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NATURAL IODINE AND IODINE-129 IN MAMMALIAN THYROIDS AND ENVIRONMENTAL SAMPLES TAKEN FROM LOCATIONS IN THE UNITED STATES

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LIST OF FIGURES

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Abstract

Iodine-129 is present in the environment from natural and man-made sources. It is produced by spontaneous fission of natural uranium and spallation reactions in the upper atmosphere. It is also one of the fission products found in the fallout of debris from nuclear tests. As a fission product ^{129}I is also produced in nuclear reactors and can be released either at the reactor or later at the irradiated fuels reprocessing plant. Because of its long half-life (1.6 x 10^7 years), ^{129}I is an excellent tracer for environmental processes involving other stable or radioactive isotopes of iodine. Environmental concentration measurements of stable natural iodine (^{127}I) and long-lived radioactive iodine (^{129}I) have been made at our laboratory.

Bovine and other mammalian thyroids, air, precipitation, water, grass, and milk samples from selected locations have been analyzed for their natural iodine and iodine-129 content. Particle filters and activated charcoal beds were used to collect the particulate and gaseous iodine fractions from the air. Anion exchange was used for the separation of iodine from liquid samples. Combustion methods were used for iodine separation from solid samples, anion exchange resins, and air filters. Iodine-129 and natural iodine concentrations were determined by neutron activation analysis with a detection limit of about 10^8 atoms of iodine-129 per sample.

Analytical results of both ^{129}I concentrations and ratios of atoms of ^{129}I per atom of ^{127}I for these samples are summarized in this paper. The small radiation doses to human thyroids projected from these low ^{129}I concentrations are also discussed.

INTRODUCTION

Numerous studies of the levels of radioactive materials in the environment have been undertaken to gain a knowledge of the release, dispersion, and accumulation of these materials from a variety of sources - natural background, fallout and nuclear facilities. The results of these measurements are also required for calculation of present and future radiation doses to people and biota. When performing these studies and dose calculations, materials with long half-lives must be considered because they are not removed by radioactive decay and can buildup in the environment. Several authors have published studies on the permissible concentration of iodine-129 (half-life 1.6 x 10^7 years) in the environment.[1-4] Knowledge of both the ^{129}I concentration and the ratio of ^{129}I to natural stable iodine, ^{127}I, in various environmental materials is required for proper assessment of the radiological impact of ^{129}I.

The long half-life (1.6 x 10^7 years), low specific activity (385 dpm per microgram), and low radiation energy limits the sensitivity of ^{129}I measurement obtained by counting the ^{129}I radiation to about 10 pCi. Neutron activation analysis can be used to measure environmental ^{127}I and ^{129}I levels with sensitivities of about 4 x 10^{-6} pCi for ^{129}I. Neutron activation analysis for ^{129}I was first applied to the analysis of uranium ore samples;[5] Studier et al. developed a method for isolation of iodine from a variety of biological materials and its subsequent iodine concentration and isotopic analysis by neutron activation.[6] Several authors have published measurements of environmental concentrations of ^{129}I by neutron activation analysis.[7-12] The use of mass spectrometry for low-level iodine isotopic analysis has also been reported.[13]

Iodine-129 is present in the environment from natural and man-made sources. An evaluation of the man-made contributions of environmental ^{129}I levels requires some knowledge of the natural production of ^{129}I. The production of ^{129}I in uranium ores by spontaneous fission and (α,n) induced fission has been measured by Purkayastha and Martin.[5] Edwards has summarized the various natural processes which can release ^{129}I to the environment.[9,14,15] The principal natural production processes are spontaneous fission of uranium and cosmic-ray-produced spallation reaction with atmospheric xenon. Because of the nature of these processes the natural $^{129}I/^{127}I$ will differ for various natural sources. Thus old mineral iodine, not in contact with the atmosphere or uranium, contains less ^{129}I than pre-1945 environmental materials that had been in contact with the atmosphere and hydrosphere. Values reported for $^{129}I/^{127}I$ in mineral iodine range from 1 x 10^{-11} to 2 x 10^{-12};[9] values reported for pre-1945 thyroid tissue range from 2.5 x 10^{-9} to 1 x 10^{-10}.[7,15]

Neutron induced fission in nuclear reactors and nuclear weapons also produces ^{129}I. Thus fallout from nuclear weapon testing introduced additional ^{129}I into the environment. The ^{129}I produced in nuclear reactors can also be partially released to the environment especially during irradiated fuel reprocessing and waste handling operations.

The levels of ^{129}I and ^{127}I in the environment of the Hanford AEC facilities have been studied for a number of years.[11] Levels at locations remote from the Hanford facilities have also been studied to arrive at background levels in the environment. This paper summarizes the results of these ^{129}I and ^{127}I analyses obtained for thyroid tissue, milk, grass, water and air samples and discusses their significance.

SAMPLE COLLECTION

Studies of ^{129}I levels in the environs of the USAEC Hanford, Washington Reservation started with the collection of vegetation samples in the 1950's. Since iodine is concentrated in thyroid tissue, collection of animal thyroids began after consideration of the results of the analysis of the local vegetation samples. Thyroids from sheep, which were raised on the Hanford Reservation but which were not being fed radioactive tracers, were obtained in 1962 for iodine isotopic analysis. Several local hog thyroids were obtained in 1963, and thyroids from both local and distant wild deer, elk and antelope were collected in 1963 and 1964 for ^{129}I analysis.

Bovine thyroids were routinely collected for ^{131}I analyses over a broad geographical area by the U.S. Public Health Service from 1964 to 1969.[16] A number of these thyroid samples were made available to our laboratory for ^{129}I analyses after the nondestructive ^{131}I measurements had been completed. Local bovine thyroids collected for ^{131}I analysis by gamma-ray spectrometry in the routine Hanford environmental surveillance program were retained for ^{129}I analysis. Arrangements were also made to routinely receive bovine thyroids from Klamath County, Oregon for studies of the ^{129}I background levels from fallout.

Samples of pre-1936 human thyroid tissue were received from the AEC and a sample of pre-1942 I_2O_5 was received from the University of Chicago for measurement of the levels of ^{129}I.

