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THE RADIOECOLOGY OF IODINE-129
AN INTERIM REPORT

by

J. K. Soldat F. P. Brauer
J. E. Cline J. E. Fager
B. Klepper W. H. Rickard
B. E. Vaughan D. G. Watson

Battelle
Pacific Northwest Laboratories
Richland, Washington 99352

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PREFACE

This is an interim report intended to inform the AEC staff about a substantial part of our research results on the radioecology of ^{129}I. Because of the interim nature of the report, data are necessarily somewhat incomplete. Data from ongoing collections and analyses should help to formulate a more complete picture. We anticipate that a second interim report will be developed at some future time with a more complete data bank.

The subsection on geographical distribution of iodine has been taken substantially from material prepared for a forthcoming monograph on radioiodine sponsored by the U.S. Atomic Energy Commission and the American Industrial Hygiene Association. The reference is: Bustad, L. K., J. K. Soldat, and P. G. Voilleque. Radioiodine: Biomedical and Industrial Hygiene Aspects, (in press).

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GEOGRAPHICAL, ECOLOGICAL AND BIOLOGICAL CONSIDERATIONS IN ANALYSIS OF RADIATION DOSE FROM ^{129}I

INTRODUCTION

Radioactive isotopes of iodine are produced during nuclear fission in reactors and in weapons tests and to a limited extent in nature. Over the past several years, considerable effort has been devoted to the study of the generation, release, dispersion, chemical forms and radiation dosimetry of ^{131}I. Recently attention has been drawn to another isotope of radioiodine, ^{129}I. Because of its extremely long half-life (1.6×10^7 yr), ^{129}I has the potential for accumulation in the environment from long-term, chronic releases from nuclear facilities.

This section discusses possible problems associated with such an accumulation, including transfer of the ^{129}I through the biosphere and resultant radiation doses to man and other organisms.

GEOGRAPHICAL DISTRIBUTION OF STABLE IODINE AND NATURALLY PRODUCED ^{129}I

Iodine has only one stable isotope, ^{127}I. It is widely dispersed in the sea, rocks and all organisms and is usually found in the iodide or iodate form. The marine environment and its inhabitants usually contain considerably more iodine than do terrestrial fauna and soil, as shown in Table 1. Marine organisms concentrate iodine to a surprising degree; certain tropical sponges may contain up to 14% iodine.[21] Some land masses, such as Chilean nitrate beds, are a rich source of iodine; other areas are so low in stable iodine that their inhabitants suffer from iodine deficiency. Large areas in the northern and western United States have been labeled as "goitrogenic" because of their low iodine content.[2,18]

This wide variation in soil iodine content has been the subject of much speculation and argument.[23] However, Goldschmidt's view[10] that the major source of iodine in soils is material released from the ocean surfaces appears now to be well substantiated.[23] The possibility that

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TABLE 1. IODINE IN MARINE AND TERRESTRIAL ENVIRONS[a]

Component	Percent by Weight	
	Terrestrial	Marine
Thyroid gland (animals and fish)	10^{-1}	10^{-1}
Animals	10^{-6}	10^{-4}
Plants (algae)	10^{-5}	10^{-3}
Soils (silts)	10^{-4}	10^{-2}
Rock	10^{-5}	----
Water	10^{-7}	10^{-6}
Salt[b]	10^{-7}	10^{-5}
Air	10^{-8}	10^{-6}
Rain and snow	10^{-7}	----

a. References 9, 10, 11, 13, 14, 26.
b. Iodized salt contains 10^{-3} to 10^{-2}% iodine.

the Chilean deposits resulted from airborne oceanic iodide[10] has been evaluated further and a general theory has been developed that these nitrate deposits resulted from precipitation scavenging of the atmosphere together with the absence of weathering and biological activity.[3]

Iodine concentration in the atmosphere generally diminishes with increasing distance from the oceans; this reduction consequently reduces the amount of iodine transferred to inland soils by precipitation scavenging and dry deposition. The period of exposure of the soil also affects the iodine deposit; hence the reduced concentrations in young postglacial soils generally and particularly in the deeper horizons of such soils.[10]

Measurements of iodine concentrations in the atmosphere and in precipitation in central Europe showed concentrations in air ranging from 0.1 to 1 $\mu g/m^3$. Higher concentrations, up to 10 $\mu g/m^3$, were found in air along coastal regions. Concentrations found in rain and snow ranged from 0.2 to 5 μg/liter. Iodine concentrations in European rivers were in the range of 1 μg/liter, while in some mineral springs there may be as much as 300 μg/liter.[10]

There are apparently at least two processes which release iodine from the ocean surface to support the levels in marine atmospheres. The first is the release of gaseous radioiodine (I_2) by photochemical oxidation of iodide. Experimental demonstrations of the I_2 production by this mechanism have been described. Release of iodine-bearing particulate material from an experimental sea water surface has been reported.[19,22] It has also been suggested that organically bound iodine may be released to the marine atmosphere.[1,4] Understanding of relationships between the various sources of airborne iodine and elucidation of the mechanisms which produce the observed concentrations and relative iodine enrichment (see 5, 6, 7) await further detailed measurements, particularly of iodine species. Some of the common inorganic chemical species in which iodine can be found are listed in Table 2.

The human body contains 20 to 50 mg of iodine; up to one-half or more may be localized in the thyroid gland. The International Commission on

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TABLE 2. COMMON INORGANIC CHEMICAL SPECIES OF IODINE[12]

Valence	Common Species
-1	I^-, HI, NaI, $HI \cdot \underline{n}H_2O$
0	I_2
+1	ICl, IBr, HOI
+5	I_2O_5, IO_3^-, HIO_3, $NaIO_3$
+7	IO_4^-, HIO_4, $NaIO_4$

Radiological Protection (ICRP)[15] has summarized data on the concentration (μg/g of wet tissue) of iodine in various body organs of a "standard" adult as follows: total body, 0.43; thyroid, 350; ovary, liver, skin, muscle, heart, ∿1; pancreas, 0.5; gastrointestinal tract, 0.4; bone, 0.3; kidney, 0.2; brain, 0.1. The concentration of iodide in the blood of man ranges from 3 to 13 μg/100 ml with a mean of 7.7; the protein-bound iodine concentration ranges from 4.0 to 8.5 μg/100 ml.[24]

Although radioiodine released into the biosphere in North America and Europe reaches man chiefly through cow's milk, the stable iodine content of cow's milk is only about 0.2 ppm with a range of 0.004 to 2 ppm.[24] Bruner[2] quotes McClendon's[20] value of about 0.05 ppm for iodine in milk and milk products. However, the iodine content of milk may vary with stage of lactation. It also depends upon dietary intake of stable iodine and possibly of other materials.

Twenty-three radioactive isotopes of iodine have been identified. The single naturally-occurring radioisotope, ^{129}I, has a half-life of 1.6×10^7 years.[30] It is produced in nature by spontaneous fission of uranium and thorium in soils and in the oceans, by the interaction of high energy particles with xenon in the upper atmosphere, and by neutron-initiated reactions with $^{128}Te(n,\gamma)$ and $^{130}Te(n,2n)$. Edwards[8] estimated that these natural reactions would produce a steady-state ratio of ^{129}I to ^{127}I of $\geq 10^{-14}$, and Kohman and Edwards[16] estimated that natural reactions should produce a current equilibrium $^{129}I:^{127}I$ value of about 2×10^{-12} in the oceans. Analyses of samples of a natural silver iodide deposit in Australia have led to an estimate that, prior to man's utilization of nuclear fission, the equilibrium terrestrial ratio was bounded as follows:

$$2.2 \times 10^{-15} \leq {}^{129}I:{}^{127}I \leq 3.3 \times 10^{-15}.$$ [25]

MAN-MADE SOURCES OF ^{129}I

Iodine-129 is produced in nuclear fission as a decay product of ^{129}Te. The fission yields of several radioiodine isotopes from thermal neutron fission of ^{235}U were tabulated by Holland.[12] The ^{129}mass chain is

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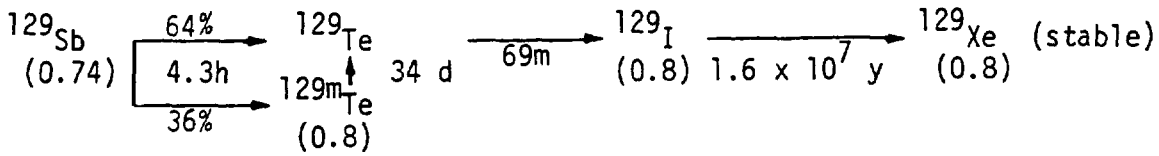
reproduced in part below with more recent values of radioactive half-lives from References 17 and 30.



The values in parentheses are cumulative fission yields in atoms per 100 fissions. More recent data[30] indicate that the cumulative yield for ^{129}Te and ^{129}I are both 1.0 rather than 0.8. Since there is essentially no direct fission yield of ^{129}I, the total quantity present in a fission product mixture will increase slowly with time after irradiation has ceased as the ^{129}I precursors decay. The peak activity is not reached for several months.

Iodine-129 is produced in nuclear explosions at approximate rates of 30 and 50 μCi/kt from fission of ^{235}U and ^{239}Pu, respectively. The transport and diffusion of radioiodine depend upon the initial height of the cloud and upon meteorological processes. A review of these factors was made by the United Nations Scientific Committee on the Effects of Atomic Radiation.[28] Megaton-size weapons exploded above ground inject their debris into the stratosphere, where an overall mean residence time of about 2 years has been estimated. The fission products reach the level of rain-bearing clouds by turbulent mixing, and, below this level, particles are rapidly deposited by precipitation. Dry deposition, affected by micro-meteorological processes and vegetation, are significant only in areas of low rainfall.

The net production of ^{129}I from fission of ^{235}U in a thermal reactor is about 1.0 μCi per megawatt-day (MWD) depending upon the neutron flux and irradiation time which effect the "burn-up" of ^{129}I by absorption of neutrons. The net production is actually higher than that quoted above due to the contribution from fission of the ^{239}Pu built up in the uranium fuel by neutron absorption in ^{238}U. The yield of ^{129}I is 1.4% in ^{239}Pu fission compared to 1.0% in ^{235}U fission. If, for example, 40% of the reactor energy comes from ^{239}Pu fission[27] the ^{129}I production will increase by about 15%.

Since nearly all of the fission products generated in nuclear fuels are retained within the fuel cladding until the time the fuel is chemically reprocessed, the principal sources of ^{129}I in the environment are the effluents from nuclear fuels reprocessing plants rather than those from nuclear reactors. Russell and Hahn[27] have estimated that by the year 2000 about 10^4 Ci of ^{129}I will have been generated by world production of electricity from nuclear power with perhaps 1% or 100 Ci actually released to the environment. By the year 2060, they estimated these values will have risen to 2×10^6 Ci accumulated and 2×10^4 Ci released.

Stable ^{127}I is also produced in nuclear fission, but with a smaller yield (0.25% in ^{235}U and 0.37% in ^{239}Pu fission).[30] It would appear, therefore, that the atom ratio $^{129}I:^{127}I$ should be about 4 when irradiation is followed by cooling times of several months to allow the ^{129}I to grow in. Tadmor[29] however quotes this ratio as being 0.75 for cooling times of 1 year without specifying the irradiation conditions. Even accounting for the fact that ^{129}I will burn-up at a faster rate in the reactor (σ = 27 barns) than will ^{127}I (σ = 6.2 barns) the ratio $^{129}I:^{127}I$ should be no less than about 3 even for irradiation times of about 1 year. It is possible that additional ^{127}I could be present as a result of iodine contaminants in chemicals employed in manufacture and reprocessing of the fuel.

The exact quantity of stable iodine and the ratio $^{129}I:^{127}I$ in the fuel when processed is important because it is related to the specific activity of ^{129}I which can exist in nature. If this ratio were 1 for example then no more than half of the iodine atoms accumulated in the thyroid of man or animals could be ^{129}I. Because of dilution with the naturally occurring stable iodine in the environment, the ratio would be considerably less than 1 even in the immediate vicinity of a nuclear facility discharging ^{129}I to the environs. For example, the highest ratios reported in samples collected in the vicinity of the Nuclear Fuels Services plant, which reprocesses irradiated reactor fuels, were in the order of 10^{-3}.[27,31]

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POTENTIAL PATHWAYS OF EXPOSURE FROM ^{129}I

Radioiodine released to the environment can expose man and other organisms via several different exposure pathways. Potential pathways are listed in Table 3 without regard to their relative importance. These are also illustrated in Figures 1 for man and 2 for other organisms. Although the figures are oriented toward releases from nuclear facilities, they could apply to fallout as well if the gaseous effluent plume were replaced by airborne fallout debris and if the liquid effluent source were replaced by rain-out and run-off from land fallout.

For exposure to man, the most significant pathways are ingestion of milk and vegetables whether contaminated via air or water. If there is no aerial deposition or irrigation of crops, then ingestion of drinking water and aquatic foods could be the significant pathways. As will be shown in the following section, certain aquatic organisms can concentrate iodine from their water environment leading to relatively high intakes of iodine by consumers of such organisms.

External pathways of exposure, whether to air, water or ground, are relatively insignificant for ^{129}I because of rather low energies of the beta and gamma radiations emitted.

BIOLOGICAL AND ECOLOGICAL PARAMETERS

The behavior of iodine and its transfer between certain portions of the environment depend heavily upon its chemical and physical form. Inorganic vapor is the most reactive form of iodine chemically but particulate forms and even organic compounds such as methyl iodide (CH_3I) are readily metabolized in the body.

