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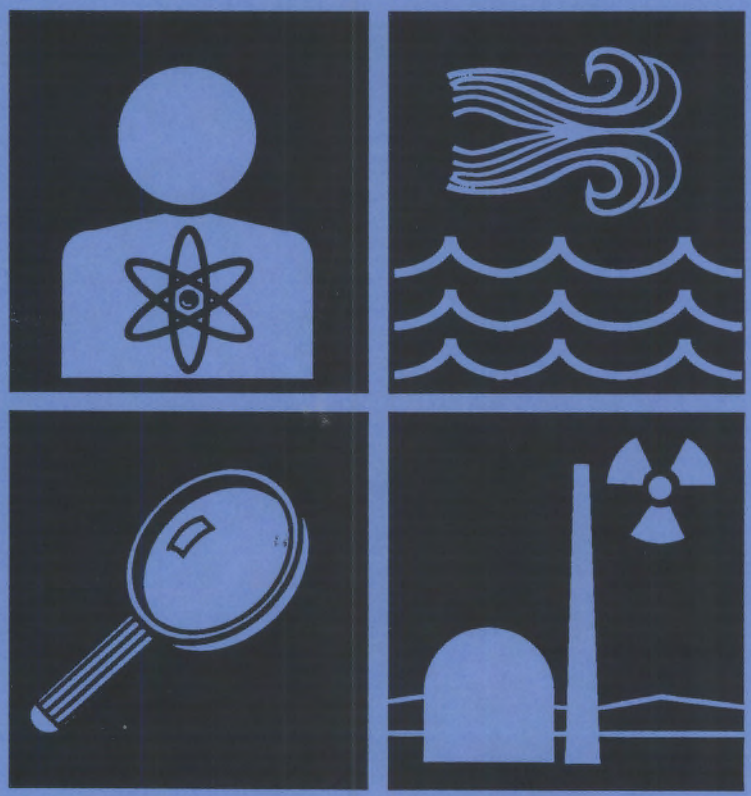
Feasibility of Using ^{129}I Concentrations in Human Tissue to Estimate Radiation Dose from ^{131}I

Hanford Environmental Dose Reconstruction Project

October 1989



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NTIS Price Codes
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Hanford Environmental Dose Reconstruction Project

Feasibility of Using ^{129}I Concentrations in Human Tissue to Estimate Radiation Dose from ^{131}I

May 1989

This document has been reviewed by the Technical Steering Panel and is approved for public release.

J. E. Till, Chair
Technical Steering Panel

August 17, 1989
Date

Battelle
Pacific Northwest Laboratories
Battelle Boulevard
P.O. Box 999
Richland, Washington 99352
Telephone (509)

February 15, 1990

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Please make the following change in your copy of the subject report:

Page 6, line 4: change 1974 to 1972

Sincerely,

B.A. Napier

Research Scientist
Hanford Environmental
Dose Reconstruction Project

PNL-6889 HEDR
UC-707

FEASIBILITY OF USING ^{129}I CONCENTRATIONS IN HUMAN TISSUE TO ESTIMATE RADIATION DOSE FROM ^{131}I

W. D. McCormack

October 1989

Prepared for
the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Richland, Washington 99352

PREFACE

This report describes the progress of a study conducted in support of the Hanford Environmental Dose Reconstruction (HEDR) Project. The objective of the HEDR Project is to develop estimates of the potential radiation doses that people may have received from past Hanford operations. The objective of this supporting study is to evaluate the feasibility of using data on ^{129}I in human tissue to estimate historical exposure of individuals to past releases of ^{131}I from the Hanford Site.

Although this report discusses the results of investigations conducted to date, additional information is required before the study can be completed and final recommendations developed. Specifically, further analyses are under way regarding the potential use of thyroid samples collected during the late 1940s and early 1950s. Evaluations of such data are included in the efforts of Or. D. E. Wrenn, University of Utah, to reconstruct the exposure of individuals to radioiodine released during nuclear weapons testing at the Nevada Test Site. However, the report of Dr. Wrenn's efforts is not scheduled to become available until later in 1989. While information obtained in direct communications with Dr. Wrenn has tended to reinforce the conclusions of this study, the report of his work will be reviewed in detail when it becomes available, and an attachment to this report will be prepared which summarizes that review.