The collection of surface water and air samples was initiated in 1965. Initially water samples were collected in large plastic carboys and returned to the laboratory for filtration and iodine separations. More recently, filtration and ion-exchange separation of iodine have been accomplished in the field as the water samples were collected.[11]

Large volume air sample collection was initiated at the Pacific Northwest Laboratory near Richland, Washington in 1965, at Spokane, Washington in 1966, and at Neah Bay on Washington's Olympic Peninsula in 1967. The samples were collected by passing 3,000 to 60,000 cubic meters of air through a membrane filter (mean pore size of 5 to 7 micrometers) backed with an activated charcoal bed. Positive displacement pumps were used to maintain a flow rate through the filters and charcoal of about 3,000 m^3/day with an air surface velocity of 150 to 300 cm/sec through the charcoal bed. Normal sample collection periods were at two-week intervals, but other intervals were occasionally used. A sufficient number of samples were collected at each location to assure a seasonal average for several years.

Airborne particles were collected on the membrane filters while volatile iodine species were collected on the activated charcoal bed. The 2.5-cm-deep activated charcoal beds used on our samplers did not trap and retain all the volatile iodine. About the same amount of iodine has been found in the front one-half of bed depth as in the back one-half of bed depth. Several experiments suggest that the fraction of iodine penetrating the charcoal bed increases with the number of days over which the samples were collected. Some iodine desorption is probably responsible for this effect.

Since 1967 several milk samples have been collected for analysis from small dairies in the area. Although the source of the cattle feed is not known for all of the samples, the feed for the cattle near the Hanford Reservation was probably locally produced.

SAMPLE ANALYSIS

The activiation analysis method developed by Studier, et al.[6] was used for the ^{129}I and ^{127}I analysis in these studies. The analytical method includes procedures for isolation of the iodine from the sample, irradiation of the separated iodine with neutrons to produce ^{126}I, ^{128}I, and ^{130}I, decontamination of the irradiated sample, measurement of the induced radioactivity in the iodine sample, and calculation of the ^{127}I and ^{129}I content. Prior to analysis some of the grass samples were freeze-dried to remove water and the thyroid glands were trimmed to remove non-thyroidal tissue. A known amount of ^{125}I spike was added to the sample prior to the iodine separation. Ionic iodine was concentrated from large liquid samples such as water and milk by absorption on a Dowex 1 ion-exchange resin column. Solid samples or the resin beds from liquid sample processing were placed in a quartz combustion apparatus and ignited at high temperatures (up to 1000°C) in a stream of oxygen for isolation of the iodine. The iodine was collected on a small bed of activated charcoal. Combustion of the charcoal trap in oxygen and trapping of the iodine on activated charcoal was used for further purification of the iodine. The iodine was then removed from the charcoal by heating in a vacuum system, trapping the iodine in a quartz tube at liquid nitrogen temperature, and sealing the tube to make a quartz irradiation ampoule.

Quartz ampoules containing the iodine separated from the samples were irradiated with reactor neutrons for eight to twenty-four hours. Comparator standards containing ^{125}I, ^{127}I, and ^{129}I were irradiated with each set of samples.

The neutron capture reactions used for the iodine activation analysis were:

(1) $^{127}I(n,\gamma)^{128}I \xrightarrow[25\ min]{\beta^-,\gamma} {}^{128}Xe$

(2) $^{127}I(n,2n)^{126}I \xrightarrow[13\ day]{\beta^-,\gamma} {}^{126}Xe$

(3) $^{129}I(n,\gamma)^{130}I \xrightarrow[12.4\ hr]{\beta^-,\gamma} {}^{130}Xe$.

Interfering reactions include:

(4) $^{125}I(n,\gamma)^{126}I \xrightarrow[13\ day]{\beta^-,\gamma} {}^{126}Xe$

(5) $^{127}I(n,\gamma)^{128}I(n,\gamma)^{129}I(n,\gamma)^{130}I \xrightarrow[12.4\ hr]{\beta^-,\gamma} {}^{130}Xe$

(6) $^{235}U(n,fission)^{131}I \xrightarrow[8\ day]{\beta^-,\gamma} {}^{131}Xe$

$^{132}I \xrightarrow[2.3\ hr]{\beta^-,\gamma} {}^{132}Xe$

$^{133}I \xrightarrow[21\ hr]{\beta^-,\gamma} {}^{133}Xe \xrightarrow[5.3\ day]{\beta^-,\gamma} {}^{133}Cs$

$^{134}I \xrightarrow[52\ min]{\beta^-,\gamma} {}^{134}Xe$

$^{135}I \xrightarrow[6.7\ hr]{\beta^-,\gamma} {}^{135}Xe \xrightarrow[9.2\ hr]{\beta^-,\gamma} {}^{135}Cs$

(7) $^{81}Br(n,\gamma)^{82}Br \xrightarrow[36\ hr]{\beta^-,\gamma} {}^{82}K$.

Following irradiation the iodine samples were further purified by distillation and solvent extraction. The samples were finally precipitated as AgI and mounted on thin plastic scintillators for counting.

The ^{126}I, ^{128}I and ^{130}I activities produced in the sample and comparator standards during irradiation were estimated by gamma-ray spectrometry using several spectra collected over time to obtain half-life information. Well-type NaI(Tl) detector systems were used for the measurements. The ^{130}I sum peaks at 1.9 MeV and 2.3 MeV were used to estimate the ^{130}I activity and to discriminate against interferences. The low-level, beta-gated, multiple-gamma coincidence spectrometry techniques described in the literature[11,17-20] were used, when required, to measure very small amounts of ^{130}I and to correct for interfering activities such as those produced by reactions (6) and (7). Post-irradiation chemical processing also limited interference from reaction (7).

Interference from reaction (4) was minimized by use of small activity levels of ^{125}I which allowed reaction (2) to predominate. Reaction (5) limited the increased sensitivity that could be obtained by increasing the exposure time and neutron flux. The neutron exposure conditions were selected, based on the expected natural iodine content of given sample types, to limit the correction required due to reaction (5) to less than 10%.

