10^{-5}	1.9×10^{-6}	3.1×10^{-5}	6.9×10^{-7}	7.0×10^{-6}

If a person's only source of iodine were this SRS groundwater, eventually all the iodine in the person's body would have the same ^{129}I/^{127}I ratio as the groundwater, 7.0×10^{-6}. The resulting dose, according to the IEA method would be:

$$3.62 \text{ rem/year} * 7.0 \times 10^{-6} * 1,000 \text{ mrem/rem} = 0.025 \text{ mrem/year}$$

Although this study is interesting, the results are not pertinent to performance assessments since they are based on observed ^{129}I concentrations in groundwater rather than those calculated in the PA. It does, however, suggest that isotopic dilution may diminish doses calculated for ^{129}I. In the example above, the person consumes water containing 0.22 pCi ^{129}I per liter. Assuming, as in a PA, the consumption of 2 liters of water per day, the person would consume 160 pCi of ^{129}I in a year. Using the ^{129}I dose conversion factor of 0.28 rem per µCi and converting units, the dose would be 0.045 mrem/year. This suggests that, taking into account the dilution of ^{129}I by stable iodine, the dose would be reduced by about a factor of 2.

Dietary Intake of Stable Iodine and Some Aspects of Radioiodine Dosimetry

Zvonova[33] showed that the uptake of iodine by the thyroid decreased as the dietary intake increased. Her study also showed that thyroid sizes measured in different geographic locations varied inversely with the quantity of iodine in the diet. Zvonova's[33] data are shown in Figure 3. At an iodine intake of 200 micrograms per day, the data in Figure 3 show that the iodine uptake by the thyroid would be about 30% and the thyroid mass would be about 20 grams; these values are consistent with the values used by the ICRP in calculating dose from radioisotopes of iodine.

As a first approximation, iodine intake of 200 µg/day results in a thyroid mass of 20 grams, while an iodine intake of 400 µg/day results in a thyroid mass of 10 grams. The radiological dose from ^{129}I is due almost entirely to a low energy beta particle that has a very short range in tissue; thus, the dose is deposited only in the thyroid tissue. Since the dose is derived from the energy deposited in a particular mass of affected tissue, it is dependent upon the concentration of iodine in the thyroid and remains constant over a reasonable range of iodine intake.

The radiation dose due to ^{129}I is largely independent of the quantity of iodine in the diet. This conclusion was supported in a personal communication with a recognized expert[36].

Figure 3 Iodine Uptake by Thyroid and Thyroid Mass vs Iodine Intake
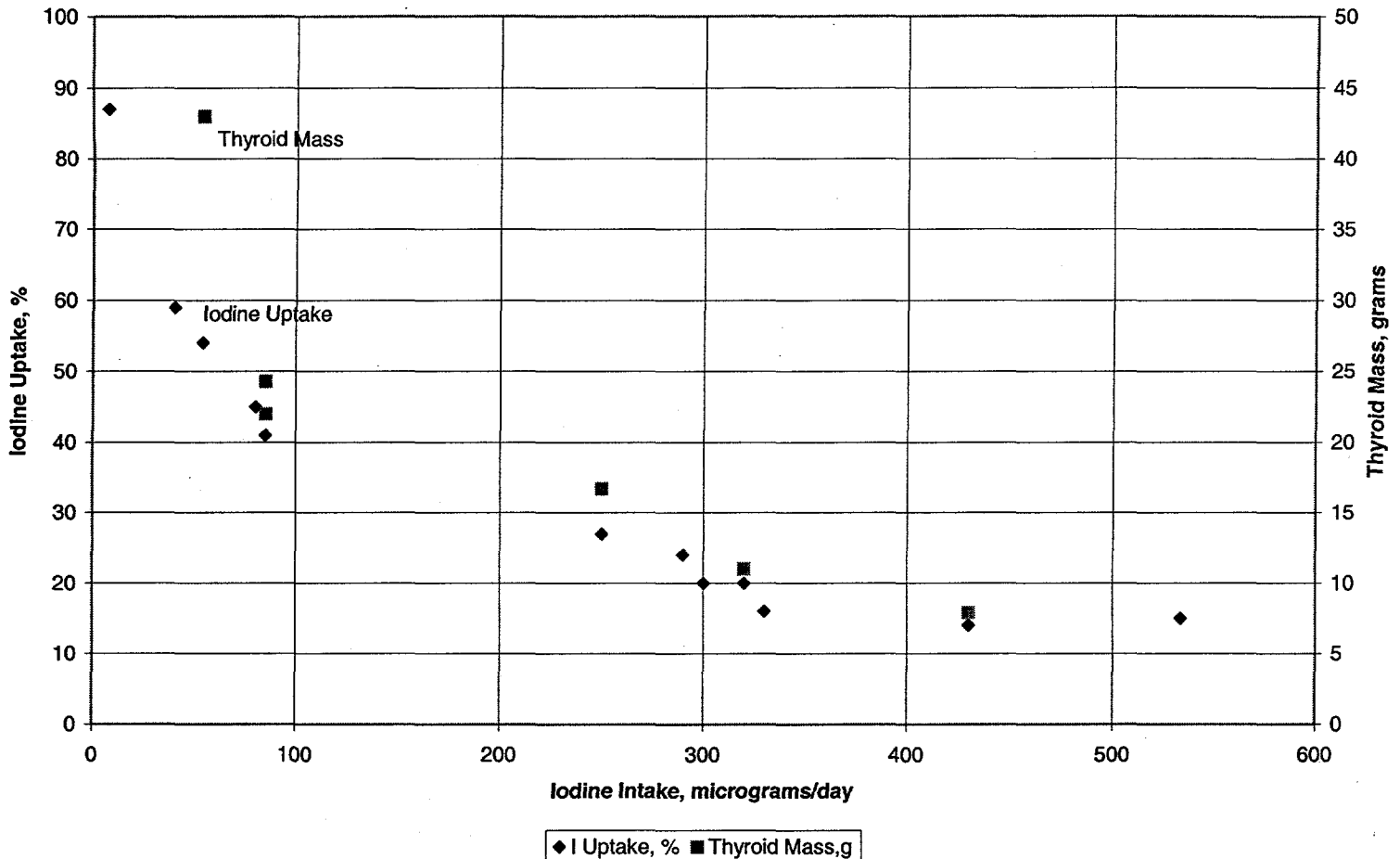
Isotopic Dilution of ^{129}I