Ecological Parameters

Most of the studies of iodine behavior have been conducted with available fallout from nuclear explosions, principally ^{131}I in particulate form, or with planned releases of elemental vapor forms of ^{131}I. Very little is known about changes in behavior which might result from changes in form and availability of ^{129}I in the long term. Analyses of the potential

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TABLE 3. PATHWAYS OF EXPOSURE TO MAN AND OTHER ORGANISMS FROM ^{129}I

Water Pathways

External

Water immersion and water surface
Exposure to shoreline

Internal

Ingestion of water
Ingestion of aquatic foods
Ingestion of irrigated food crops
Ingestion of products from animals fed irrigated foods

Air Pathways

External

Air submersion
Exposure to deposited materials

Internal

Inhalation
Ingestion of food crops
Ingestion of animal products

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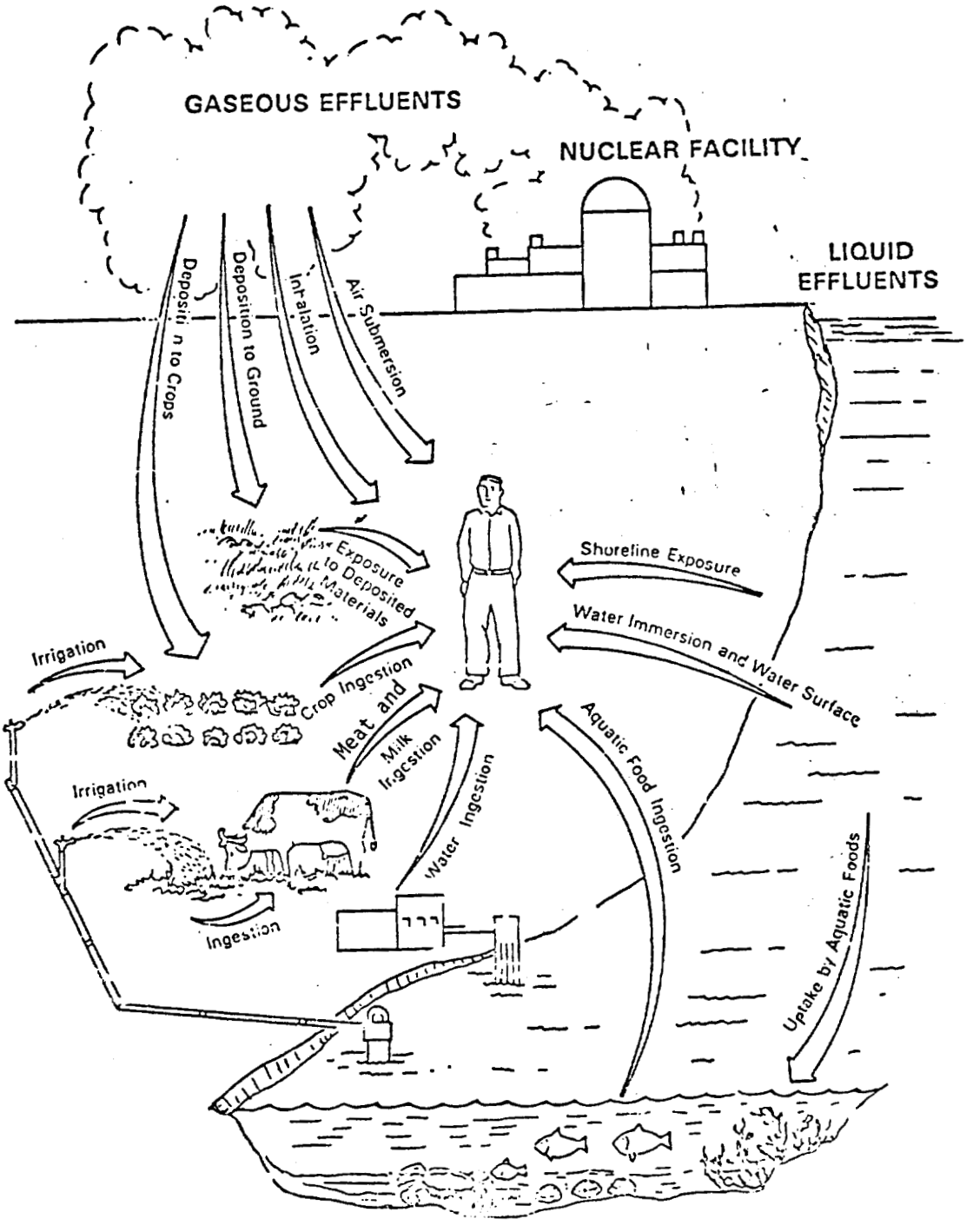

FIGURE 1. Exposure Pathways to Man from Iodine-129

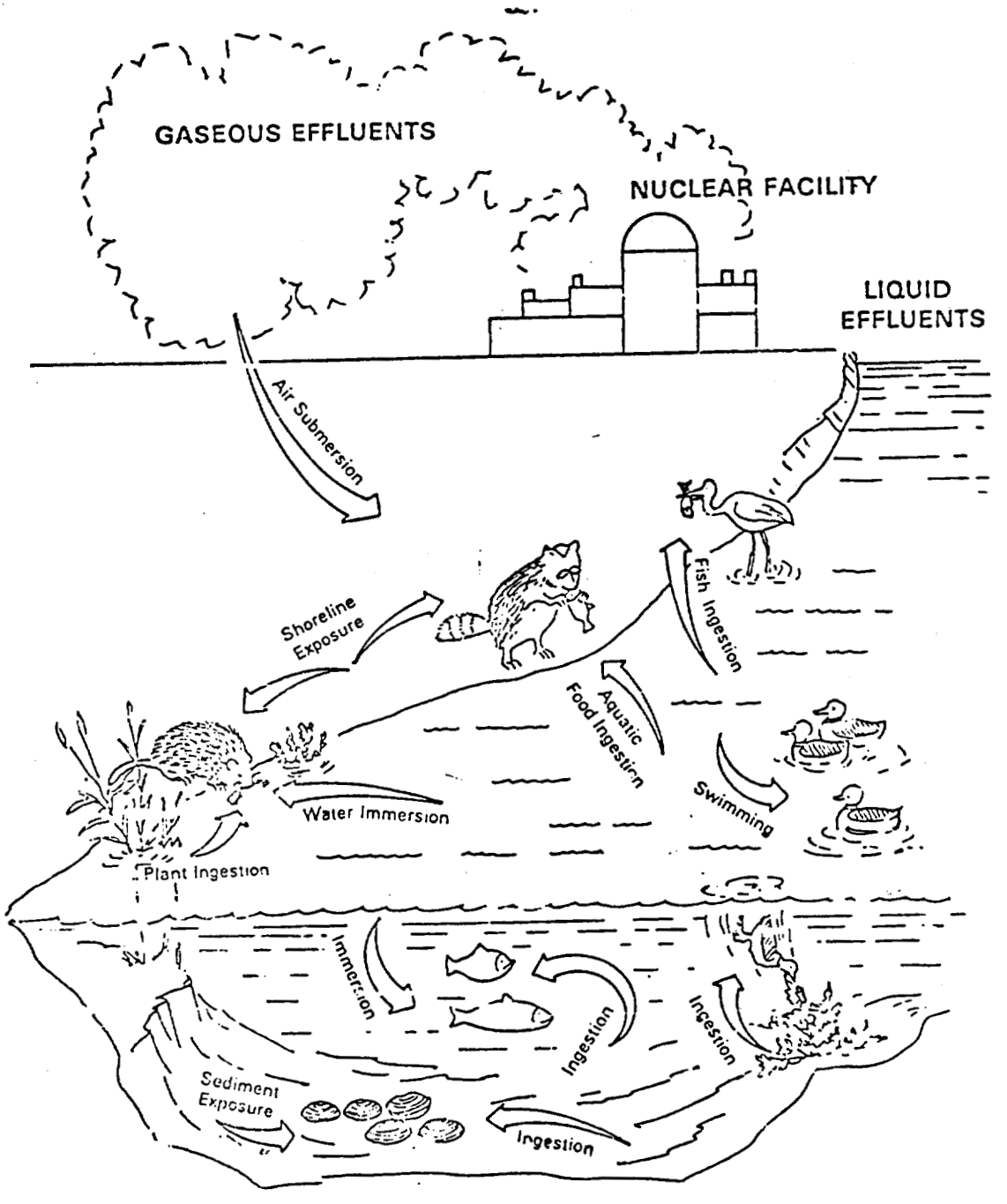

FIGURE 2. Exposure Pathways to Organisms Other than Man from Iodine-129

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accumulation and radiation doses resulting from releases of ^{129}I to the environment must, therefore, be based upon studies of ^{131}I for the short-term behavior and stable iodine for the long-term behavior.

Behavior of iodine in the aquatic environment, while far from completely understood, can be described in terms of bioaccumulation factors--ratios of concentration in an organism to concentration in the water at equilibrium. Such factors have been calculated[32] for all elements by Thompson, et al., from literature data on concentrations in water and in different organisms. Frecke[33] has tabulated bioaccumulation factors for numerous radionuclides in the marine environment. These bioaccumulation factors for iodine are listed in Table 4.

In addition to their use in calculating the concentrations in and radiation doses to the organisms listed in Table 4, the bioaccumulation factors are necessary input for calculation of body burdens and doses to organisms including man which consume the listed organisms, as explained in the next section.

Factors for transfer of radionuclides from soil to plant and from animal feeds to animal products were derived by Soldat[34] for a study of the potential doses to people from a nuclear power complex in the year 2000. The ecological factors selected for iodine are summarized in Table 5.

The concentration in a plant resulting from direct deposition onto the plant surfaces is limited by the 14-day "ecological" half-life. The long-term accumulation of ^{129}I in the plant is then given by the expression

$$C_p = \frac{R\ D}{Y} \frac{\left(1-e^{-\lambda_E t'}\right)}{\lambda_E}$$

where: C_p = concentration on plant in pCi/kg

D = deposition rate in pCi/m^2/day

R = fraction of deposition retained on plant = 0.25

Y = plant yield in kg/m^2

λ_E = effective removal constant

t' = time of exposure of crop to airborne contamination.

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TABLE 4. AQUATIC BIOACCUMULATION FACTORS FOR IODINE
(Concentration in organism ÷ concentration in water)

Organism	Fresh Water Reference 32	Salt Water Reference 32	Salt Water Reference 33
Fish	15	10	20
Invertebrates	5	50	100
Algae	40	4000(1000*)	10,000

* Measured value.

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TABLE 5. ESTIMATED ECOLOGICAL PARAMETERS FOR IODINE[a]

Deposition velocity from air to ground ($pCi/m^2/sec$ per pCi/m^3 = m/sec)		10^{-2}[b]
Fraction of aerial deposition retained on plant		0.25
"Ecological" half-life on plant (days)		14
Fraction of deposited iodine translocated to part of plant eaten	- leafy vegetables	1.0
	- other vegetables & grain	0.1
Soil uptake [pCi/kg plant (wet) per pCi/kg soil(dry)]		0.02
Transfer from feed to milk (pCi/liter milk per pCi/day intake)	- cow	0.01
	- goat	0.2[c]
Transfer from feed to meat (pCi/kg meat per pCi/day intake)	- beef	0.02
	- pork	0.09
	- chicken	0.004
Transfer from feed to chicken eggs (pCi/egg per pCi/day intake)		0.03[d]

a. Unless otherwise indicated values were taken from Reference 34.
b. Approximate mid-range of literature values.
c. From Reference 35.
d. Recalculated from data originally used to get the value of 0.08 given in Reference 34.

The effective removal constant is given by:

$$\lambda_E = \lambda_R + \lambda_e = \frac{0.6932}{T_R} + \frac{0.6932}{14}$$

where: T_R = radiological half-life of ^{129}I in days

14 = the "ecological" half-life of the deposited material on the plant.

Calculations based on the parameters given in Table 5 indicate that the uptake of ^{129}I via the soil-root pathway is insignificant for short-term deposition. This pathway would add about 1% per year of accumulation to the concentration in leafy vegetables, if the iodine were deposited from the air and if all of the iodine in soil were retained in the root zone of the plant. It would take about 100 years of soil accumulation before the contribution of the soil-root pathway equaled the contribution from one year's direct deposition on the plant. For other vegetables where only 10% of the aerial deposition may reach the portion of the plant consumed, it would take only 10 years' accumulation in the soil at the root zone to yield a concentration equal to that present from direct deposition. For milk contamination via fresh forage, grain, and stored feed, the soil pathway contributes nearly 2% per year of accumulation to the concentration present via aerial deposition.

Parameters Affecting Human Uptake and Retention

The International Commission on Radiological Protection (ICRP) has published reference data on a standard adult[15] including uptake and retention parameters for radioiodines in various body organs. Data for other ages were reviewed by Soldat for use in the year 2000 study.[34,36] Recently the ICRP has published a newer value of 100 days for the biological half-life of iodine in the adult thyroid.[37] Further review of the literature indicated that the parameters previously selected for other ages in the year 2000 study could also be updated.[38,39,40,41] The revised parameters are summarized in Table 6. These data were utilized to calculate the factors for converting rate of intake of ^{129}I to organ burden and to radiation dose given in the following section.

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TABLE 6. PARAMETERS USED IN CALCULATING THYROID DOSE FACTORS FOR ^{129}I

Parameter	1 yr	4 yr	14 yr	Adult
Fractional uptake via ingestion	0.3[41][a]	0.3[41]	0.3[41]	0.3[37]
Fractional uptake via inhalation	0.23	0.23	0.23	0.23[37]
Biological half-life in thyroid (days)	20[38-41]	20[38-41]	50[38-41]	100[37]
Thyroid mass (grams)	2[34]	5[34]	15[34]	20[15]
Concentration of ^{127}I in thyroid (ppm)	90[42]	180[42]	280[42]	350[15]
Weight of ^{127}I in thyroid (mg)	0.18	0.90	4.2	7.0
Thyroid radius (cm)	1.4[34]	2[34]	2.7[34]	3[15]
Inhalation rate (m^3/day)	5.6[40]	7.0[40]	13.5[40]	20[15]
Effective MeV per disintegration of ^{129}I[b]	0.047	0.047	0.048	0.048

a. Numbers in ( ) are references.
b. Calculated from formulas given on Page 28 of Reference 15 and decay scheme given on Pages 70, 273, 274 of Reference 17.

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RADIATION DOSE TO MAN AND BIOTA

Calculation of radiation doses to man from internally deposited radionuclides requires a knowledge of the distribution of the nuclide within the body, the metabolic behavior of the nuclide, its decay scheme, and the general theory of beta and gamma radiation dosimetry. The biological parameters related to radioiodine in the human body organs were tabulated in the previous section.

Several empirical methods,[43] developed for calculating doses delivered from internally deposited materials, yield reasonably accurate results. The Medical Internal Radiation Dosimetry (MIRD) Committee of the Society of Nuclear Medicine has published several pamphlets[44] as supplements to their Journal, which describe the results of Monte Carlo calculations for estimating the dose in a target organ from a radionuclide contained in a source organ. The pamphlets also tabulate the contributions to absorbed dose from each component in the decay schemes of several nuclides of interest in nuclear medicine. Included in the tabulations were seven radioiodine nuclides, but not ^{129}I. Because of its very long half-life and low energy radiation, ^{129}I is not suitable for either diagnostic or therapeutic applications.