The low-level ^{125}I measurements required to estimate the chemical yield of the sample iodine and to measure the amount of the ^{127}I and ^{129}I in the irradiated comparator standard were performed with the special gamma-x-ray coincidence system described in reference 17.

The $^{129}I/^{127}I$ in the comparator standard was calibrated by activation analysis relative to a mixed ^{129}I-^{127}I standard prepared by dilution of mass-spectrometric-analyzed ^{129}I with natural iodine.[21] The amount of natural iodine relative to the ^{125}I activity in the comparator standard was determined by activation analysis with a C_6I_6 standard.

The components in the time-dependent gamma-ray spectra resulting from the measurements on a sample were calculated by a weighted least squares method.[20] The data handling programs outlined in reference 21 were used for calculation of analytical results from the sample and the standard and for preparation of final data tabulations. Corrections were made for interferences, chemical yield, laboratory blanks, and sampling blanks where applicable. The results obtained were the ^{127}I and ^{129}I concentrations per unit amount of sample and the $^{129}I/^{127}I$ ratio. The detection limit for ^{129}I is about 4 x 10^{-6} pCi per sample.

Several precautions should be noted in interpretation of the analysis of some sample types. When ion exchange is used to separate iodine from liquid samples, only the ionic iodine is separated. Organic bound iodine may not be completely separated from the sample. The same is true for air sampling. The air sampler used for these studies did not necessarily completely trap all forms of atmospheric iodine.

RESULTS AND DISCUSSION

Estimates of the ^{129}I environmental levels in pre-atomic era samples are required to evaluate the man-made contributions to the environment. Table I summarizes values reported in the literature and measurements by the authors on several iodine samples from pre-1945 thyroid sources, mineral sources and laboratory reagent sources. When several values were reported or obtained on the iodine source, they are shown as a range. The data show that the isotopic $^{129}I/^{127}I$ in pre-atomic era thyroid glands was probably less than 10^{-9}. Mineral iodine sources contain an even lower concentration of ^{129}I.

Table II summarizes iodine concentration and isotopic analysis results for sheep and deer thyroids collected from animals which lived on, near, and remote from the Hanford Reservation. The concentration results are

TABLE I. SUMMARY OF $^{129}I/^{127}I$ VALUES MEASURED FOR PRE-1945 THYROID TISSUE, REAGENT AND MINERAL IODINE SAMPLES

Iodine Source	Year	$^{129}I/^{127}I$ Atom Ratio (10^{-9})	Reference
Composite Human Thyroids	Pre-1945	2.5 ± 0.9	[7]
Human Thyroid	Pre-1936	≤0.04	This report
Hog Thyroid	Pre-1945	0.8 ± 0.2	[7]
Hog Thyroid	Pre-1945	~0.1	[15]
Brine, Long Beach, California	1930	0.005 to 0.01	[9]
Brine, Michigan	1966	0.002 to 0.003	[9]
Chile Nitrate	1939	0.005	[9]
Chile Nitrate	1966	0.0003 to 0.003	This report
Merck I_2	1920	0.005	[9]
Merck I_2O_5	Pre-1942	0.004 ± 0.004	This report
I_2 Reagent	Pre-1954	≤0.002	This report
Reagent	Not given	0.06 to 0.18	[12]

TABLE II. SUMMARY OF THE RESULTS OF ANALYSES FOR ^{127}I AND ^{129}I IN THYROID TISSUES OTHER THAN BOVINE

Location	Year	Number of Thyroids	Type	^{127}I Concn (mg/g of Thyroid)			^{129}I Concn (10^{-3} pCi/g of Thyroid)			^{129}I/^{127}I Atom Ratio (10^{-9})		
				Avg	Min.	Max.	Avg	Min.	Max.	Avg	Min.	Max.
Hanford Reservation, Wash.	1962	11	Sheep	0.51	0.12	1.2	420	140	905	5700	861	9050
Wyoming	1964	3	Sheep	1.7	1	2.4	34	17	49	100	87	112
Walla Walla, Wash.	1963	2	Hog	2.6	2.6	2.6	7	7	7	15	15	15
Ringold, Wash.	1963	1	Hog				330					
Maryland	1963	2	Deer	3.4			23	6.4	40	65		
Blue Mountains, Ore.	1963	2	Adult Deer	0.48	0.23	0.72	400	276	525	5400	4061	6721
Blue Mountains, Ore.	1963	4	Fetus from above	0.34	0.13	0.56	420	142	681	7000	6038	8792
Hanford Reservation, Wash.	1964	2	Adult Deer				8600	6360	10800			
Hanford Reservation, Wash.	1964	4	Fetus from above	0.29	0.22	0.34	2000	1610	2370	39000	31000	44000
Cascade Mountains, Wash.	1964	1	Elk	0.9			320			1900		
Quinault, Wash.	1964	2	Elk	1.5	0.88	2.2	27	15	42	100	66	148
Wyoming	1963	4	Elk	4.7	3	7.6	160	97	248	180	180	185
Wyoming	1964	2	Antelope	2.4	0.6	4.2	80	20	140	190	184	194
United States	Pre-1936	5	Human							≤0.04		

listed in units of milligrams of ^{127}I per gram of thyroid tissue and picocuries of ^{129}I per gram of thyroid tissue. Average, minimum and maximum values are listed when results from more than one thyroid were obtained. All the animal thyroid samples were found to contain considerably more ^{129}I than that observed in the pre-1936 human thyroid tissue. The highest ^{129}I concentration and ^{129}I/^{127}I atom ratios were observed in samples collected on or near the Hanford Reservation. Fetal thyroid tissue ^{129}I levels were found to be similar to the levels observed in the mothers.