Physiological processes cannot discriminate between radioisotopes of iodine. Thus, assuming chemical equilibrium between iodine released from disposed waste and iodine present in the groundwater (i.e., all of the iodine is in the same chemical form), the iodine taken up by the thyroid from ingested groundwater will have the same isotopic (i.e., ^{129}I/^{127}I) mass ratio as that in the groundwater. Assuming long-term ingestion of the groundwater, and the groundwater is the predominant source of iodine in the diet (i.e., the assumptions in performance assessments of disposed LLW) the iodine in the thyroid would eventually have the same isotopic mass ratio as the groundwater.

The dose from ingestion of ^{129}I can be expressed as the isotopic mass ratio of ^{129}I to the total iodine ingested in a year. The standard metabolic parameters (i.e., daily ingestion of 200 µg iodine, fraction of ingested iodine taken up by the thyroid of 0.3, dose conversion factor of 0.28 rem per µCi ^{129}I ingested) from Table 2 are assumed. Table 4 shows selected values.

Table 4. ^{129}I Dose and Isotopic Mass Ratio			
Dose, mrem	µCi ^{129}I ingested	µg ^{129}I ingested	g ^{129}I per g I
0.01	3.6×10^{-5}	0.2	2.8×10^{-6}
0.025	8.9×10^{-5}	0.5	6.9×10^{-6}
0.1	3.6×10^{-4}	2.0	2.8×10^{-5}
1	3.6×10^{-3}	20.	2.8×10^{-4}
2	7.1×10^{-3}	40.	5.5×10^{-4}
3	1.1×10^{-2}	60.	8.3×10^{-4}
4	1.4×10^{-2}	81.	1.1×10^{-3}
10	3.6×10^{-2}	200	2.8×10^{-3}
20	7.1×10^{-2}	400	5.5×10^{-3}
30	1.1×10^{-1}	600	8.3×10^{-3}

This analysis suggests that, if the disposed waste has an isotopic mass ratio of ^{129}I of less than 0.001 g ^{129}I per gram of iodine, the dose, in the drinking water scenario, from ^{129}I cannot exceed 4 mrem/year. For example, a dose of 4 mrem/year, calculated via the drinking water scenario, requires that the water contain 20 pCi ^{129}I per liter. This is equivalent to 0.1 µg ^{129}I per liter. The drinking water scenario assumes that a person consumes 2 liters of water per day for a year. The ICRP metabolic model for iodine assumes a daily ingestion of 200 µg of iodine, or, in the drinking water scenario, a water concentration of 100 µg iodine per liter. Thus, in this example, the ^{129}I isotopic mass ratio is 0.001.

This isotopic dilution, however, will not affect doses from ^{129}I calculated in a PA, if the constraints of the iodine metabolic model are adhered to. Assume the isotopic mass ratio in groundwater were 2.8×10^{-4}, per table 4 this would be commensurate with a dose of 1 mrem/year. Also assume the groundwater contained 0.1 µg ^{129}I per liter, commensurate with a dose of 4 mrem/year. Thus, the groundwater would contain 357 µg of iodine per liter. The drinking water scenario requires that the person consume 2 liters of water per day. The ingested water would contain 714 µg of iodine. Because this is several times greater than the normal daily ingestion of 200 µg, the absorption of iodine by the thyroid will decrease, as shown by Zvanova[33]. Assuming a linear proportionality, the fraction of ingested iodine absorbed by the thyroid would be about 0.08. In other words, over the span of the year's ingestion of groundwater, the thyroid would take up

$$0.1 \mu g\ ^{129}I/L * 2L/day * 365 day/year * 0.08 = 5.8\ \mu g\ ^{129}I$$

This is only 27% of the ^{129}I taken up by the normal thyroid to yield a dose of 4 mrem/year. However, the increased iodine ingestion not only decreases the iodine uptake by the thyroid, it also causes the thyroid mass to decrease. Again, according to Zvanova's data, assuming a linear proportionality, the decrease in thyroid mass would be equivalent to the decrease in iodine uptake. Therefore, the calculated dose from ^{129}I would be the same. This demonstrates that isotopic dilution cannot be used to reduce doses calculated for ^{129}I in a PA.

Conclusion

The above analyses show that the maximum dose from ^{129}I ingestion is limited by the isotope's low specific activity. However, they also demonstrate that, at the relatively low levels of ^{129}I ingestion addressed in a PA, isotopic dilution does not limit the dose from ^{129}I. Thus, adjustment of ^{129}I dose calculated in a PA to account for isotopic dilution is not appropriate.

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