A very simple model has been constructed by the International Commission on Radiological Protection (ICRP) for purposes of estimating radiation doses to man.[15] This model is diagrammed in Figure 3 and discussed below.

When material is ingested, a fraction, f_1, is absorbed from the GI tract and a fraction f_2' of that in the blood is deposited in the organ of interest. Combining f_1 and f_2' into one parameter, f_w we have

$$f_w = f_1 f_2' \qquad (1)$$

f_w is the fraction of the amount ingested which finally reaches the organ of interest. Similarly, f_a is defined as that fraction of the amount of soluble material originally inhaled which reaches the organ.

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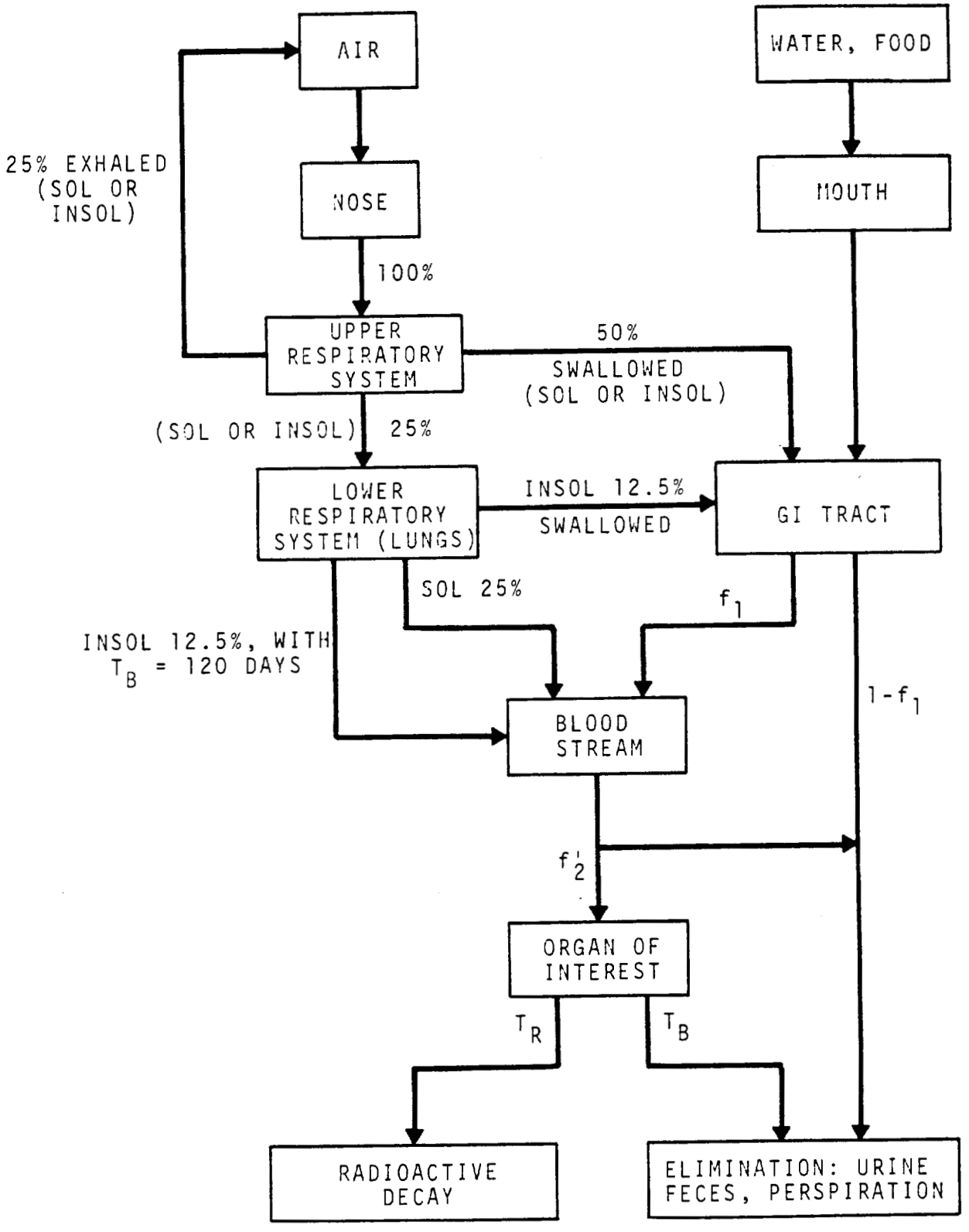

FIGURE 3. ICRP Model for Internal Radiation Dosimetry

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$$f_a = f_2' \, (0.25 + 0.5 \, f_1) \tag{2}$$

Essentially all of the compounds of radioiodine encountered in the environment are completely soluble in body fluids so that $f_1 = 1.0$ and $f_a = 0.75 \, f_w$.

If insoluble material were to be ingested none of it would be absorbed and only the GI tract would be irradiated. If insoluble material were to be inhaled only 12.5% would finally be cleared from the lung to the blood with a biological half-life, T_B, of 120 days.

A biological removal constant, λ_B, analogous to the radiological decay constant, λ_R, is defined as $(\ln 2)/T_B$, and an effective removal constant, λ, can be calculated as

$$\lambda = \lambda_B + \lambda_R = \frac{\ln 2}{\tau} \tag{3}$$

where τ is the effective half-life in the organ of interest.

Since the radiological half-life of ^{129}I is so long, the value of τ will equal T_B and λ will equal λ_B for this nuclide.

The model assumes that the radionuclide is all concentrated in the center of a spherical organ whose radius was estimated on the basis of its shape and size by the ICRP. Under this assumption, all of the particulate radiation (alpha, beta, conversion electrons) is absorbed within the organ. A fraction of the gamma photons is absorbed depending upon the effective radius, x, and the absorption coefficient, μ, specific to the energy and medium (muscle, bone, etc.). The fraction of the gamma energy absorbed is then $1-e^{-\mu x}$.

Using these assumptions together with a detailed decay scheme[17] for the nuclide of interest, the total absorbed energy per disintegration, ε, can be calculated. The assumption of a point source in the center of the organ does not yield results significantly different from those obtained assuming a uniform distribution of the nuclide. However, the contribution to the dose of photons from adjacent organs is not considered as is the case in the MIRD system.

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Dose from a Single Intake of a Radionuclide

If a single intake, Q_o, of a radionuclide occurs via ingestion, the activity initially reaching the organ of interest is $Q_o f_w$. The activity, Q, present at any time, t, after ingestion is

$$Q = f_w Q_o \, e^{-\lambda t} \tag{4}$$

The dose rate DR at time t is directly proportional to the activity concentration present and the effective energy absorbed per disintegration, ε.

$$DR = K \, \varepsilon \, f_w Q_o \left(e^{-\lambda t}\right) / m \tag{5}$$

where DR = dose rate in mrem per day

$\varepsilon = \left[\text{MeV} \left(\frac{\text{rem}}{\text{rad}}\right)\right]$ absorbed per disintegration

Q_o = pCi initially ingested

λ = effective decay constant in days^{-1}

t = exposure time in days

m = mass of organ in grams

K = a conversion constant

$$= \frac{2.22 \text{ dis/min}}{\text{pCi}} \times \frac{1440 \text{ min}}{\text{day}} \times \frac{1.602 \times 10^{-8} \text{ g}\cdot\text{rad}}{\text{MeV}} \times \frac{10^3 \text{ mrem}}{\text{rem}} = 0.0512$$

The total dose delivered to the organ over a time, t, following ingestion is simply the integral of Equation (5) over the time from 0 to t.

$$D = 0.0512 \int_0^t \frac{\varepsilon \, f_w Q_o \left(e^{-\lambda t}\right) dt}{m}$$

$$= 0.0512 \, \varepsilon \, f_w Q_o \left(1 - e^{-\lambda t}\right) / m \, \lambda \tag{6}$$

where D = total dose in mrem.

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If λ is replaced by $0.6932/\tau$ then Equation (6) becomes

$$D = \frac{0.074\ \varepsilon\ f_w\ \tau\ Q_o\ \left(1-e^{-\lambda t}\right)}{m} \tag{6a}$$

and the dose per unit intake becomes

$$D/Q_o = \frac{0.074\ \varepsilon\ f_w\ \tau\ \left(1-e^{-\lambda t}\right)}{m}\ \text{mrem/pCi} \tag{7}$$

As t becomes very long relative to τ the expression $\left(1-e^{-\lambda t}\right)$ approaches unity and the total dose per unit intake becomes

$$D/Q_o = \frac{0.074\ \varepsilon\ f_w\ \tau}{m} \qquad (t \rightarrow \infty) \tag{8}$$

If the mode of intake were inhalation instead of ingestion, the dose equations would be similar to those given above except that f_w would be replaced with f_a throughout.

The thyroid doses received per unit intake of ^{129}I via ingestion and inhalation for four age categories have been calculated from Equation (8) and are listed in Table 7. The results in this table indicate that the dose decreases with increasing age until adulthood when the large increase in τ (50 to 100 days) overshadows the relatively small increase in m (15 to 20 g) between the teen and the adult.

In addition, the effective half-life of ^{129}I in the adult thyroid is long enough so that only 92% of the total thyroid dose from an acute intake will be received in the first year.

Dose Rate from a Chronic Intake of a Radionuclide

If a radionuclide is ingested at a constant chronic rate of P pCi/day, then the rate of change of activity, Q, in an organ will be

$$\frac{dQ}{dt} = -\lambda Q + P\ f_w \tag{9}$$

Integration of Equation (9) yields

$$Q = \frac{P\ f_w\ \left(1-e^{-\lambda t}\right)}{\lambda}\ \text{pCi} \tag{10}$$

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TABLE 7. THYROID DOSE FACTORS FOR ^{129}I

	1 yr	4 yr	14 yr	Adult
Dose per Unit Intake				
mrem/pCi ingested	1.05(-2)[a]	4.22(-3)	3.52(-3)[b]	5.28(-3)[c]
mrem/pCi inhaled	8.07(-3)	3.23(-3)	2.70(-3)[b]	4.05(-3)[c]
Equilibrium Dose Rate				
mrem/yr per pCi maintained in the thyroid	4.43(-1)	1.78(-1)	5.94(-2)	4.45(-2)
weight of ^{127}I in the thyroid, μg	1.8(2)	9.0(2)	4.2(3)	7.0(3)
Maximum Possible ^{129}I in Thyroid				
μg	1.8(2)	9.1(2)	4.3(3)	7.1(3)
pCi[d]	3.17(4)	1.58(5)	7.45(5)	1.23(6)
corresponding dose, mrem/yr	1.41(4)	2.80(4)	4.42(4)	5.47(4)
Thyroid Content of ^{129}I to Yield				
1500 mrem/yr pCi	3.39(3)	8.43(3)	2.53(4)	3.37(4)
^{129}I:^{127}I	1.14(-1)	5.68(-2)	3.61(-2)	2.91(-2)
pCi ^{129}I/μg I	1.88(1)	9.37(0)	6.02(0)	4.81(0)
pCi ^{129}I/g thyroid	1.69(+3)	1.69(+3)	1.69(+3)	1.69(+3)

a. 1.05(-2) = 1.05×10^{-2}
b. 99.4% of this dose is delivered in the first year following the intake.
c. 92.0% of this dose is delivered in the first year following the intake.
d. At a specific activity of 1.73×10^5 pCi ^{129}I per mg ^{129}I.

As before the dose rate is

$$DR = \frac{Q\ \varepsilon\ K}{m}\ \text{mrem/day} \tag{11}$$

$$= \frac{0.0512\ P\ f_w\ \varepsilon}{m\ \lambda}\left(1-e^{-\lambda t}\right) \tag{11a}$$

$$= \frac{0.074\ P\ f_w\ \varepsilon\ \tau\left(1-e^{-\lambda t}\right)}{m}$$

and the dose rate per unit intake rate is

$$DR/P = \frac{0.074\ f_w\ \varepsilon\ \tau\ \left(1-e^{-\lambda t}\right)}{m}\ \text{mrem/day per pCi/day intake.} \tag{12}$$

As $t \to \infty$, Equation 12 reduces to

$$\frac{DR}{P} = \frac{0.074\ f_w\ \varepsilon\ \tau}{m} \tag{13}$$

As before, replacement of f_w with f_a yields the corresponding equations for inhalation.

The similarity between Equations (7) and (12) [and between (8) and (13)] is obvious. Equation (7) can be used to calculate the total <u>dose</u> out to time t following a single intake in units of mrem/pCi. Equation (12) can be used to calculate the <u>dose rate</u> at time t following the start of a chronic intake rate of P pCi/unit time. The result of application of Equation (7) will have units of mrem/pCi intake while the results from Equation (12) will have units of mrem/day per pCi/day (or mrem/yr per pCi/yr) intake.

The total dose over a period of time during which a constant chronic intake is maintained can be obtained by integrating Equation (11a).

$$D = \frac{0.0512\ P\ f_w\ \varepsilon}{m\ \lambda} \int_0^t \left(1-e^{-\lambda t}\right)dt$$

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$$= \frac{0.0512\ P\ f_W\ \varepsilon}{m\ \lambda} \left[t - \frac{e^{-\lambda t}}{-\lambda} \right]_0^t$$

$$= \frac{0.0512\ P\ f_W\ \varepsilon}{m\ \lambda} \left[t - \left(\frac{1-e^{-\lambda t}}{\lambda} \right) \right]$$

$$= \frac{0.0512\ P\ f_W\ \varepsilon}{m\ \lambda^2} \left(\lambda t + e^{-\lambda t} - 1 \right) \text{ mrem} \tag{14}$$

or normalizing to unit intake rate

$$\frac{D}{P} = \frac{0.0512\ f_W\ \varepsilon}{m\ \lambda^2} \left(\lambda t + e^{-\lambda t} - 1 \right) \frac{\text{mrem}}{\text{pCi/day}} \tag{15}$$

As t becomes very large compared to τ, Equation (15) reduces to

$$\frac{D}{P} = \frac{0.0512\ f_W\ \varepsilon\ t}{m\ \lambda} = \frac{0.074\ f_W\ \varepsilon\ \tau\ t}{m} \tag{16}$$

<u>Dose Rate Per Unit Concentration in Thyroid and Limiting Specific Activity</u>

Another dose relationship of interest is the dose rate per unit activity maintained in an organ. This is calculated by the following equation.

$$\frac{DR}{Q^*} = \frac{2.22\ \text{dis/min}}{\text{pCi}} \times \frac{60\ \text{min}}{\text{hr}} \times \frac{8766\ \text{hr}}{\text{yr}} \times 1.602 \times 10^{-8}\ \frac{\text{g}\cdot\text{rad}}{\text{MeV}} \times 10^3\ \frac{\text{mrem}}{\text{rem}} \left(\frac{\varepsilon}{m}\right)$$

$$= 18.70\ \varepsilon/m \quad \text{mrem/yr per pCi in organ.} \tag{17}$$

The dose rate per unit activity in the thyroid was calculated for four age categories and the results are listed in Table 7. Since the effective half-life does not enter into Equation (17), the dose rates decrease steadily with increasing age.