Results of iodine analyses on cattle (bovine) thyroid samples are summarized by states in Table III. The Eastern Washington samples were all collected within 100 miles of the Hanford Reservation and analyzed as composites of fourteen to forty-one thyroids from the same month. The samples collected in Northeastern Oregon near the Hanford Reservation are listed separately from the Southern Oregon samples because of the large differences in ^{129}I content in thyroids from these two locations. Median values as well as average values are given for ^{129}I/^{127}I. Some of the thyroid samples were analyzed separately and some samples were run by compositing two to nine thyroids from the same state and on the same date. No apparent trends in the data with date sampled were observed. The dependence of the ^{129}I/^{127}I results on geographic location is shown in Fig. 1. The highest ^{129}I concentrations and ^{129}I/^{127}I ratios were observed in locations nearest the AEC reservations at Hanford, Washington and Savannah River, South Carolina. The ^{129}I content of all areas, however, appeared to be higher than pre-1945 levels.

Iodine analysis results are listed by county in Table IV for bovine thyroids collected in Eastern Washington and Umatilla County, Oregon. The ^{129}I concentration and ^{129}I/^{127}I atom ratios observed in a single county were found to vary by a factor of 1000. Fig. 2 shows the locations from which the samples were collected and the ^{129}I/^{127}I estimates. The Hanford Reservation is located in Benton County.

The average air concentrations of ^{127}I and ^{129}I for three locations in the Northwestern United States are listed in Table V. The highest measured concentrations of ^{129}I were observed near Richland, Washington. Lower concentrations were observed at Spokane, Washington located about 120 miles from the Hanford Reservation. Significantly lower concentrations were obtained on the west side of Washington's Olympic Peninsula which is upwind and separated from the Hanford Reservation by two mountain ranges. These latter values can be used as indicative of background ^{129}I levels. The gaseous (charcoal absorbed) species of iodine in the atmosphere were found to represent more than 50% of the atmospheric ^{127}I and ^{129}I collected by the samplers at all locations.

Table VI lists iodine measurements obtained on precipitation samples. The highest ^{129}I/^{127}I values were observed at Eastern Washington locations.

TABLE III. SUMMARY BY STATES OF THE RESULTS OF ANALYSES FOR ^{127}I AND ^{129}I IN BOVINE THYROID TISSUE COLLECTED FROM 1964 TO 1969

State	Year	Number of Thyroids	^{127}I Concn (mg/g of Thyroid)			^{129}I Concn (10^{-3} pCi/g of Thyroid)			$^{129}I/^{127}I$ Atom Ratio (10^{-9})			
			Avg	Min.	Max.	Avg	Min.	Max.	Median	Avg	Min.	Max.
Arizona	1967	1	1.7			0.42				1.3		
California	1965-1967	54	2.2	0.63	5.3	5.3	0.20	44	7	18	0.55	240
Colorado	1965-1967	13	1.8	0.41	4.2	3.6	1.0	12	13	14	2.1	55
Georgia	1965-1967	29	1.9	0.78	5.8	210	0.55	908	268	690	2.0	2650
Iowa	1965-1966	5	2.3	0.48	3.5	4.3	1.6	10	13	14	3.9	35
Idaho	1965-1967	45	1.9	0.46	4.2	17	0.84	99	58	73	3.3	528
Illinois	1966	1	3.2			7.9				14		
Indiana	1967	1	3.7			0.28				0.4		
Kansas	1966	2	5.1	2.2	7.9	8.1	2.1	14		7.6	5.4	9.9
Minnesota	1965-1966	6	2.7	0.89	5.7	3.2	0.72	9.7	8	12	0.70	45
Mississippi	1965-1966	32	1.6	0.31	3.3	11	3.3	23	30	38	13	71
North Carolina	1966-1969	6	2.8	0.57	5.8	84	20	212	140	440	21	2070
New Mexico	1965-1966	32	2.3	0.66	7.0	3.4	0.29	12	6.4	8.7	0.63	34
New York	1967	1	2.9			5.7				11		
Oregon	1964-1969	358	1.2	0.14	7.5	12	0.26	1180		70	0.52	4480
Southern Oregon	1966-1969	339	1.2	0.14	7.5	4.3	0.26	26	21	25	0.52	116
Northeastern Oregon	1964-1967	9	1.4	0.20	2.3	290	8.5	1180	1280	1700	58	4480
South Carolina	1966-1968	15	2.0	1.3	3.7	260	5.3	1090	705	800	22	3550
South Dakota	1965-1968	93	2.3	0.19	5.7	31	0.09	448	16	75	0.37	876
Tennessee	1965-1968	5	1.7	1.0	3.0	28	21	33	98	110	50	174
Texas	1965-1967	4	2.1	1.0	4.7	3.5	0.66	11	7	8.3	2.7	15
Utah	1965-1967	65	1.4	0.25	4.6	11	0.36	313	23	50	1.8	888
Vermont	1966	12	1.6	0.93	2.3	3.3	0.12	8.0	10	11	0.5	34
Eastern Washington	1964-1966	162	0.74	0.11	1.4	140	75	350		1200	460	3040
Wisconsin	1966-1967	40	1.9	0.46	3.6	2.4	0.15	18	2	8.7	0.40	75
Wyoming	1966	1	0.34			0.87				14		