SUMMARY

To use data on ^{129}I in human tissue to estimate an individual's past exposure to that radionuclide, it is necessary to know when and how the exposure occurred, and to know about any other prior and/or ongoing exposures. Moreover, to use ^{129}I data to estimate past exposure to ^{131}I, it is also necessary to know the relationship of the two radionuclides at the time of exposure. The relative quantities of ^{131}I and ^{129}I in gaseous effluents from Hanford Site facilities varied significantly because of the large variations in elapsed time between discharge of irradiated fuel from Hanford production reactors and initiation of chemical processing. The relationship of the two radionuclides also varied in the environment because the shorter-lived ^{131}I decayed and the longer-lived ^{129}I accumulated. Because of its extremely long half-life, ^{129}I from both Hanford and non-Hanford sources (such as fallout from weapons testing) has accumulated in the environment. Without an associated exposure to ^{131}I, chronic exposure to ^{129}I deposited in the environment has contributed and continues to contribute to thyroid burdens.

Based on investigations conducted to date, measured levels of ^{129}I in human tissue will not provide a viable alternative for reconstruction of historical exposure to ^{131}I. Final recommendations on the possible use of ^{129}I data will not be developed until receipt of results of the efforts of Dr. D. E. Wrenn, University of Utah, to reconstruct the exposure of individuals to radioiodine released during nuclear weapons testing at the Nevada Test Site.

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INTRODUCTION

Past operations on the Hanford Site have resulted in the release of much greater quantities of radionuclides to the environment than operations during more recent years. Radioactive materials were released in the gaseous and liquid effluents from operation of the production reactors and during chemical processing of the irradiated reactor fuels. In 1986, the Hanford Health Effects Review Panel recommended that a study be performed to reconstruct the radiation doses that people may have received from radioactive releases since Hanford operations began in 1944.

The Hanford Environmental Dose Reconstruction (HEDR) Project was initiated to develop these estimates. The project involves, in part, the review of historical information to identify relevant and useful environmental monitoring data, development of estimated quantities of radionuclides that may have been discharged in the gaseous and liquid effluents, and reconstruction of local meteorological and other pertinent environmental conditions, including the dietary and living habits of the people living near Hanford.

The radiation dose that a person receives is the sum of doses received from exposure to external and internal sources of radiation. The dose that a person receives from external sources is a function of the dose rate and the duration of the exposure. The dose from internally deposited radionuclides is a function of the quantity and length of time that radionuclides remained in the individual's body. The amount of a particular radionuclide in the body at any given time is dependent on a number of parameters--the time, rate, and mode in which it entered the body (e.g., via inhalation, ingestion, absorption, or punctures through the skin), the chemical behavior of the radioactive material in the body (which determines the amount initially taken up and deposited in the various tissues of the body, rates of transfer between organs, and elimination from the body), and its physical half-life.

Estimates of radiation doses potentially received by the general public from exposure to radioactive materials from nuclear facilities can be developed from direct measurements of radionuclides in environment. Such measurements can be used to develop intake rates of the radionuclides and/or

external exposure rates. However, most estimates are typically based on measurements or projections of radionuclides in a nuclear facility's effluents and on environmental pathway models. The models are used to predict the movement of the radionuclides through the environment and to predict the levels of radionuclides in the air, water, soil, and foodstuffs. Metabolic and dosimetric models are then used to estimate the uptake, deposition, and elimination of the radionuclides in the human body and the resulting potential radiation dose.

Under certain circumstances, measured levels of radionuclides in the human body can also be used to estimate potential doses. The radiation dose estimates can be based on direct measurements of radioactive materials in the body using sensitive radiation detectors (i.e., in vivo measurements) or indirect measurements based on laboratory analyses of excreta, body fluids, or other tissue samples (i.e., in vitro measurements). These measurements, along with knowledge of when and how the exposure occurred and how the radionuclides behave in the body, can be used to develop estimates of potential dose.