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The results of the last set of calculations and the parameters given previously in Table 6 can be used to calculate the dose rate expected if all of the iodine in the thyroid were ^{129}I and also to calculate the atom ratio $^{129}I:^{127}I$ in the thyroid which would yield the limiting dose rate of 1500 mrem/yr specified for the maximum permissible dose rate to an individual member of the general public. These results have also been entered into Table 7. From the values in the table, it can be seen that, if all of the iodine in the thyroid were ^{129}I, the 1-year-old infant would receive a dose rate of about 9 times the guide of 1500 mrem/yr while the adult would receive a dose rate of about 34 times the guide. The reason for the higher adult dose rate is the higher concentration of iodine in the adult thyroid (350 ppm) as compared to the 1-year-old (90 ppm).

It appears, therefore, that the limiting atom ratio, $^{129}I:^{127}I$, must be based on the adult thyroid and should be 1:34 or 0.03 if the guide of 1500 mrem/yr is not to be exceeded. Such a ratio corresponds to 4.8×10^6 pCi ^{129}I per gram of total iodine (4.8 μCi/g).

For all ages the thyroid concentration of ^{129}I which would yield 1500 mrem/yr is 1.7×10^3 pCi per gram of total thyroid (1.7×10^{-3} μCi/g). This value is constant with age because the effective energy absorbed per disintegration is essentially constant with age for ^{129}I.

Thyroid Doses from Selected Pathways

By making reasonably conservative assumptions as to the environmental behavior of radioiodine and human dietary habits, it is possible to estimate the thyroid doses potentially received from selected pathways of exposure.

1. Doses Associated with Releases to Air

Using the inhalation rates given in Table 6 and the inhalation dose factors given in Table 7, the thyroid dose rate from continuous exposure to a unit concentration of ^{129}I in air were calculated. The results of these calculations are given in Table 8 in units of mrem/yr per pCi/m^3.

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TABLE 8. THYROID DOSES FROM UNIT CONCENTRATION OF ^{129}I IN AIR

Exposure Pathway	mrem/yr per pCi/m^3 [a]			
	1 yr	4 yr	14 yr	Adult
Inhalation [b]	16.5	8.28	13.3	29.6
Milk consumption [c]	4640	1860	1550	2330
Leafy vegetable consumption [d]	0	388	547	1110
Beef consumption [e]	0	250	375	1000

(a) Maximum dose rate after equilibrium is reached between thyroid and intake rate. Soil-root pathway would add an additional ∿1.5% (per year of accumulation) to the milk pathway and ∿1.0% (per year of accumulation) to the vegetable pathway.
(b) Inhalation rates were listed in Table 6.
(c) One liter per day of fresh milk from cow grazing on contaminated pasture 12 months per year.
(d) Leafy vegetable consumption was assumed to be 0, 32, 54 and 73 kg/yr for the 1, 4, 14 and adult ages, respectively.
(e) Beef consumption was assumed to be 0, 25, 45 and 80 kg/yr for the 1, 4, 14 and adult ages, respectively.

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The same unit concentration of ^{129}I in air will lead to somewhat higher thyroid dose rates via the milk and leafy vegetable pathways. Calculation of the dose rates from these pathways are illustrated below and the results are also summarized in Table 8.

The concentration on fresh forage is

$$C_f = \frac{C_a V_D R\,(86{,}400\ \text{sec/d})}{Y_f} \left(\frac{1-e^{-\lambda_E t'}}{\lambda_E}\right) \text{pCi/kg} \tag{18}$$

and the concentration in milk is

$$C_m = C_f\ Q_f\ S_d$$

$$= \frac{Q_f\ S_d\ C_a\ V_D R\,(86{,}400)}{Y_f} \left[\frac{\left(1-e^{-\lambda_E t'}\right)}{\lambda_E}\right] \text{pCi/}\ell \tag{19}$$

where C_a = ^{129}I air concentration, pCi/m^3
C_f = ^{129}I concentration in fresh forage, pCi/kg
C_m = ^{129}I concentration in fresh milk, pCi/liter
V_D = deposition velocity to ground, 0.01 m/sec
R = fraction of deposited radioiodine initially retained on vegetation = 0.25
Y_f = forage yield = 1.8 kg/m^2
Q_f = forage consumption by cow = 55 kg/d
S_d = fraction of daily iodine consumption appearing per each liter of milk = 0.01
λ_E = effective removal constant for deposited material as explained in previous section
t' = time between successive removal of forage by cow from same part of pasture = 30 days

Applying the above values in Equation (19) and using ^{131}I as a test nuclide the results imply a milk-to-air concentration ratio of 478.

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The average of six reported values for the ratio of C_m/C_a for ^{131}I was 560[45-49] or 1.17 times the value calculated above. On the assumption that the difference in the two values results solely from differences in linear factors such as forage yield, deposition velocity or forage consumption rate, a factor of 1.17 can be multiplied into Equation (19) to normalize the results.

When this is done and the value of λ_E for ^{129}I (0.0495) is used, Equation (19) yields a value of C_m/C_a of 1210. Assuming a milk consumption rate of 365 liters/year and the ingestion dose factors listed in Table 7, the dose from the milk pathway per unit air concentration can be calculated. These doses are listed in Table 8.

The concentration of ^{129}I on leafy vegetables can be calculated the same way as the concentration on forage was except that the yield is 1.5 kg/m^2. The consumption rate of leafy vegetables is a function of age. Maximum rates of 0, 32, 54 and 73 kg/yr were assigned to the 1 yr, 4 yr, 14 yr and adult, respectively. The doses from consumption of vegetables per unit air concentration are also tabulated in Table 8.

The ^{129}I concentrations in beef can be estimated using the parameters derived for the Year 2000 Study.[34] That is, cattle are fed 54 kg/day of stored feed, 14 kg/day of grain and 50 liters/day of water; the yield of stored feed is 2.3 kg/m^2 (fresh weight), and the yield of grain is 0.35 kg/m^2; each crop has a four-month growing season. Ten percent of the iodine deposited on the leaves will be translocated to the edible part of the grain. Two percent of the daily intake of the cattle will appear in each kg of meat. Applying these parameters we can define the following equations.

$$C_s = \frac{C_a\ V_D\ R\ (86{,}400\ \text{sec/d})}{Y_s} \left(\frac{1-e^{-120\lambda_E}}{\lambda_E}\right) e^{-\lambda_R t''}\ \text{pCi/kg} \qquad (20)$$

where C_s = concentration in stored feed, pCi/kg

Y_s = yield of stored feed (alfalfa) = 2.3 kg/m^2

t'' = decay time between harvest and consumption

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$$\frac{1-e^{-120\lambda_E}}{\lambda_E} = 20.0 \text{ for an ecological half-life of 14 days}$$

$$C_g = \frac{C_a\ V_D\ R\ (86{,}400\ \text{sec/d})}{\lambda_g}\left(\frac{1-e^{-120\lambda_E}}{\lambda_E}\right) e^{-\lambda_R t''}\ \text{pCi/kg} \qquad (21)$$

where C_g = concentration in grain, pCi/kg
Y_g = grain yield = 0.35 kg/m^2

$$C_b = (S_{kb})\ [Q_s\ C_s + Q_g\ C_g]\ \text{pCi/kg} \qquad (22)$$

where C_b = concentration in beef, pCi/kg
Q_s = consumption rate of stored feed, kg/day
Q_g = consumption rate of grain, kg/day
S_{kb} = transfer factor from feed to meat = 2×10^{-2} pCi/kg per pCi/day intake.

Assuming 1 pCi $^{129}I/m^3$ air, the concentration in beef would be:

$$C_b = (1\ \text{pCi/m}^3)(10^{-2}\text{m/s})(86{,}400\ \text{sec/d})\ (0.25)(20\ \text{days})(2 \times 10^{-2})$$
$$\times\left[\frac{54\ \text{kg/d}}{2.3\ \text{kg/m}^2} + \frac{(14\ \text{kg/d})(0.1)}{0.35\ \text{kg/m}^2}\right]$$
$$= 2370\ \text{pCi/kg}.$$

Assuming consumption rates of 0, 25, 45 and 80 kg/yr of beef for the 1, 4, 14 and adult ages, respectively, and using the dose factors given in Table 7 for ingestion, the doses from beef consumption were calculated and are included in Table 8.

Contributions from long-term accumulation in the soil of ^{129}I via airborne deposition can be calculated from Equation (23).

$$C_f = \frac{C_a\ V_D\ (86{,}400\ \text{sec/d})\ B_f}{p}\left(\frac{1-e^{-\lambda_R t^*}}{\lambda_R}\right)\text{pCi/kg} \qquad (23)$$

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where t^* = the total soil accumulation time in days

p = 224 kg dry soil/m^2 of plow layer

B_f = plant uptake factor = 0.02 pCi/kg plant (wet) per pCi/kg soil (dry)

Equation (23) is based on the assumption that all of the material deposited is retained in the plow layer where it is available to the roots and that the plant is at equilibrium with the soil at the time of harvest. For long-term accumulation, C_a may not remain constant. In that instance the following modification of Equation (23) applies:

$$C_f = \frac{V_D(86{,}400)B_f}{p} \sum_{n=1}^{t^*} (C_a)_n \, e^{-\lambda_R(t^*-n)} \text{ pCi/kg} \tag{24}$$

Because of the long half-life of ^{129}I the exponential expression $e^{-\lambda_R t^*}$ can be replaced by $1-\lambda_R t^*$ in Equation (23) yielding

$$C_f = \frac{C_a \, V_D(86{,}400 \text{ sec/d})B_f t^*}{p} \text{ pCi/kg} \tag{23a}$$

Applying Equation (23a) for a 1-year accumulation time and a constant air concentration of 1 pCi $^{129}I/m^3$ yields a forage concentration of

$$C_f = \frac{1 \text{ pCi}}{m^3} (10^{-2} \text{ m/sec})(86{,}400 \text{ sec/d})\left(2 \times 10^{-2} \frac{\text{pCi/kg plant}}{\text{pCi/kg soil}}\right)$$

$$\times (365 \text{ d/yr})/(224 \text{ kg/m}^2 \text{ soil})$$

$$= 28.2 \text{ pCi/kg.}$$

By comparison, the concentration of ^{129}I in the same forage due to direct deposition onto the leaves would be [from Equation (18)]

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$$C_f = \frac{(1\ pCi/m^3)(10^{-2}\ m/sec)(0.25)(86{,}400\ sec/d)}{1.8\ kg/m^2}\left(\frac{1-e^{-30\lambda_E}}{\lambda_E}\right)$$

$$= 1870\ pCi/kg.$$

Therefore, the soil-root pathway will contribute about 1.5% per year of accumulation to the ^{129}I concentration present in fresh forage from direct deposition. The same ratio would also apply to milk produced by cows grazing on such forage.

Assuming a growing season of 90 days and a yield of 1.5 kg/m^2, fresh leafy vegetables exposed to an air concentration of 1 pCi $^{129}I/m^3$ would contain 2870 pCi/kg via foliar deposition and 28.2 pCi/kg (1% per year of accumulation) via root uptake.

2. Doses Associated with Releases to Water

The thyroid dose from consumption of ^{129}I with drinking water or aquatic foods can be calculated from the concentration in the food or water and the dose factors given in Table 7. The concentration in the aquatic foods can be obtained from Equation (25).

$$C_A = C_W\ B_A \tag{25}$$

where

C_A = concentration in aquatic foodstuff, pCi/kg
C_W = concentration in water, pCi/liter
B_A = bioaccumulation factor from Table 4, pCi/kg per pCi/liter

An avid fisherman might consume 18 kg/yr each of fish and invertebrates (shellfish and molluscs) in addition to about 2 liters/day of drinking water. If all of these items were derived from fresh water containing 1 pCi ^{129}I/liter the resultant thyroid doses would be as follows.

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Drinking Water

$$(DR)_W = (2\ \ell/d)(365\ d/yr)(5.28 \times 10^{-3}\ mrem/pCi)$$

$$= 3.85\ mrem/yr$$

Fish

$$(DR)_F = (18\ kg/yr)(15\ pCi/kg)(5.28 \times 10^{-3}\ mrem/pCi)$$

$$= 1.43\ mrem/yr$$

Invertebrates

$$(DR)_I = (18\ kg/yr)(5\ pCi/kg)(5.28 \times 10^{-3}\ mrem/pCi)$$

$$= 0.475\ mrem/yr$$

Total dose rate from all three sources is 5.8 mrem/yr.

If in addition the same water was used to irrigate farm land, an additional intake of ^{129}I would result from the consumption of farm products. Assume sprinkler irrigation applied at the rate of 110 $\ell/m^2/month$[34] for a 12-month forage season and for a 3-month vegetable growing season. In addition, assume radionuclides reaching the plant and soil via sprinkler irrigation behave the same as those deposited from the air.[34] Under these assumptions, the following thyroid doses can be calculated.

$$C_m = \frac{(110\ \ell/m^2/month)(1\ pCi/\ell)(0.25)}{(1.8\ kg/m^2)(30\ d/month)} \left(\frac{1-e^{-30\lambda_E}}{\lambda_E}\right)$$

$$(55\ kg/d)\left(0.01\ \frac{pCi/\ell}{pCi/d}\right)(1.17)$$

$$= 5.12\ pCi/\ell$$

$$(DR)_m = (5.12\ pCi/\ell)(365\ \ell/yr)(5.28 \times 10^{-3}\ mrem/pCi) = 9.87\ mrem/yr.$$

Leafy Vegetables

$$C_v = \frac{(110\ \ell/m^2/\text{month})(1\ \text{pCi}/\ell)(0.25)}{(1.5\ \text{kg}/m^2)(30\ \text{d/month})}\left(\frac{1-e^{-90\lambda_E}}{\lambda_E}\right) = 12.2\ \text{pCi/kg}$$

At a consumption rate of 73 kg/yr, the adult thyroid dose would be

$(DR)_v$ = (12.2 pCi/kg)(73 kg/yr)(5.28 x 10^{-3} mrem/pCi) = 4.70 mrem/yr if all the vegetables consumed during the year contained 12.2 pCi/kg.