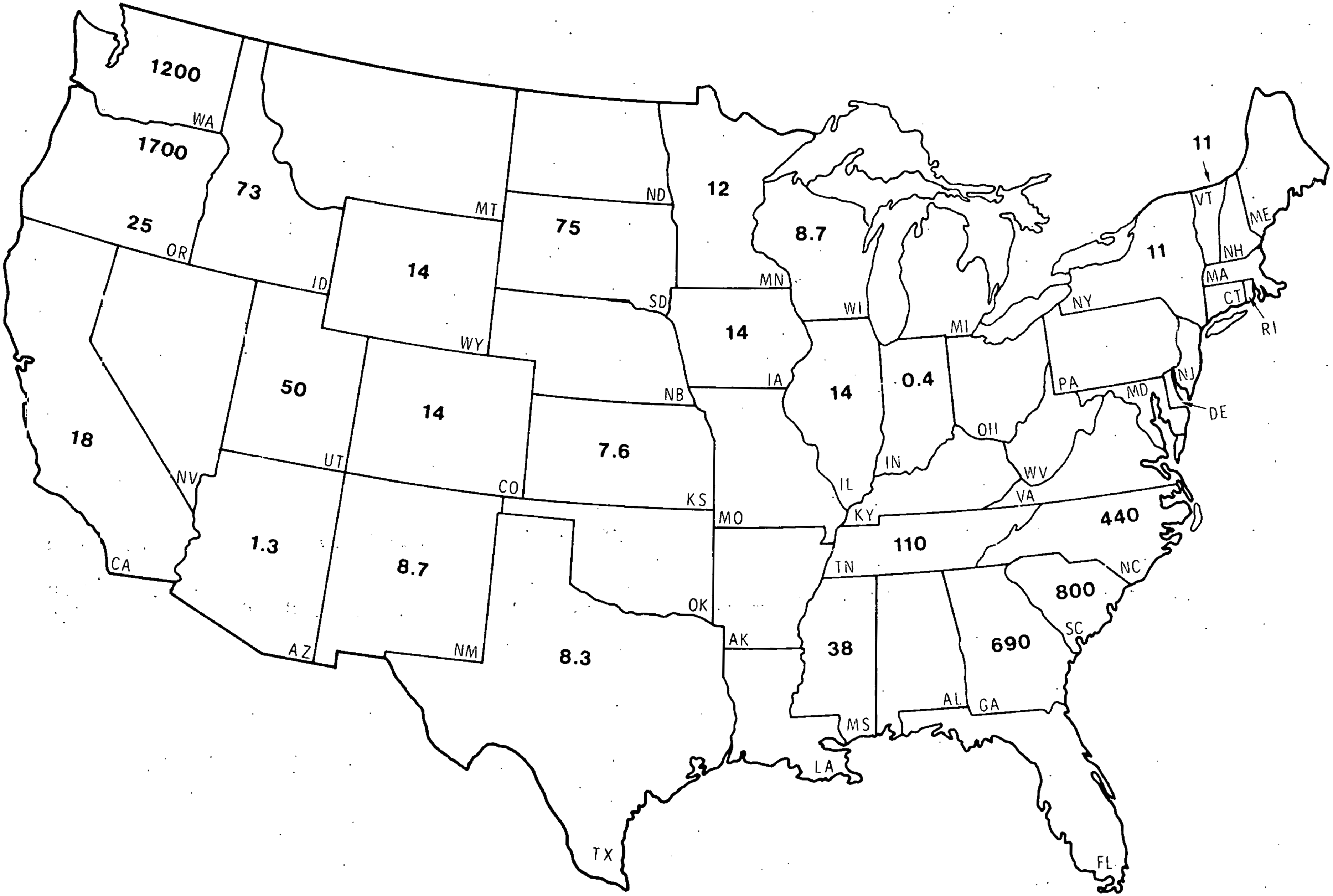

FIG. 1. Average ratio by states of ^{129}I atoms per 10^9 atoms of ^{127}I in bovine thyroids collected between 1964 and 1969.

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FIGURE 1

TABLE IV. SUMMARY BY EASTERN WASHINGTON COUNTIES OF THE RESULTS OF ANALYSES FOR ^{127}I AND ^{129}I IN BOVINE THYROID TISSUE COLLECTED FROM 1964 TO 1966

County	Number of Thyroids	^{127}I Concn (mg/g of Thyroid)			^{129}I Concn (10^{-3} pCi/g of Thyroid			$^{129}I/^{127}I$ Atom Ratio (10^{-9})			
		Avg	Min.	Max.	Avg	Min.	Max.	Median	Avg	Min.	Max.
Adams	9	1.5	0.41	3.0	270	65	637	635	1500	123	4900
Benton	12	1.2	0.081	2.9	320	26	914	1030	1600	623	3800
Chelan	39	1.1	0.13	6.0	110	8.7	922	582	690	16	3900
Columbia	3	0.85	0.28	1.7	245	65	515		1500	1300	1700
Douglas	13	0.96	0.30	3.7	260	1.5	1520	960	2300	2.3	8300
Franklin	19	0.78	0.16	2.2	279	54	1030	2180	2940	158	10900
Grant	34	1.2	0.19	4.8	320	20	1010	1760	2060	128	6206
Kittitas	7	1.1	0.35	2.1	218	20	668	515	2060	52	10700
Lincoln	7	0.96	0.48	3.0	195	11	443	1340	1900	20	4780
Spokane	1	0.31			30				530		
Walla Walla	56	1.4	0.11	14	193	4.8	1030	882	1300	45	7270
Whitman	1	2.3			282				680		
Yakima	33	1.2	0.11	9.5	73	5.3	305	277	480	36	1990
Umatilla, Oregon	9	1.4	0.20	2.3	290	8.5	1180	705	1700	58	4480