Iodine-131 (^{131}I) was one of the radionuclides released to the environment in the largest quantities during the early years of Hanford operations and may have been a major contributor to the potential radiation dose received by people living near the Site. In situations in which all of the information and parameters relevant to the estimation of radiation doses are known, the dose received by an individual can be determined fairly accurately. However, adequate information is not currently available to the HEDR Project for many of these parameters. Because radiation detection equipment was in its infancy during those years, few measurements were made on the levels of ^{131}I in effluents, foodstuffs, or people. In addition, ^{131}I does not remain in the environment for very long because of its relatively short 8-day physical half-life. As a result, very few data are available on the measured levels of ^{131}I in environmental media or in people during the early years of Hanford operations.

Other radioactive isotopes of iodine are also produced within reactor fuel during the fission process. However, iodine-129 (^{129}I), which has a physical half-life of 1.7×10^7 years (Table 1), is the only other isotope of

TABLE 1. ^{129}I and ^{131}I Radiological, Biological, and Effective Half-Lives (in days)

	Radiological	Biological	Effective
^{131}I	8.0	120	7.5
^{129}I	5.8×10^9	120	120

iodine that has a physical half-life long enough to be present in the fuel, effluents, or environment for any length of time. Sample archives from the early years of Hanford operations may be available and helpful in determining the levels of general population exposure to ^{129}I which, in turn, may be useful in estimating past exposures to ^{131}I. Most notable is the possibility of using thyroids collected from people who lived in the area during the early Hanford years. No ^{131}I would be present; however, ^{129}I might be because of its long physical half-life. This report presents the results of an evaluation of the feasibility of using data on ^{129}I to reconstruct the potential radiation doses that people may have received from the ^{131}I that was released to the environment from Hanford operations since 1944.

BACKGROUND

A summary of the quantities of ^{129}I and ^{131}I contained in Hanford gaseous effluents for the years 1944 through 1972 is provided in Table 2 (Price et al. 1981, Anderson 1974). The quantity of iodine in the gaseous effluents varied significantly from year to year. The quantity of ^{131}I in gaseous and liquid effluent is directly related to the elapsed time between fuel discharge from the reactor and the initiation of chemical separations. The extremely long half-life of ^{129}I means it is essentially not subject to removal from the irradiated fuel, effluents, or environment by radioactive decay.

Particularly important to this discussion is the wide range of values for the ratio of ^{131}I to ^{129}I: approximately 4 orders of magnitude during the period 1944 to 1972. Variations in the ratio of ^{131}I to ^{129}I are largely the result of variable cooling times between the discharge of irradiated fuel from the reactors and initiation of chemical processing.

An evaluation of ^{129}I and nonradioactive ^{127}I in the tissue of pigs raised in the vicinity of the Hanford Site was conducted by J. K. Soldat (1974). Tissue samples from pigs as well as samples of feed, ground cover, and soil beneath the ground cover were collected and analyzed for ^{129}I and ^{127}I. The results of analyses on pig tissue revealed different ratios of ^{129}I to ^{127}I in different tissues. An explanation suggested by Soldat was that ratios of ^{129}I to ^{127}I in the environment varied with time and that sampled pigs were exposed to environmental iodine via ingestion of soil. An additional factor that may have caused non-uniform organ uptake of iodine was the high content of ^{127}I in the pig's feed. This suggested the possibility that high intake of ^{127}I affected the further intake of iodine from other sources.

An evaluation of the levels of ^{129}I in Pacific Northwest forage and deer tissue was conducted by Price et al. (1981) "demonstrated the longevity of ^{129}I in the biosphere following gaseous release from a nuclear facility." The average ^{129}I concentrations in Hanford deer thyroid samples collected in 1978 were nearly the same as values reported previously by Brauer et al. (1973) for samples collected from the site in 1964. Although other

TABLE 2. Estimated Radioiodine Releases to the Atmosphere with Gaseous Effluents at Hanford, 1944 Through 1972