Radiation Doses to Biota Other than Man

Radiation doses to the thyroid of certain mammals from radioiodine intake have been calculated from the concentrations (activity per unit mass) measured either *in vivo* or after sacrifice of experimental animals. Doses to aquatic organisms such as algae, invertebrates and fish, are generally calculated as doses to the total organisms, since aquatic bio-accumulation factors are given in those terms, and since many of the organisms do not have a discernible thyroid.

If the concentration of ^{129}I in the thyroid (or total body) of an animal is known, Equation (17) can be used with a simple modification. Re-writing the equation in terms of concentration rather than total activity, yields:

$$DR = 18.7\ \varepsilon\ (Q^*/m)\ \text{mrem/yr} \qquad (26)$$

where Q^*/m = the concentration in pCi/g in the thyroid (or total body)

ε = the effective energy per disintegration in the organ (or total body)

If the biological parameters f_a, f_w, τ are known for the animal in question, then the concentration in the organ could be calculated from the daily intake of radioiodine. Of course, the animal's diet and the concentrations in its air, water and food supply would also need to be known or estimated by equations given in the previous section.

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Radiation doses to aquatic organisms can also be obtained from Equation (26) by first utilizing Equation (25) to calculate concentration in the organism, provided the water concentration and bioaccumulation factors are known. Combining these two equations yields Equation (27).

$$DR = 18.7\ C_W\ B_A\ \varepsilon\ \text{mrem/yr} \tag{27}$$

where DR = the average dose rate to the entire organism.

External doses to terrestrial and aquatic organisms from ^{129}I are generally insignificant compared to internal doses, because of the low energy of the beta and gamma radiations emitted.

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FIELD SAMPLING PROGRAM

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FIELD SAMPLING PROGRAM

INTRODUCTION

Field sampling has been undertaken at four locations: two in the arid steppe region of the western United States and two in deciduous forests located in higher rainfall areas of the eastern United States. Sites were chosen so that one landscape which had been exposed to airborne ^{129}I from nearby nuclear fuels reprocessing plants could be compared to another landscape of similar climate remote from nuclear facilities. The objective of the sampling program was to determine, for undisturbed natural field conditions, the potential sites of accumulation of airborne ^{129}I in the landscape, that is, in the soil-plant-waterway part of the ecosystem. A more sophisticated sampling program, intended to elucidate iodine relationships of food webs and iodine movement and accumulation characteristics in other parts of the ecosystems studied can be developed later on the basis of information obtained in these early studies.

Samples generally fell into three categories: 1) long-standing (persistent) above-ground vegetation such as mosses, lichens and leaves of evergreen trees, 2) recent litter and humus and underlying mineral soil in areas of minimal disturbance, mostly deciduous forests and old fields, and 3) bogs, ponds and streams with associated biota which would be expected to receive iodine washed from terrestrial areas. The sampling programs for the two regions were conducted concurrently by the same team of investigators with the analyses being done by the same procedures in order to allow valid comparisons of the sites.

COLLECTION METHODS

Where practicable, terrestrial vegetation samples were taken on a per unit area basis in order to allow comparisons to be made from site to site. Soil profiles were studied, generally in fairly mature second growth deciduous forests or abandoned fields or pastures. Separate collections were made of the herb-litter layer, of the upper 2.5 cm of soil and of 2.5 cm

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layers at deeper depths in the soil profile. Most soils in the forest sites were shallow, i.e., less than 30 cm. Precautions were taken to prevent contamination of the deeper soil samples by overlying material. All samples were sealed into plastic bags at the time of sampling to prevent contamination by airborne ^{129}I. They were frozen as soon as practicable and were stored frozen until they could be analyzed. Some large soil samples were not frozen for lack of space.

COLLECTION SITES AND SAMPLE DESCRIPTIONS

Arid Steppe Sites

Two arid sites were studied. One of these was located within the Rattlesnake Hill Research Natural Area (Arid Lands Ecology Reserve) which is a part of the USAEC Hanford Reservation, Benton County, southeastern Washington. A companion site approximately 200 miles southeast of the Hanford Reservation was on land owned by the Bureau of Land Management five miles north of the town of Vale, Oregon.

On the Hanford Reservation, vegetation and soil samples were taken at sites located about 10 miles west of the chemical separations plants (200 E and 200 W). One site is located at an altitude of 1000 feet (305 m) (Figure 4A) and the other at 1700 feet (518 m) (Figure 4B). The site at the higher altitude receives slightly more precipitation than that at the lower altitude. However, the two soils are similar as to texture, pH, and organic matter content. The parent material is loess and is at least 1 m deep. Both sites are unique in that they have been mostly ungrazed and unburned for 30 years, the age of the Hanford Plant. Soil moisture profile studies have shown that water penetrates about 1 m at the lower elevation site and about 1.5 m at the higher site. Both sites are abandoned fields that have supported more or less uniform stands of cheatgrass for 30 years. Cheatgrass effectively reduces soil erosion by wind and water action, and, over the years, a substantial layer of dead plant material (mulch) has accumulated over the mineral soil. Because of the low rainfall and the lack of use of these fields in recent history it is expected that most of the

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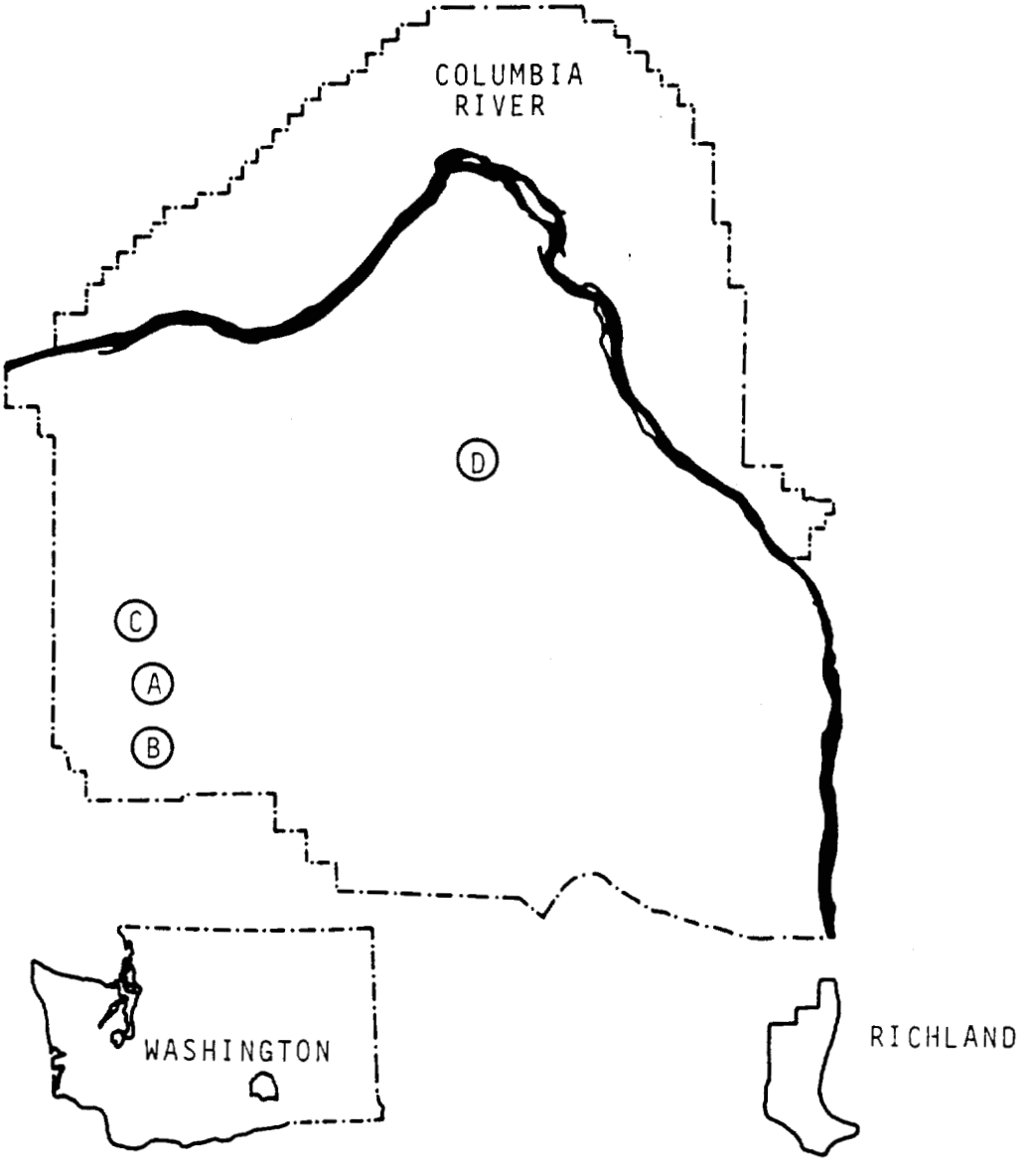

FIGURE 4. Map of the Hanford Reservation Showing Sampling Sites

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non-volatile ^{129}I that has been deposited over these fields is still present between the top of the vegetation canopy and the maximum depth of soil water penetration.

At each elevation, three replicate sets of samples were taken. All live and dead plants were collected from a 0.5 m^2 circular plot. A second collection was made of the upper 2.5 cm layer of surface soil from the same plot. Then two deeper samples were taken, one at 15 cm and one at 60 cm below the soil surface. To obtain these deep samples, a sharpened 15-cm-diameter pipe was driven into the soil at two locations within the 0.5 m^2 plots to the desired depth. The sampler was then twisted to break continuity at the bottom and was removed carefully to retain the core within the tube. Then from the center of the bottom of the soil column, a disc-shaped sample, 7.5 cm in diameter and 2.5 cm thick, was removed with a clean spatula. This technique gave samples uncontaminated by the passage of the tube through overlying soil layers. The samples from each depth from the two cores were pooled at the laboratory before analysis. Identical samples were taken from the pasture site at Vale, Oregon.

Aquatic plants and animals were collected at two locations on the Hanford Reservation, one at Rattlesnake Springs (Figure 4C) and the other at Gable Mountain Pond (Figure 4D). Rattlesnake Springs is one of the two major springs occurring in this otherwise arid area. The spring produces a small stream which varies in length from 2.4 to 3.2 km before it seeps back into the ground. Flow is approximately 1 ft^3/sec (28 liters/sec) except during winter when snowmelt creates flash floods of many times this volume. The stream is about 6.4 km southwest of one of the nuclear chemical processing plants and would be expected to receive airborne effluents from this plant. The stream bed is a mixture of basalt fragments, sand-clay sediments and organic detritus. Vegetation samples were taken from the lower portion of the stream; sediment and cattail samples were taken from a catch-basin type pool. Sampling was done in spring when growth was just beginning. Watercress was only 7 to 10 cm long and cattail leaves

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were only 60 to 90 cm long. Both roots and leaves were included in the plant samples. The sediment sample was taken from the top 7 to 10 cm of the bottom profile.

Plants, sediments and goldfish were taken from Gable Mountain Pond. This pond, located about 1.6 km north of the chemical processing facility, is approximately 50 acres (20 hectares) in surface area with depths ranging up to 2.7 m. Bottom materials range from sand-clay to organic detritus composed primarily of plant materials. The shoreline and islands of the pond are heavily populated with cattails and rushes. This is an artificial pond developed to receive low-level radioactive liquid effluents from the chemical processing facility. It contains measurable quantities of a number of fission products.

Samples were taken from near the southwest shore of Gable Mountain Pond in spring when the year's growth was just beginning. Cattail leaves were only 60 to 90 cm in length. Both roots and leaves were included in the cattail sample; leaves and stems of water milfoil were collected. Only the throat or thyroid region (tip of the lower jaw to the base of the pectoral fins) was sampled from goldfish. Tissues from 22 fish were excised in the field immediately after the fish were caught and were pooled for a single analysis. Sediment was taken from the top 7 to 10 cm of the pond bottom.

Deciduous Forest Sites

Two collection locales were chosen in the eastern United States, one in the environs of the Midwest Fuel Recovery Plant (MFRP) at Morris, Grundy County, Illinois (near Joliet) and the other in the environs of the Nuclear Fuels Service (NFS) plant in Cattaraugus County, western New York. The sampling in New York was done in early May and that in Illinois in early June.

The MFRP had not begun operation at the time of field collection so that the samples will serve to give baseline data on the ^{129}I levels in the environment. Since this reprocessing plant is very close to the

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Dresden reactors, which could have released small amounts of ^{129}I over the past several years, a forest system was sampled in Pilcher Park near Joliet, Illinois, to determine ^{129}I levels at a distance of at least 24 km from the Dresden reactors. These Pilcher Park samples will serve as comparisons for both the New York samples and the samples taken nearer to the MFRP.

Three main sampling sites were chosen: one, the Pilcher Park Site, an oak-maple-beech forest about 14 km east of Joliet, Illinois; two, an oak-hickory forest across the Illinois River from the MFRP site (Figure 5C); and, three, a pasture on the MFRP property on which cows are maintained (Figure 5A). We got samples of milk from these cows.

Litter, herb and soil samples were collected as follows: standing herbs and litter were taken from a measured circle with a radius of 40 to 60 cm, then the upper 2.5 cm of soil was collected from a smaller radius circle, then a hole was dug to the 15 cm depth and a third collection of a 2.5 cm layer of soil was made, and, finally, a deeper hole was dug (where possible) to allow the collection of a 2.5 cm layer at a deeper depth. This sampling procedure was adopted because rocky soils would not allow the coring technique to be used. At each forest collection site, a bag of understory shrub leaves was collected. Approximately 250 cm^2 of a moss mat was sampled at the Pilcher Park site, and a juniper bush was sampled from a lawn immediately across the Des Plaines River from the Dresden reactors (Figure 5D). Two water samples were collected: one from the Illinois River downstream of the plant and, two, from the Michigan-Illinois canal across the river from the plant (Figure 5B).