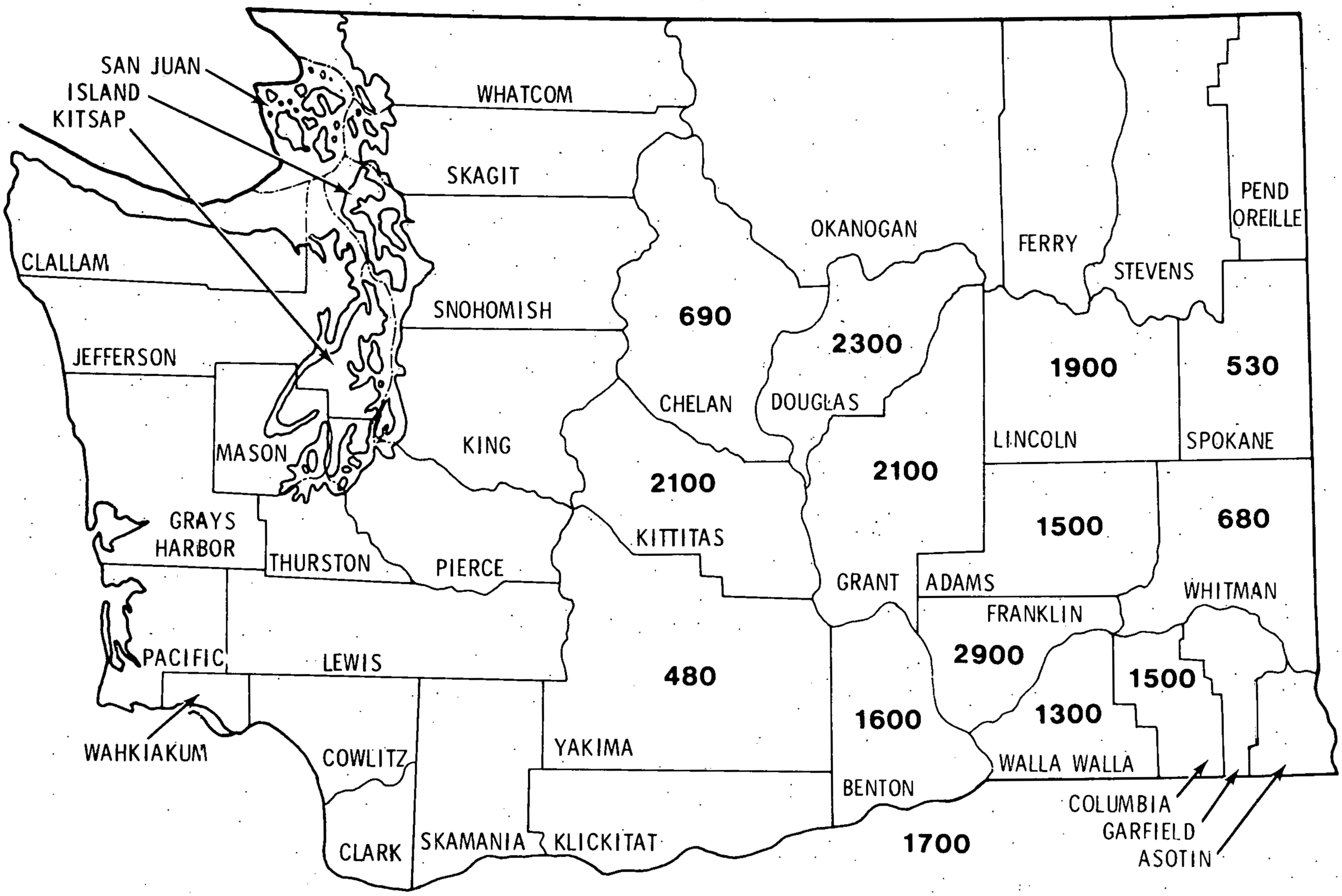

FIG. 2. Average ratio by Eastern Washington counties of ^{129}I atoms per 10^9 atoms of ^{127}I in bovine thyroids collected between 1964 and 1966.

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FIGURE 2

TABLE V. SUMMARY OF THE RESULTS OF ANALYSES FOR ^{127}I AND ^{129}I IN AIR SAMPLES COLLECTED IN NORTHWESTERN UNITED STATES IN 1965-1970

Location	Number of Samples	Filter Collected Fraction			Activated Charcoal Collected Fraction		
		^{127}I Avg Concn (ng/m^3)	^{129}I Avg Concn (pCi/10^{6}m^3)	^{129}I/^{127}I Atom Ratio (10^{-9})	^{127}I Avg Concn (ng/m^3)	^{129}I Avg Concn (pCi/10^{6}m^3)	^{129}I/^{127}I Atom Ratio (10^{-9})
Richland, Wash.	52	1.1	6.5	35000	4.7	43	51000
Spokane, Wash.	47	1.1	1.6	8000	6.6	2.7	2200
Olympic Peninsula, Wash.	24	1.3	0.071	220	2.2	0.096	250

TABLE VI. SUMMARY OF THE RESULTS OF ANALYSES FOR ^{127}I AND ^{129}I IN PRECIPITATION SAMPLES COLLECTED FROM NORTHWESTERN UNITED STATES BETWEEN 1966 AND 1971

Location	Number of Samples	Year	^{127}I Concn (ng/litre)			^{129}I Concn (10^{-6} pCi/litre)			^{129}I/^{127}I Atom Ratio (10^{-9})		
			Avg	Min.	Max.	Avg	Min.	Max.	Avg	Min.	Max.
RAIN											
Richland, Wash.	10	1966	920	270	2700	4800	100	22000	27000	1800	89000
Richland, Wash.	1	1968	340			650			11000		
Richland, Wash.	1	1971	1800			26000			81000		
Spokane, Wash.	23	1966-1967	1200	500	2300	3400	43	46000	11000	410	11000
White Pass, Wash.	1	1969	640			580			5100		
Olympic Peninsula, Wash.	1	1967	1100			67			340		
Copalis, Wash.	1	1969	3600			2800			4400		
Troutdale, Ore.	4	1969	4000	1500	6600	2900	900	7500	3600	1500	6300
Boise, Idaho	1	1967	2500			410			920		
Rocksprings, Wy.	1	1967	1200			370			170		
Ogden, Utah	1	1968	1100			150			760		
SNOW(a)											
Richland, Wash.	1	1970	1100			4300			23000		
Pullman, Wash.	1	1971	510			1700			19000		
Olympic Peninsula, Wash.	1	1970	310			6.1			110		
Boise, Idaho	1	1970	470			6.6			78		
Bend, Ore.	1	1970	830			120			800		

(a) Concentration values for snow refer to amounts per litre of melted snow.

The small number of samples analyzed from most locations must be considered when interpreting these results. Thus a single sample from a location is insufficient to establish a reliable estimate of the value. The total number of results, however, does indicate that levels observed in Eastern Washington were above background levels.

Lake and river samples to the extent that they represent precipitation and not ground water should establish ^{129}I levels in precipitation integrated over a long time period. Data on the ^{129}I content of lake and river samples from the Northwestern United States are listed in Table VII. The ^{129}I levels observed were found to be as low or lower than the lowest found in precipitation samples. No evidence of increased ^{129}I levels in the Columbia River as a result of Hanford operation was detected.

The number of grass samples taken for analysis was smaller than the number of thyroids analyzed since thyroids should represent a better integrated sample both over time and location. The results obtained by iodine analysis of grass samples are listed in Table VIII. Higher ^{129}I levels were observed in Benton County, Washington than at other locations.