Year	^{129}I (Ci)(a)	^{131}I (Ci)(b)	Ratio $^{131}I/^{129}I$
1944	1.0×10^{-5}	1.7×10^{3}	1.7×10^{8}
1945	1.0×10^{-1}	3.4×10^{5}	3.3×10^{6}
1946	1.6×10^{-1}	7.6×10^{4}	4.9×10^{5}
1947	1.1×10^{-1}	2.4×10^{4}	2.2×10^{5}
1948	6.5×10^{-2}	1.2×10^{3}	1.9×10^{4}
1949	8.0×10^{-5}	4.6×10^{3}	5.8×10^{10}
1950	1.5×10^{-4}	2.1×10^{3}	1.4×10^{7}
1951	2.1×10^{-4}	1.9×10^{4}	8.9×10^{7}
1952	2.9×10^{-4}	9.7×10^{2}	3.3×10^{6}
1953	3.9×10^{-4}	7.3×10^{2}	1.9×10^{6}
1954	5.0×10^{-4}	5.4×10^{2}	1.1×10^{6}
1955	5.7×10^{-4}	1.2×10^{3}	2.1×10^{6}
1956	9.1×10^{-4}	3.7×10^{2}	4.0×10^{5}
1957	1.4×10^{-3}	3.7×10^{2}	2.7×10^{5}
1958	1.4×10^{-3}	4.3×10^{2}	3.0×10^{5}
1959	1.8×10^{-3}	2.9×10^{2}	1.6×10^{5}
1960	1.9×10^{-3}	3.5×10^{2}	1.8×10^{5}
1961	2.1×10^{-3}	2.6×10^{2}	1.2×10^{5}
1962	2.0×10^{-3}	1.2×10^{2}	6.0×10^{4}
1963	1.8×10^{-3}	1.4×10^{2}	7.6×10^{4}
1964	2.0×10^{-3}	7.9×10^{1}	3.9×10^{4}
1965	2.1×10^{-3}	9.9×10^{1}	4.8×10^{4}
1966	1.6×10^{-3}	7.1×10^{1}	4.6×10^{4}
1967	1.3×10^{-3}	3.0×10^{1}	2.3×10^{4}
1968	1.7×10^{-3}	5.6×10^{0}	3.5×10^{3}
1969	1.3×10^{-3}	1.6×10^{0}	1.2×10^{3}
1970	6.0×10^{-4}	5.0×10^{-1}	8.2×10^{2}
1971	1.1×10^{-3}	2.0×10^{-1}	1.9×10^{2}
1972	4.5×10^{-4}	2.1×10^{-1}	4.7×10^{2}

(a) Price 1981.
(b) Anderson 1974.

explanations for this nearly constant level of ^{129}I in deer thyroid may exist, the authors suggested the possibility that the environmental availability of ^{129}I to deer did not change during the intervening 14 years despite the cessation of chemical processing activities in ~~1974~~ 1972.

The several different pathways by which individuals can be exposed to radioiodine released to the environment are illustrated in Figure 1, the most significant of which are ingestion of milk and vegetables. A review of those pathways (Soldat 1976) indicated that for short-term exposure situations, the intake of ^{129}I via the soil-root pathway is insignificant relative to intakes via foliar deposition. However, when releases of ^{129}I to the environment continue over a period of several years, the contribution of the soil-root pathway to foodstuff concentrations becomes increasingly significant. For example, iodine deposited on the soil and retained in the root zone of the plant contributes about 1% (via the soil-root pathway) of the ^{129}I in leafy vegetables. After 10 years of ongoing (constant) release to the environment and deposition on the soil, the soil-root pathway would be expected to contribute approximately 10% of the leafy vegetable iodine concentrations. For other vegetables, where only 10% of the aerially deposited material is expected to reach the edible portions of the plant, the soil pathway contributes approximately 10% to the plant's concentration (per year of release). The soil pathway contributes about 1.3% (per year of release) of the ^{129}I present in milk from aerial deposition on fresh forage.

The environmental dose calculation procedure indicated in Figure 2 demands extensive input data and often requires that assumptions be made at each stage of the calculations. Valid calculational results require accurate characterization of the exposure environment. It is necessary that only the pathways applicable to a given exposure situation be identified, described, and included in the calculations. For example, consumption of cow's milk is a primary pathway for iodine intake. Calculations based on consumption of milk produced locally would produce significantly different estimates of iodine intake relative to calculations based on milk obtained from outside the area. The season of the year during which iodine is released to the environment will affect the availability of certain exposure pathways. For example, most agricultural pathways are active during the spring, summer, and autumn seasons but are reduced or inactive during the winter.