In the first week in May, 33 samples were taken from the environs of the NFS plant in Cattaraugus County, western New York State. Generally, samples were taken from near the plant boundaries within sight of roads which penetrate the general boundaries of the plant.

The vegetation mosaic of the New York site consists of deciduous forest stands interspersed among open fields of pasture grasses. Steep, north-facing slopes support stands of conifers, especially eastern hemlock. The important deciduous trees are american beech and sugar maple. The stands

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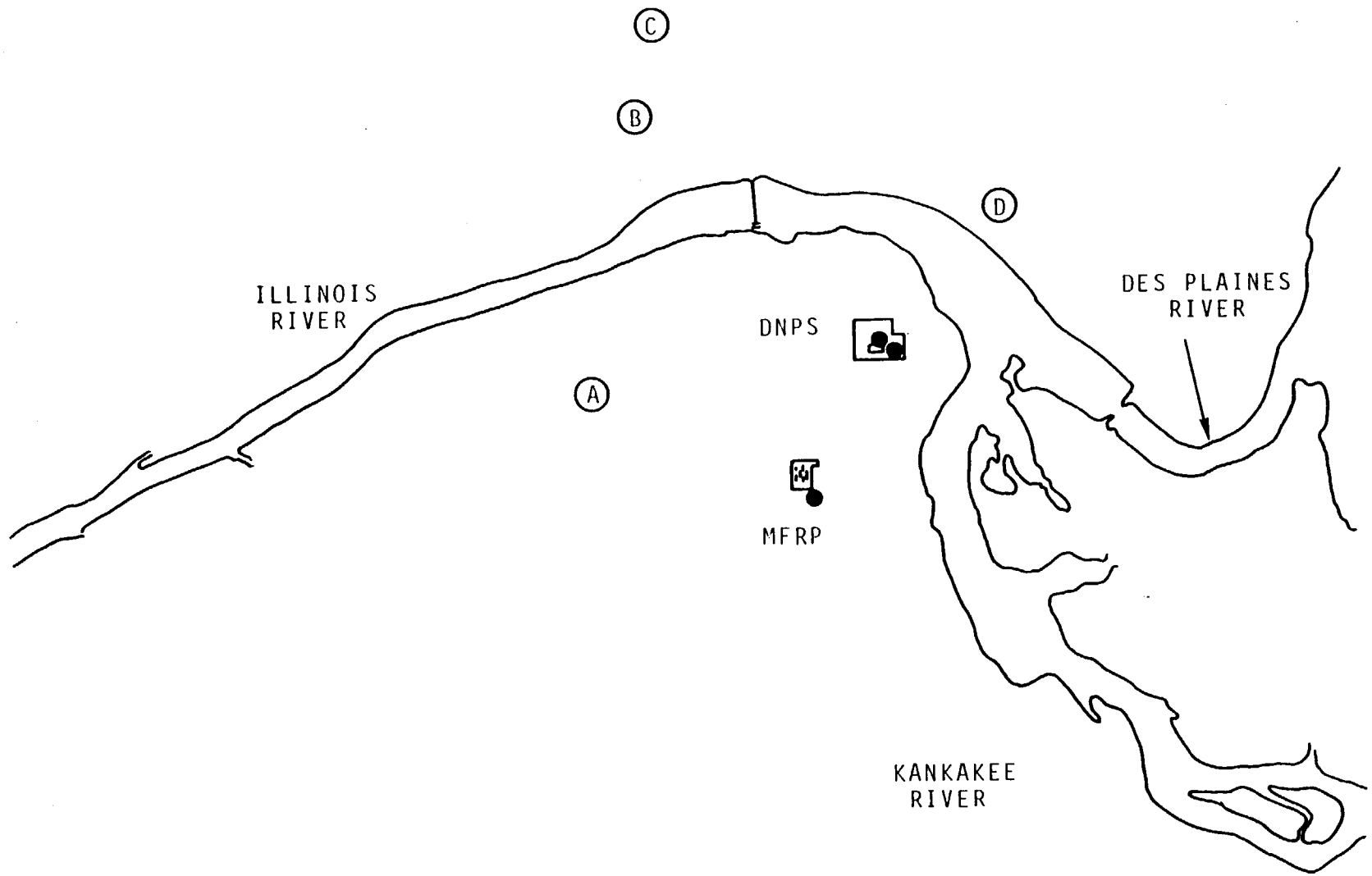

FIGURE 5. Map of the Environs of the Midwest Fuels Recovery Plant (MFRP) Showing Sampling Sites

all have a logging history. At the time of sampling, deciduous trees were putting out new leaves. Some of the early flowering herbs such as Trillium were in bloom. The understory of the stands sampled consisted of sparse herbs and shrubs; however the forest floor was completely covered by a layer of matted tree leaves. The soil in these forest stands was shallow (less than 30 cm deep) over fractured rock. At the time of sampling the soil was saturated with water from recent rainstorms. One would expect water movement through these shallow soils into the rocky substrate would be a relatively rapid process and much of the ^{129}I would be moved into ground water and out of the system.

Most samples were taken from within a 3 km radius of the stack. Sites included two second-growth stands in beech-maple forests (Figure 6C and F), a hemlock stand (Figure 6D), and an abandoned pasture (old field) (Figure 6B). Litter, herbs and soils were taken as for the Illinois samples.

A collection was made of a lichen, Physcia millegrana Degel., and a moss mat, consisting of about three parts of Platygyrium repens (Brid.) B.S.G. and one part Brachythecium salebrosum (Web. and Mohr) B.S.G. These were both growing on a tree trunk about 2 km east of the stack (Figure 6E). A sample of Hypnum curvifolium Hedw. on a unit area basis was taken from a ridge near the confluence of Buttermilk Creek and Cattaraugus Creek (Figure 6A). Samples of honey (fall collection of 1972) and maple syrup (spring of 1973) were purchased from a farm less than 8 km southeast of the stack.

Aquatic samples were taken from Cattaraugus Creek about 1 km downstream from its confluence with Buttermilk Creek (Figure 6A). Buttermilk Creek flows within 0.8 km of the NFS plant and has received radioactive liquid and gaseous effluents from the plant. The average flow of Buttermilk Creek is 46.5 ft^3/sec (1.3 m^3/sec) and of Cattaraugus Creek, 354 ft^3/sec (10 m^3/sec). Cattaraugus Creek discharges into Lake Erie approximately 39 miles (62 km) downstream of its confluence with Buttermilk Creek.

Samples were collected upstream of Felton Bridge near the north perimeter fence. According to NFS personnel, Cattaraugus Creek is typically quiet,

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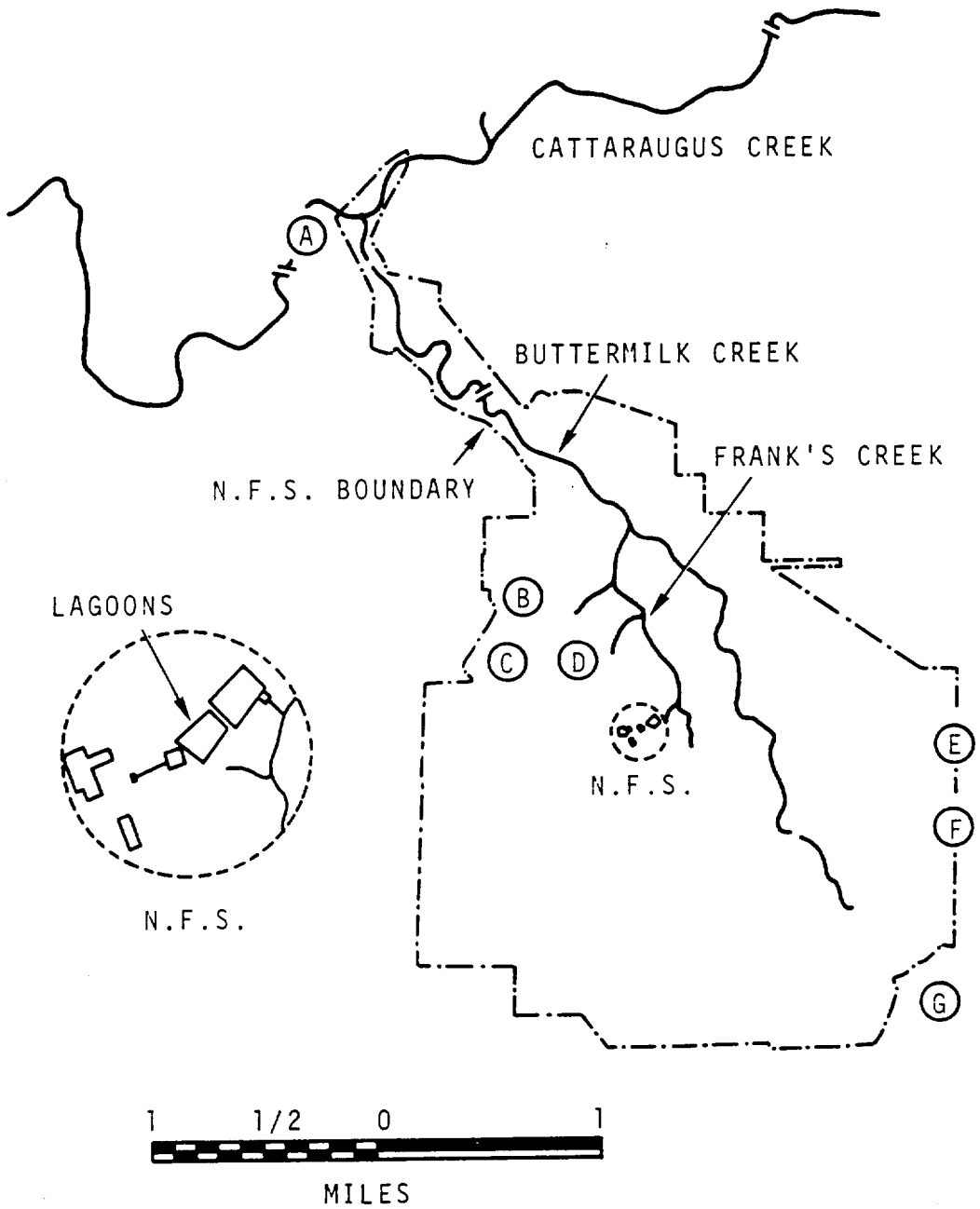

FIGURE 6. Map of the Environs of the Nuclear Fuels Service (NFS) Plant Showing Sampling Sites

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turbid and subject to wide fluctuations in flow with scouring of the stream bed. At the point of sampling, the stream bottom is composed of a mixture of angular stones and fine silts with the deeper holes having sand-silt bottoms. The stones were covered with a 0.3 cm thick layer of fine silt of the consistency of glacial flour. Stream turbidity was high with visibility less than 30 cm. Fish and crayfish were collected with a seine from the slower moving sections of the stream. Throat ("thyroid") tissues were removed in the field in the manner described for Gable Mountain Pond. Tissues from 13 white suckers ranging in length from 7.6 to 25 cm were pooled for one analysis, and tissues from 24 common shiners, from 5 to 10 cm in length, were combined for another analysis. Two crayfish were collected for another sample. Sediments were taken near the downstream side of the right pier of Felton Bridge. Fish samples were kept on ice for about 36 hours before being frozen.

A cattail sample (*Typha latifolia*) and a collection of *Brachythecium rutabulum* (Hedw.) B.S.G. were taken from a small pond located near the southeast corner of the plant perimeter fence (Figure 6G). This pond was approximately a quarter of an acre in area with a maximum depth of less than 1 m. The bottom was composed of a putrid-smelling mixture of fine silts and organic debris. Plant growth was just beginning; leaves were less than 60 cm long. Both roots and leaves were combined in a single sample.

ANALYTICAL METHODS

Methods available for measuring the ^{129}I content of environmental samples include procedures based on neutron activation analysis, mass spectrometric analysis, and liquid scintillation counting (1-8). All of these methods require that iodine be separated from the sample and isolated in a relatively pure form prior to measurement.

Activation analysis and mass spectrometric procedures determine directly the ratio of ^{129}I to ^{127}I. To calculate the ^{129}I concentration in the sample requires a measure of the chemical yield of iodine in the

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separation steps and a determination of the amount of either ^{129}I or ^{127}I separated. Chemical yield can be determined by spiking the raw sample with a known activity of another iodine ratioisotope (e.g., ^{125}I) prior to separation and then measuring the recovery of that isotope after separation. Thus, the activation analysis procedure can measure both the ^{129}I:^{127}I ratio and the total amount of each of the iodine isotopes separated. As little as 4×10^{-6} pCi of ^{129}I can be measured by neutron activation analysis.

Direct counting methods such as liquid scintillation counting for ^{129}I are not applicable to most environmental samples due to their limited sensitivity of about 10 pCi. Other limitations include the inability to use radioactive spikes for yield measurements, the need to measure ^{127}I separately when ^{129}I is counted, and the interference of radioactive impurities remaining in the separated iodine fraction which can result in a high estimate of ^{129}I content.

The steps used by our laboratory for processing solid environmental field samples are based on the activation procedure developed by Studier et al.[2] These steps are listed below:

1. Total sample weight is determined.
2. An aliquot of the sample to be used for analysis is freeze-dried. The weight loss is measured.
3. The gamma radioactive radionuclide content of the sample is measured by gamma-ray spectrometry.[9-15]
4. A known amount of ^{125}I spike is added to the sample and iodine is separated from the sample by combustion in oxygen and trapping of the iodine on activated charcoal.[2,19]
5. The separated iodine is irradiated with an iodine comparator standard for 8 to 24 hours in a nuclear reactor.
6. The irradiated iodine sample is purified by distillation and solvent extraction techniques.[2,19]

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7. The purified irradiated iodine fractions from the sample and comparator standard are counted by low-level gamma-ray spectrometry.[9,16,17]

8. Computer programs are used to process counting data, calculate ^{129}I and ^{127}I estimates, and tabulate results.[12,18]

Liquid samples, such as water and milk, are normally spiked with ^{125}I and processed by adsorption of the ionic iodine on a Dowex-1 ion exchange resin column. The iodine is removed from the resin by combustion of the resin as outlined above in Step 4.