Only a limited number of milk samples have been analyzed for ^{129}I. The results are listed in Table IX. High values for the ^{129}I in milk were not restricted to Eastern Washington. This may be related to the shipment of feed from Eastern Washington or may be related to the relatively high levels of ^{129}I in rain observed at other locations, (Table VI). Additional analyses are required to establish the background levels of ^{129}I in milk samples.

The ratio of $^{129}I/^{127}I$ is critical in any evaluation of potential population doses from ^{129}I.[3] A comparison of $^{129}I/^{127}I$ for the various sample types and the dilution of the ^{129}I with natural iodine in the pathway of iodine from air to milk or bovine thyroids from three Washington locations are presented in Table X. The apparent buildup of ^{129}I in the thyroids of undomesticated animals requires further investigations.

RADIATION DOSES FROM ^{129}I

The concentrations of ^{129}I measured in the environs of the Hanford project were used to estimate the potential radiation dose to the thyroids of local residents. Although the measurements did not include every common dietary item and were not always correlated with each other in time, it was still possible to make approximations of the ^{129}I intake rate and dose. Most of the assumptions required in these approximations have previously been published.

Equations and parameters describing the transfer of radionuclides from air and water to human and animal foods and to milk, meat and eggs were developed for a study of the radiation doses to people in the year 2000 from a large nuclear power complex.[23] The factors for conversion of

TABLE VII. SUMMARY OF THE RESULTS OF ^{127}I AND ^{129}I ANALYSES OF LAKE AND RIVER WATER SAMPLES FROM NORTHWESTERN UNITED STATES

Location	Number of Samples	Year	^{127}I Concn (ng/litre) Avg	Min.	Max.	^{129}I Concn (10^{-6} pCi/litre) Avg	Min.	Max.	$^{129}I/^{127}I$ Atom Ratio (10^{-9}) Avg	Min.	Max.
Lake Chelan, Wash.	1	1969	450			23			290		
Lake Pend Oreille, Idaho	1	1969	2100			40			100		
Priest Lake, Idaho	1	1969	560			40			400		
Yakima River, Wash.	4	1965-1971	4400	2200	8400	41	15	61	54	38	83
Columbia River, Wash. Upstream from AEC Reservation	9	1965-1971	1300	680	2300	87	32	190	470	80	1000
Columbia River, Wash. Downstream from AEC Reservation	12	1965-1971	1700	230	9500	47	22	96	280	15	570
Snake River, Wash.	4	1966-1971	6200	2600	8000	38	16	90	37	21	63
Olympic Peninsula Rivers, Wash.	2	1969-1971	740	520	950	10	20	17	60	21	99

TABLE VIII. SUMMARY OF THE RESULTS OF ANALYSES FOR ^{127}I AND ^{129}I IN GRASS SAMPLES COLLECTED AT VARIOUS LOCATIONS BETWEEN 1958 AND 1971

Location	Number of Samples	Year	^{127}I Concn (ng/g)			^{129}I Concn (10^{-6} pCi/g)			^{129}I/^{127}I Atom Ratio (10^{-9})		
			Avg	Min.	Max.	Avg	Min.	Max.	Avg	Min.	Max.
Benton County, Wash.	15	1958-1963	400	46	1100	12000	1600	39000	190000	44000	930000
Umatilla County, Ore.	3	1961-1963	250	190	350	100	44	190	2100	1200	3100
Kittitas County, Wash.	15	1961-1962	240	17	790	110	3.8	560	2400	400	6800
Spokane County, Wash.	9	1961-1966	450	15	3200	50	44	460	8900	790	22000
Olympic Peninsula, Wash.	9	1963-1971	960	20	2200	19	0.7	38	120	54	200
Idaho	7	1963-1968	280	20	690	38	6.7	140	1400	110	3600
Montana	6	1966-1968	1700	14	9300	68	14	190	1700	110	5400
Wyoming	2	1968	300	57	540	14	13	14	720	150	1300
Texas	2	1963-1966	590	290	890	2.3	1.7	2.9	26	18	34
Maryland	2	1966	460	210	700	12	2.3	23	130	73	180
New York	3	1967	70	31	100	2.0	1.6	2.6	200	110	290

TABLE IX. RESULTS OF ANALYSES FOR ^{127}I AND ^{129}I IN MILK SAMPLES COLLECTED IN THE NORTHWESTERN UNITED STATES FROM 1967 TO 1971

Location	Year	^{127}I Concn (mg/litre)	^{129}I Concn (pCi/litre)	^{129}I/^{127}I Atom Ratio (10^{-9})
Richland, Wash.	1971	4.8	0.084	97
Benton City, Wash.	1967	0.21	0.021	670
Pullman, Wash.	1971	1.7	0.031	100
Hoh River, Wash.	1968	0.031	0.00011	19
Hoquiam, Wash.	1969	0.40	0.0056	79
Chehalis, Wash.	1971	1.1	0.034	180
Portland, Ore.	1970	1.2	0.021	93

TABLE X. COMPARISON OF THE $^{129}I/^{127}I$ RATIOS IN VARIOUS MATERIALS FROM THREE WASHINGTON LOCATIONS

	Typical $^{129}I/^{127}I$ Atom Ratios (10^{-9})		
	Benton County	Spokane County	Olympic Peninsula
Air			
Particle Fraction	35000	8000	220
Gaseous Fraction	51000	2200	250
Snow	23000		110
Rain Water	27000	11000	340
River Water	280		60
Grass	190000	8900	120
Milk	97 to 670		19
Thyroids			
Bovine	1600	530	
Deer and Elk	39000		104

radionuclide intake rate to thyroid dose were derived from the equations and methods of the International Commission on Radiological Protection (ICRP),[23-25] utilizing the new value of 100 days for the biological half-life of iodine in the thyroid of adults.[26] The diets utilized in the calculations were those currently employed at Hanford for annual estimates of radiation doses to local residents.[27] The results of these calculations are tabulated in Table XI.

The total dose listed in Table XI is only ∿0.4 mrem/yr to the hypothetical individual with maximized dietary habits and ∿0.2 mrem/yr to the average adult. These doses were calculated for the situation where the radionuclide content of the body has come to equilibrium with the intake rate.

Similar calculations were performed for the 1-year-old infant with maximized diet (2045 m^3/yr of air, 292 litre/yr of water, and 365 litre/yr of milk) and for the infant with average dietary habits (2045 m^3/yr of air, 146 litre/yr of water, and 219 litre/yr of milk). The total doses to the maximum and average infant were only ∿0.3 and 0.1 mrem/yr, respectively. Nearly the entire thyroid dose to the infant comes from ingestion of milk. Less than half of the adult dose comes from milk, with significant contributions from beef and vegetables. None of these doses are significant when compared to the current guide of 1500 mrem/yr to the thyroid of an individual member of the general public or 500 mrem/yr to the average of a suitable sample of the public.

Similar estimates of the radiation dose to the thyroid of an average adult residing on the Olympic Peninsula yielded 3×10^{-4} mrem/yr, ∿0.2% of that estimated for the average Richland resident.

The significance of the $^{129}I/^{127}I$ atom ratios measured can be judged by comparing them with the ratios required to deliver a dose rate of 1500 mrem/yr to the thyroid. The limiting ratio in an adult thyroid is 2.9×10^{-2} atoms ^{129}I per atom ^{127}I. For a 1-year-old child the limiting ratio is 1.1×10^{-1}, much less restrictive because the child's thyroid has a much lower concentration of ^{127}I than does the adult thyroid. By comparison the highest ratio of $^{129}I/^{127}I$ in any single sample was 9.3×10^{-4} for a grass sample collected from Benton County, Washington; this is 3% of the limiting specific activity for the thyroid. The highest ratios measured in items of human diet were 5×10^{-5} in air, 1×10^{-6} in Columbia River water and 6.7×10^{-7} in milk, all much less than 1% of the limiting atom ratio.

The highly variable ratios of $^{129}I/^{127}I$ found in this study parallel the extreme variability recently reported for milk samples collected in the vicinity of the Nuclear Fuels Services fuels reprocessing plant in New York state.[29] The wide variability precludes the extrapolation of the $^{129}I/^{127}I$ ratio from one sample to the next and between sample types to potential thyroid content.

TABLE XI. ESTIMATED RADIATION DOSES FROM ^{129}I IN THE HANFORD, WASHINGTON ENVIRONMENT

Item	Maximum Individual Adult Dose				Average Richland Adult Dose			
	Annual Intake[27]	^{129}I Concn pCi/(m^3,litre,kg)	Total ^{129}I Intake pCi/yr	Dose rate mrem/yr(a)	Annual Intake[27]	^{129}I Conc pCi/(m^3,litre,kg)	Total ^{129}I Intake pCi/yr	Dose rate mrem/yr
Air	7300 m^3	5.0×10^{-5}	0.36	1.5×10^{-3}	7300 m^3	5.0×10^{-5}	0.36	1.5×10^{-3}
Water	730 litre	9.6×10^{-5}	7.0×10^{-2}	3.7×10^{-4}	680 litre	4.7×10^{-5}	3.2×10^{-2}	1.7×10^{-4}
Milk	380 litre	8.4×10^{-2}	32	0.168	130 litre	0.05(b)	6.5	3.4×10^{-2}
Beef	80 kg	~0.24	19	0.100	74 kg	~0.12	~8.8	~4.6×10^{-2}
Chicken	8 kg	~3.0×10^{-5}	~2.4×10^{-4}	1.2×10^{-6}	5.4 kg	~3.0×10^{-5}	~1.6×10^{-4}	~8.5×10^{-7}
Eggs	30 kg	~4.5×10^{-3}	~0.13	7.1×10^{-4}	15 kg	~4.5×10^{-3}	~6.7×10^{-2}	~3.5×10^{-4}
Columbia River Fish	40 kg	~1.4×10^{-3}(c)	~0.058	3.0×10^{-4}	0.48 kg	~7.0×10^{-4}	~3.4×10^{-4}	~1.8×10^{-6}
Game Birds	-	-	-	-	1.2 kg	~3.7×10^{-4}(d)	~4.4×10^{-4}	~2.3×10^{-6}
Leafy Vegetables	73 kg	~0.14	~10	5.5×10^{-2}	36.5 kg	~0.14	~10	~5.5×10^{-2}
Other Vegetables and Fruits	530 kg	~0.014	~7.4	3.9×10^{-2}	200 kg	~0.014	~2.8	~1.5×10^{-2}
Total				0.366				0.152

(a) After equilibrium is achieved between intake rate and thyroid levels
(b) For two samples (0.084 and 0.021 pCi/litre) collected four years apart
(c) Assuming a bioaccumulation factor of 15 pCi/kg per pCi/litre[28]
(d) Assuming 1/2 the game birds are waterfowl which have concentrations similar to those in fish and the other half are upland game birds similar to chickens

CONCLUSIONS

The current levels of ^{129}I in the environment throughout the United States are higher than can be explained by natural processes and are the result of man-produced ^{129}I being released to the environment. The areas adjacent to the AEC reservations at Hanford, Washington and Savannah River, South Carolina were found to contain levels of ^{129}I higher than found in most other areas of the United States. Above background levels of ^{129}I also exist in the environment of the nuclear fuel reprocessing plant at West Valley, New York.[10] The radiation doses to human thyroids projected from this study are 0.4 mrem/yr in the vicinity of the Hanford Reservation and 3×10^{-4} mrem/yr in the Olympic Peninsula in Western Washington. Such doses are substantially below established limits and are insignificant in comparison to the 100 mrem or more the thyroid receives each year from natural radiation.

Surveillance of the ^{129}I levels in the environment, especially near nuclear fuel reprocessing facilities, should be continued. Further studies of possible mechanisms for reconcentration in various pathways of exposure to man are required. Since the ratio of $^{129}I/^{127}I$ is highest in the atmosphere, potential mechanisms for uptake of atmospheric iodine without dilution by natural iodine require evaluation.

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