Solid samples or resin beds are ignited in a quartz combustion apparatus at high temperatures (up to 1000°C) in a stream of oxygen to free the iodine. The iodine is collected on a small bed of activated charcoal using a procedure developed by H. Tenny.[19] Combustion of the charcoal trap in oxygen and trapping of the iodine on activated charcoal gives further purification of the iodine. Iodine is then removed from the charcoal by heating in a vacuum system and is trapped in a quartz tube at liquid nitrogen temperature. The tube is sealed to make a quartz irradiation ampoule.

Quartz ampoules containing iodine separated from the samples are irradiated for eight to twenty-four hours. Irradiations are performed at the Hanford N-reactor or at the Washington State University (WSU) Triga reactor. Comparator standards containing ^{125}I, ^{127}I, and ^{129}I are irradiated with each set of samples.

The neutron capture reactions used in iodine activation analysis are:

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

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(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, iodine is further purified by distillation and solvent extraction according to procedures similar to those given in Reference (2). It is then precipitated as AgI and mounted on thin plastic scintillators for counting.

The ^{126}I, ^{128}I and ^{130}I activities produced in samples and in comparator standards during irradiation are measured by gamma-ray spectrometry using several spectra over time to obtain half-life information. Well-type NaI (Tl) detector systems are generally used for the measurements. The ^{130}I sum peaks at 1.9 MeV and 2.3 MeV are used to estimate ^{130}I activity and to discriminate against interferences. The low-level, beta-gated, multiple-gamma coincidence spectrometry techniques described previously (7,9,16,17,18) are used when required to measure very small amounts of ^{130}I. Measurements are corrected for interfering activities produced by reactions (6) and (7). The post-irradiation purification also limits interference from reaction (7). Interference from reaction (4) is minimized by using small levels of ^{125}I such that reaction (2) predominates.

Reaction (5) limits the increase in sensitivity that can be obtained by increasing the exposure time and neutron flux. Neutron exposure conditions are 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 percent.

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Low-level ^{125}I measurements required for estimation of chemical yield, which is needed to measure the amount of the ^{127}I and ^{129}I in irradiated comparator standards, are performed with a special gamma-X-ray coincidence system.[9]

The ^{129}I:^{127}I ratio in the comparator standard has been calibrated by activation analysis relative to a mixed ^{129}I:^{127}I standard prepared by M. H. Studier who diluted a known amount of ^{129}I (analyzed by mass spectrometry) with a known amount of natural iodine.[20] The amount of natural iodine relative to the ^{125}I activity in the comparator standard is determined by activation analysis using a weightable C_6I_6 standard. A ^{129}I standard has recently been announced by the National Bureau of Standards and will be used to check our calibrations. Sensitive mass spectrometric techniques for checking our standard calibrations are also being developed.

Components in the time-dependent gamma-ray spectra of a sample are calculated using a weighted least-squares method.[18] Data handling programs outlined in Reference (12) are used for calculation of analytical results from the sample and standard activity ratios and for preparation of final data tabulations. Corrections are made for interferences, chemical yield, laboratory blanks, and sampling blanks, where applicable. The results obtained are the ^{127}I and ^{129}I concentrations per unit amount of sample and the ^{129}I:^{127}I ratio. Ratios to other fission and activation products can be calculated from gamma-ray spectrometric measurements on the 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 ionic iodine is separated. Organic bound iodine may not be completely separated from the sample. The same is true for air sampling. No air sampler has been developed which has been shown to completely trap all forms of atmospheric iodine.

The elapsed time required for analyses and the man-hours required for analyses depend on sample characteristics. Operational and computing

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economics favor large batch operations and also extend the time required for analyses. Elapsed time required for the various analysis steps are listed below:

Operation	Elapsed Time Days
Freeze drying of sample	0-14
Isolation of iodine	1-7
Activation in nuclear reactor (including transportation)	1-7
Post-irradiation chemistry	1
Counting and calculating	14-180

Additional time is required between some steps to accumulate sufficient batch sizes for economical operation. The long counting times result from the need to check the ^{126}I 13-day and ^{125}I 60-day half-lives. The need for this varies with the natural iodine levels of the samples and the chemical yield. Results can generally be calculated within two to four weeks of the reactor irradiation depending on the computer processing schedule. The most economical operation requires three to four months elapsed time from sample receipt to results if samples are received in sufficiently small numbers that all analyses can be initiated immediately. Elapsed analysis times as rapid as one or two weeks for a small number of samples (3-4) are possible, but not economical.

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RESULTS AND DISCUSSION

The results presented in the following tables were calculated on the basis of the counting completed at the time this report was prepared. The numbers presented are not expected to be changed significantly with future counting. However, some samples have not been sufficiently processed to permit calculations of meaningful numbers and results will be available at a later date.

Arid Steppe Sites

Of the samples taken from arid sites, only the ones from the Hanford Reservation have had analyses completed. The results from the litter and soil samples are presented in Table 9.

As shown in Table 9, the greatest concentrations of ^{129}I were measured in the herb-litter layer and in the surface 2.5 cm of soil. The largest quantities of ^{129}I per m^2 were accumulated in the upper 2.5 cm of soil that contained a large amount of old organic matter. Penetration of ^{129}I into the soil profile appeared to be limited. The concentration measured at the 60 cm depth was approximately 1 percent of that measured in the surface soil.

This gradient of ^{129}I concentration with soil depth can be contrasted with the gradient for ^{127}I, which presumably has been in the soil profile for a very long time. The ^{127}I concentration at the 60 cm depth was approximately twice that in the surface soil at the lower elevation (Figure 4A) and approximately three-fold greater at the upper elevation (Figure 4B). However, for practical purposes, the stable iodine can be considered to be distributed throughout the upper 60 cm of soil. It has been leached downward in the profile to only a moderate degree with the annual rain water penetrations of this arid area.

The total sum of ^{129}I (calculated from these results and from bulk density estimates) in the upper 60 cm of the profile was about 89 pCi/m^2 and 50 pCi/m^2 at the lower and upper fields respectively. The lower field has 45 percent more ^{129}I. This may result in part from its closer location

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TABLE 9. IODINE CONTENTS OF SOIL PROFILE OF LOWER AND UPPER SNIVELY RANCHES

Sample	Atom Ratio $^{129}I:^{127}I$	pCi of ^{129}I Per g*	pCi of ^{129}I Per m^{2**}	ng ^{127}I Per g*	ng ^{127}I Per m^{2**}
		Lower Snively Ranch (Figure 4A)			
Litter-herb	$4.5 \times 10^{-5} \pm 4.0 \times 10^{-6}$	$2.9 \times 10^{-3} \pm 1.0 \times 10^{-4}$	2.0	360 ± 20	2.5×10^{5}
Surface 2.5 cm of soil	$3.6 \times 10^{-5} \pm 2.1 \times 10^{-6}$	$3.3 \times 10^{-3} \pm 2.6 \times 10^{-4}$	67.2	520 ± 45	1.1×10^{7}
A 2.5 cm layer at 15 cm	$7.4 \times 10^{-7} \pm 1.3 \times 10^{-7}$	$5.8 \times 10^{-5} \pm 4.9 \times 10^{-6}$	1.9	410 ± 15	6.7×10^{7}
A 2.5 cm layer at 60 cm	$8.1 \times 10^{-8} \pm 1.1 \times 10^{-8}$	$1.9 \times 10^{-5} \pm 2.3 \times 10^{-6}$	0.6	1300 ± 42	7.6×10^{8}
		Upper Snively Ranch (Figure 4B)			
Litter-herb	$3.1 \times 10^{-5} \pm 2.9 \times 10^{-6}$	$1.9 \times 10^{-3} \pm 3.0 \times 10^{-4}$	0.9	350 ± 35	1.8×10^{5}
Surface 2.5 cm of soil	$1.9 \times 10^{-5} \pm 8.8 \times 10^{-7}$	$2.1 \times 10^{-3} \pm 8.8 \times 10^{-5}$	39.2	600 ± 38	1.1×10^{7}
A 2.5 cm layer at 15 cm	$3.5 \times 10^{-7} \pm 6.4 \times 10^{-8}$	$4.5 \times 10^{-5} \pm 9.4 \times 10^{-6}$	1.5	720 ± 78	1.2×10^{8}
A 2.5 cm layer at 60 cm	$1.1 \times 10^{-8} \pm 1.5 \times 10^{-9}$	$5.3 \times 10^{-6} \pm 7.8 \times 10^{-7}$	0.2	2700 ± 120	1.6×10^{9}

* Basis is grams freeze-dried weight.

** Calculated from the average bulk density (1.3).

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to the chemical separations plant but may also be related to the differences in moisture penetration patterns at the two levels. The opposite was found for stable ^{127}I between the two sites with more ^{127}I being present, especially in the deeper soil layers at the upper site. At present no good reason can be given for the differences in accumulation of these two iodine isotopes at the two sites. Also there were not enough samples collected over a large enough area to prove if the differences measured were significant.

Values for the atom ratio (^{129}I:^{127}I) in Table 9 show the distribution relationships to be expected from the results discussed above for the two isotopes. There was a definite decrease of ^{129}I content with increasing soil depth and a slight increase of ^{127}I with depth so that the atom ratio fell from the order of 10^{-5} at the surface to 10^{-8} at the 60 cm depth.

There appears to have been no rapid downward movement in the soil profile of the ^{129}I with soil water as might be expected. Soils are generally considered to be cation exchangers. The negatively-charged exchange sites would reject the iodide anion, theoretically allowing the iodine to move with the soil water. It must be assumed at this time with the evidence presented that, if ^{129}I is moving as an anion, its movement through the soil profile is slowed by some soil component, possibly organic matter or positively-charged sites. Possibly the erratic upward and downward movement of soil water during the wet and dry seasons of this arid region give iodine movements which are not readily elucidated.

It can be postulated that over a very long period of time, if no more ^{129}I is added to the system, the ^{129}I now in the soil will distribute itself throughout the upper zones of the soil profile in a similar way to that shown for ^{127}I in the table. The accumulation of ^{127}I at the 60 cm depth is not unlike that for cations such as calcium that accumulate near the depth of the annual soil moisture penetration.

The aquatic samples (Table 10) were taken to act as a guide for more extensive sampling to be done at the end of the growing season (September 1973). As was expected, Gable Mountain Pond sediment and biota showed

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TABLE 10. SUMMARY OF ^{129}I AND ^{127}I IN AQUATIC SAMPLES OF THE HANFORD RESERVATION, 1973

Sample	Atom Ratio $^{129}I:^{127}I$	pCi ^{129}I Per g	ng ^{127}I Per g
	Rattlesnake Springs		
Sediment	7.1×10^{-7}(1)*	9.9×10^{-4}(1)*	7.9×10^{3}(1)*
Watercress	2.8×10^{-7}(1)	2.8×10^{-4}(1)	5.7×10^{3}(1)
Cattails	4.2×10^{-7}(1)	3.6×10^{-5}(1)	4.8×10^{2}(1)
	Gable Mountain Pond		
Sediment	5.3×10^{-4}(1)	1.2×10^{-2}(1)	1.3×10^{2}(1)
Cattails	2.1×10^{-3}(1)	3.0×10^{-2}(1)	8.2×10 (1)
Myriophyllum	3.4×10^{-3}(1)	1.7 (1)	2.8×10^{3}(1)
Gold fish "thyroids"	1.2×10^{-2}(1)	8.3 (1)	4.0×10^{3}(1)

* Numbers in brackets refer to the number of replicates.

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higher levels of ^{129}I than did Rattlesnake Springs materials. Cattails, which had been chosen as a plant likely to be available at all collection sites so that comparative information could be obtained, unfortunately does not appear to be a good indicator species for iodine. In the Gable Mountain Pond samples, there appears to be an accumulation of ^{127}I from the sediment to Myriophyllum and from the Myriophyllum to the gold fish "thyroids." Curiously enough, the accumulation of ^{129}I appears to be even greater and consequently the atom ratio, ^{129}I:^{127}I, increases up the food chain. Of course, these samples are unreplicated and were taken to do no more than indicate the sampling design which should be used for the next study. Obviously more data are needed before any conclusions can be drawn.

Deciduous Forest Sites

Data from the collections made at Morris, Illinois, are given in Table 11. As expected, ^{129}I was detectable with the methods used. The oak-maple-beech forest was sampled at Pilcher Park, about 24 km away from the MFRP site. Any differences between the values obtained at Pilcher Park and those obtained from near the Dresden reactors (oak-hickory forest and grassy pasture) are slight and probably are not significant. For example, at all sites there was of the order of 10^{-6} pCi ^{129}I per gram of soil and atom ratios were of the order of 10^{-8} to 10^{-9} (^{129}I:^{127}I).

In all collections there is a marked decrease in ^{129}I concentrations with depth. Generally, this decrease is one order of magnitude for every 15 cm of depth. One curious fact emerges. In both forest collections, the young leaves in the understory trees had higher ^{129}I:^{127}I ratios than occurred in the soil. This is in part caused by the fact that these leaves have lower ^{127}I contents than other materials (for example, the herb-litter layer). However, the reason for this apparent "enrichment" in ^{129}I for understory leaves is not at all clear on the basis of presently available data.

Values for the area around the Nuclear Fuels Service plant at West Valley, New York, are presented in Table 12. Three forest habitats

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TABLE 11. SUMMARY OF ^{129}I AND ^{127}I IN VEGETATION AND SOILS IN THE ENVIRONS OF THE MIDWEST FUEL RECOVERY PLANT AT MORRIS, ILLINOIS, 1973

Sample	Atom Ratio $^{129}I:^{127}I$	pCi ^{129}I Per g	pCi ^{129}I Per m^2	ng ^{127}I Per g	ng ^{127}I Per m^2
		Oak-Maple-Beech Forest			
Litter-herb layer	1.5×10^{-8}(2)	4.2×10^{-6}(2)	2.8×10^{-3}(2)	1.7×10^{3}(2)	1.0×10^{6}(2)
Surface 2.5 cm layer of soil	1.9×10^{-9}(2)	6.5×10^{-6}(2)	6.0×10^{-2}(2)	1.9×10^{4}(2)	1.7×10^{8}(2)
2.5 cm layer of soil at 15 cm	1.3×10^{-10}(1)	8.3×10^{-7}(1)	1.5×10^{-2}(1)	3.6×10^{4}(1)	6.3×10^{8}(1)
Understory maple leaves	7.1×10^{-8}(1)	2.2×10^{-6}(1)	----	1.7×10^{2}(1)	----
Moss mat.	7.9×10^{-8}(1)	1.2×10^{-5}(1)	3.9×10^{-3}(1)	8.6×10^{2}(1)	2.8×10^{5}(1)
		Oak-Hickory Forest (Figure 5C)			
Litter-herb layer	9.7×10^{-8}(2)	8.4×10^{-6}(2)	6.9×10^{-3}(2)	4.9×10^{2}(2)	4.1×10^{5}(2)
Surface 2.5 cm layer of soil	1.2×10^{-8}(2)	4.2×10^{-6}(2)	5.0×10^{-2}(2)	2.0×10^{3}(2)	2.4×10^{7}(2)
2.5 cm layer of soil at 15 cm	2.8×10^{-9}(1)	8.1×10^{-7}(1)	1.4×10^{-2}(1)	1.6×10^{3}(1)	2.8×10^{7}(1)
2.5 cm layer of soil at 30 cm	3.7×10^{-10}(1)	8.5×10^{-8}(1)	3.3×10^{-3}(1)	1.3×10^{3}(1)	5.0×10^{7}(1)
Understory leaves	3.7×10^{-7}(1)	2.6×10^{-6}(1)	----	3.9×10 (1)	----
		Grassy Pasture (Figure 5A)			
Litter-herb	2.5×10^{-8}(2)	3.2×10^{-6}(2)	2.3×10^{-3}(2)	7.2×10^{2}(2)	4.8×10^{5}(2)
Surface 2.5 cm layer of soil	3.6×10^{-9}(2)	4.6×10^{-6}(2)	5.9×10^{-2}(2)	7.2×10^{3}(2)	9.1×10^{7}(2)
2.5 cm layer of soil at 15 cm	$<4.0 \times 10^{-10}$(2)	$<6.2 \times 10^{-7}$(2)	$<2.6 \times 10^{-2}$(2)	4.4×10^{3}(2)	1.5×10^{8}(2)

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TABLE 12. SUMMARY OF ^{129}I and ^{127}I IN VEGETATION AND SOIL IN THE ENVIRONS OF THE NUCLEAR FUEL SERVICE PLANT, WEST VALLEY, NEW YORK, 1973

Sample	Atom Ratio $^{129}I:^{127}I$	pCi ^{129}I Per g	pCi ^{129}I Per m^2	ng ^{127}I Per g	ng ^{127}I Per m^2
Beech-Cottonwood Forest (Figure 6F)					
Litter-herb layer	3.6×10^{-4}(2)	7.6×10^{-2}(2)	4.7×10 (2)	1.4×10^{3}(2)	7.9×10^{5}(2)
Surface 2.5 cm layer of soil	1.8×10^{-5}(2)	6.6×10^{-2}(2)	5.5×10^{2}(2)	2.1×10^{4}(2)	1.8×10^{8}(2)
2.5 cm layer of soil at 15 cm	6.0×10^{-7}(2)	1.2×10^{-3}(2)	2.8×10 (2)	1.4×10^{4}(2)	4.4×10^{8}(2)
Earthworms in upper 15 cm of soil	2.6×10^{-5}(1)	9.0×10^{-2}(1)	----	1.9×10^{4}(1)	----
Viburnum leaves	7.8×10^{-4}(1)	1.3×10^{-2}(1)	----	9.6×10 (1)	----
Apple leaves	5.4×10^{-4}(1)	1.2×10^{-2}(1)	----	1.2×10^{2}(1)	----
Beech-Maple Forest (Figure 6C)					
Litter-herb layer	1.2×10^{-3}(2)	2.3×10^{-1}(2)	1.2×10^{2}(2)	1.2×10^{3}(2)	5.2×10^{5}(2)
Surface 2.5 cm layer of soil	6.8×10^{-5}(2)	5.9×10^{-2}(2)	8.0×10^{2}(2)	5.9×10^{3}(2)	8.5×10^{7}(2)
2.5 cm layer of soil at 15 cm	1.4×10^{-7}(2)	8.8×10^{-4}(2)	1.8×10 (2)	1.1×10^{4}(2)	2.3×10^{8}(2)
2.5 cm layer of soil at 30 cm	2.6×10^{-7}(1)	2.9×10^{-4}(1)	----	6.2×10^{3}(1)	----
Hemlock Forest (Figure 6D)					
Litter-herb layer	3.8×10^{-3}(1)	4.8×10^{-1}(1)	4.0×10^{2}(1)	6.9×10^{2}(1)	5.7×10^{5}(1)
Surface 2.5 cm layer of soil	3.8×10^{-4}(1)	3.2×10^{-1}(1)	1.3×10^{3}(1)	4.7×10^{3}(1)	1.9×10^{7}(1)
2.5 cm layer of soil at 15 cm	2.9×10^{-6}(1)	2.1×10^{-3}(1)	----	4.2×10^{3}(1)	----
Hemlock twigs	2.9×10^{-3}(1)	2.4×10^{-1}(1)	----	4.6×10^{2}	----
Grassy Pasture (Figure 6B)					
Litter-herb layer	5.4×10^{-5}(2)	7.0×10^{-3}(2)	1.6×10 (2)	6.7×10^{2}(2)	3.4×10^{5}(2)
Surface 2.5 cm layer of soil	1.3×10^{-5}(2)	5.6×10^{-3}(2)	1.9×10^{2}(2)	4.4×10^{3}(2)	8.0×10^{7}(2)
2.5 cm layer of soil at 15 cm	5.7×10^{-7}(1)	4.1×10^{-4}(1)	----	4.9×10^{3}(1)	1.3×10^{8}(1)
Miscellaneous Samples					
Hypnum (Figure 6A)	3.0×10^{-5}(1)	6.7×10^{-3}(1)	----	1.2×10^{3}(1)	----
Honey	----	1.9×10^{-6}(1)	----	----	----
Maple syrup	1.7×10^{-6}(1)	3.2×10^{-6}(1)	----	1.1×10 (1)	----
Physcia (Figure 6E)	6.0×10^{-4}(1)	4.3×10^{-1}(1)	----	4.0×10^{3}	----
Platygyrum/Brachythecium (Figure 6E)	5.2×10^{-4}(1)	5.3×10^{-1}(1)	----	5.6×10^{3}	----

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(beech-cottonwood, beech-maple, and hemlock) were sampled and one meadow site was studied. The locations of these sites are designated in Figure 6. Notice that not all sites are the same distance from the stack. Consequently the numbers in the table may in part reflect differences in aerial distribution of stack effluent.

Meadow vegetation had lower ^{129}I concentrations than forest vegetations expressed as pCi/gram or as pCi/m^2; the same is true for the surface layers of soil in the two locations. Deeper layers of soil had less ^{129}I than the surface layer as has also been shown in the previous tables.

The data suggest that ^{129}I is "trapped" by foliage. It reaches the forest floor via falling leaves and is retained in the surface soil for a time before moving downward, possibly through leaching, through translocation within root systems, or both. The highest concentrations of ^{129}I were associated with the hemlock forest. Hemlock, Tsuga canadensis, is an evergreen, needle-leaved tree as tall as the surrounding trees in the deciduous forest. Leaves of hemlock remain attached to their twigs for a year or more. Leaves are numerous and small in size and thus present a great deal of surface area per gram of leaf weight. This is in contrast to the deciduous tree leaves which are broad and flat and persist for only a few months each year (May-October). The leaves of hemlock trees are resinous and probably more resistant to microbial decay than the leaves of deciduous trees and shrubs. The soil beneath these hemlock trees was acidic having a pH of 4.0 in the surface 10 cm.

The meadow had less ^{129}I associated with the vegetation and soil than the forested sites. A grassy sward is exposed to a smaller volume of air than forest stands because of differences in stature. It is also likely that more leafy material is contributed to the soil surface by forests than by grass swards. Thus it is reasonable to expect that forest stands would be more efficient at sorbing airborne materials than adjacent grassy swards. The agricultural history of the grassy pasture is not known with certainty, but the grass was probably planted before the NFS plant became operational.

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The sampling was done on the highway right-of-way where the pasture has probably not had a grazing history and has not been cut for hay. The forest sites have probably changed little since the NFS plant was constructed.

The newly-emerged leaves and twigs of Viburnum and apple, understory shrubs in the area, had ^{129}I concentrations lower than those in the leaf litter collected from the forest floor, but ^{129}I:^{127}I ratios that were greater than those in the leaf litter. It is not yet clear whether the ^{129}I in these young leaves represents material sorbed from the air, remobilized from the tree, or taken up from the soil via the roots. However, the same trend was noted in the samples taken at Morris, Illinois. Only further research will clarify this matter.

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CONCLUDING DISCUSSION

This research program was initiated at least in part as the result of unexpectedly high levels of ^{129}I being found in deer thyroids in the environs of the NFS plant in New York. We have given some thought to trying to explain how deer might have gotten high levels of ^{129}I. Leaving aside the obvious explanations of experimental errors and of the fact that some deer may have browsed or drunk from "forbidden" areas, we have tried to find other alternative explanations. First of all, it is clear that iodine appears to be more persistent in the environment than many of us previously thought. The litter in these samples has been leached by rains and snowmelt in an unusually wet year (for the New York and Illinois samples) and yet the ^{129}I and ^{127}I both remain there in significant quantity. This persistence can help to explain the high levels in deer thyroids. Furthermore, we considered the odd kinds of vegetation which might be a part of the deer diet, especially in the early spring before much plant material has grown. It occurred to us that young tree leaves and buds, lichens and mosses on tree trunks and rocks, and even watercress in streams might be appealing when other food was not available. We also considered sampling conifer nuts (such as pine nuts) since persistent evergreen leaves may accumulate high levels of stack effluents. From the results presented here, it appears that persistent vegetation (Physcia and Platygyrum samples and hemlock twigs in Table 12) may have higher ^{129}I levels than surrounding deciduous leaves and also that emerging leaves in the spring may have unusually high ^{129}I:^{127}I ratios. We have not yet collected samples of evergreen nuts and fruits. We also believe that the forested, multistoried areas where deer often browse may have more ^{129}I than will be found in an open cow pasture. We have also given some thought to the NFS site from a geological or geographical point of view. The stack is in a valley. Iodine trapped by surrounding trees may be washed from higher areas in the valley to lower places and eventually it might be accumulated in the waters of Buttermilk Creek which drains the valley. Since this stream is also used to dispose of liquid effluents,

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it could be that the gaseous iodine effluents and the liquid iodine effluents are both ending up in Buttermilk Creek to some extent. We hope soon to clarify this point by determining the ^{129}I levels in samples of sessile biota collected upstream of the plant. We also have given some consideration to the fact that, in a humid area, iodine would tend to be dropped nearer to the stack than it would (on the average) in an arid area. If there is a relative decrease in plume dispersion in a humid climate, then release criteria developed over the years in an arid zone cannot necessarily be applied without careful reconsideration to releases in a humid climate. We realize that we have not answered the question of why deer thyroids showed high levels of ^{129}I in previous years, but we have presented some ideas and some data giving possible explanations. Other samples will be collected soon to help clarify some of these points further. We are especially curious to obtain some air samples. We initially felt that we did not need to take air samples since the plant has not been in operation for over two years and no gaseous effluents were being emitted. However, some of our results could be explained if the iodine in the air has a high ^{129}I:^{127}I ratio. Only further sampling will clarify this point.

We would like to turn next to the question of whether the present levels of ^{129}I in the environment warrant concern with regard to the dose information developed in the first section of this report.

The data collected to date do not permit direct calculation of human thyroid doses. Samples of items related to human exposure such as air, water, animal feed crops or human foods have not yet been collected. However, data reported here for atom ratios (^{129}I:^{127}I) in the Hanford samples are the same order of magnitude as those previously reported by Brauer, et al.[21] In that study the highest ^{129}I:^{127}I ratios found in items related to direct human intake were 5×10^{-5} in air; 1×10^{-6} in Columbia River water; and 6.7×10^{-7} in milk. Since the dose via inhalation of radioiodine is insignificant compared to the dose via ingestion, a typical limiting ratio in the thyroid can be taken to be 1×10^{-6} atoms of ^{129}I per atom of ^{127}I for Brauer's data. From the parameters listed

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in Table 7, it can be seen that such a ratio implies a dose of about 5 mrem/yr to the adult thyroid and 1 mrem/yr to the infant.

However, if actual ^{129}I activities in the samples reported by Brauer rather than atom ratios are used to calculate the doses, results are significantly lower. For example, the thyroid dose to an adult with maximized dietary habits was ∿0.4 mrem/yr, while the dose to a more typical adult was about one-third of that. The doses to the maximum and average infants were estimated to be 0.3 and 0.1 mrem/yr, respectively.

The highest atom ratios (10^{-3}) found in the vicinity of the NFS plant in the current study are also similar to those previously reported[27,31] for that site, although the types of samples collected were different in the two studies. An unexpected finding was that samples collected from the litter-herb layer at Hanford and at the grassy pasture site at NFS had similar atom ratios, (3 to 5) x 10^{-5}.

Milk samples from the vicinity of the NFS plant have been collected and analyzed by New York State personnel since September 1971.[22] Although a peak concentration of about 2 pCi ^{129}I per liter of milk was detected in March 1972, concentrations fell to less than 0.3 pCi/liter in May 1972 and to less than 0.01 pCi/liter in March 1973. Again, these values are not significantly higher than those reported for two samples collected in the vicinity of Hanford[21] (0.02 in 1967 and 0.08 in 1971), considering the fact that the vast majority of the results of the NFS samples were below the detection limit (typically 0.3 to 0.5 pCi/liter milk). On this basis one would not expect the average thyroid doses for the past year at the NFS site to be significantly different than those estimated above for the Hanford site.

The atom ratio in the litter-herb layer collected from the grassy pasture in the environs of the MFRP at Morris, Illinois, was about 0.1% of those measured at the Hanford and NFS sites so that thyroid doses to residents around Morris, Illinois, would be expected to be about 0.1% of those given above for the other two sites.

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None of these estimated doses are significant in comparison to the existing radiation standard of 1500 mrem/yr to the thyroid or in comparison to the annual dose of about 100 mrem from natural background radiation.

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