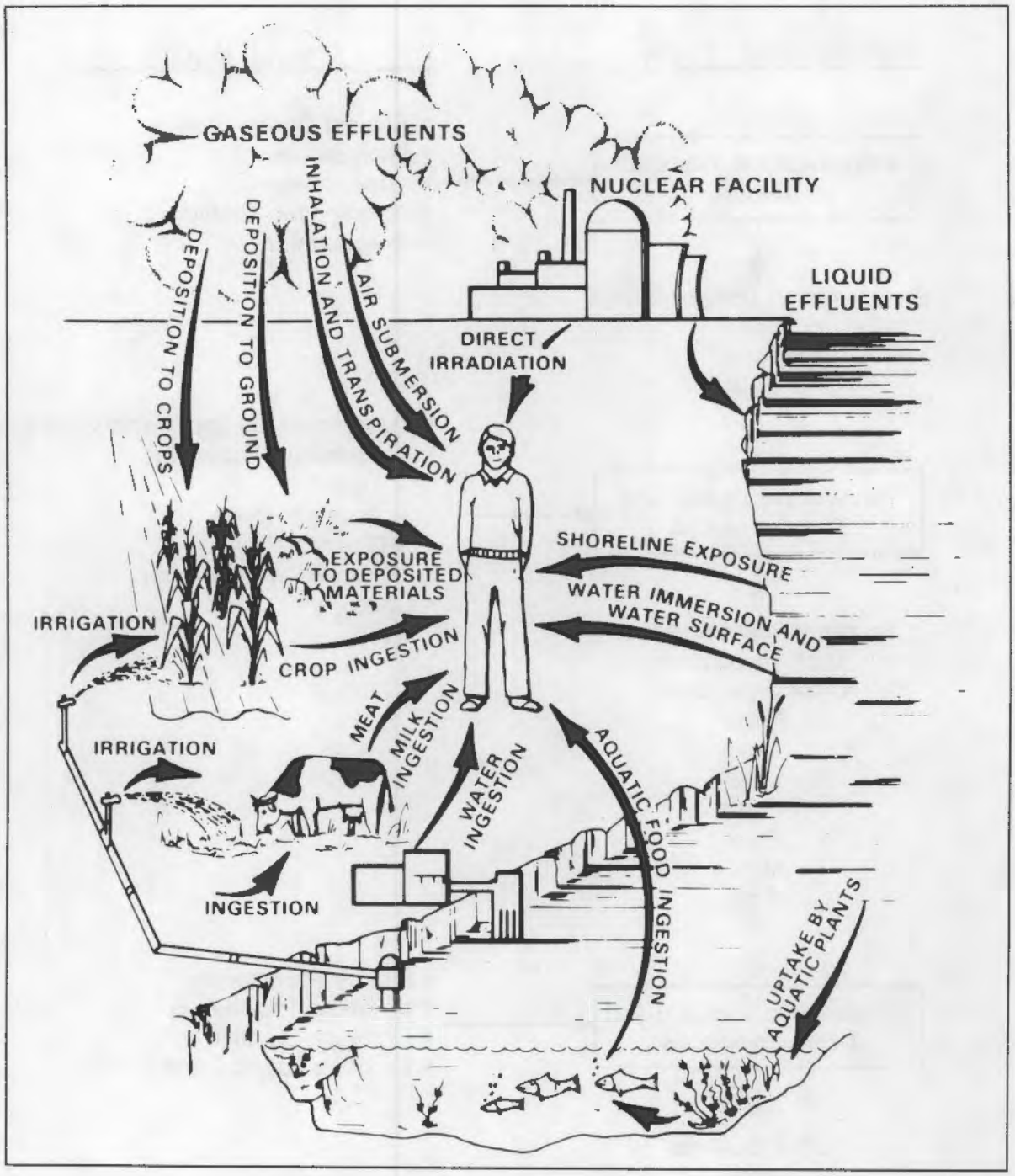

FIGURE 1. Potential Radiological Dose Pathways

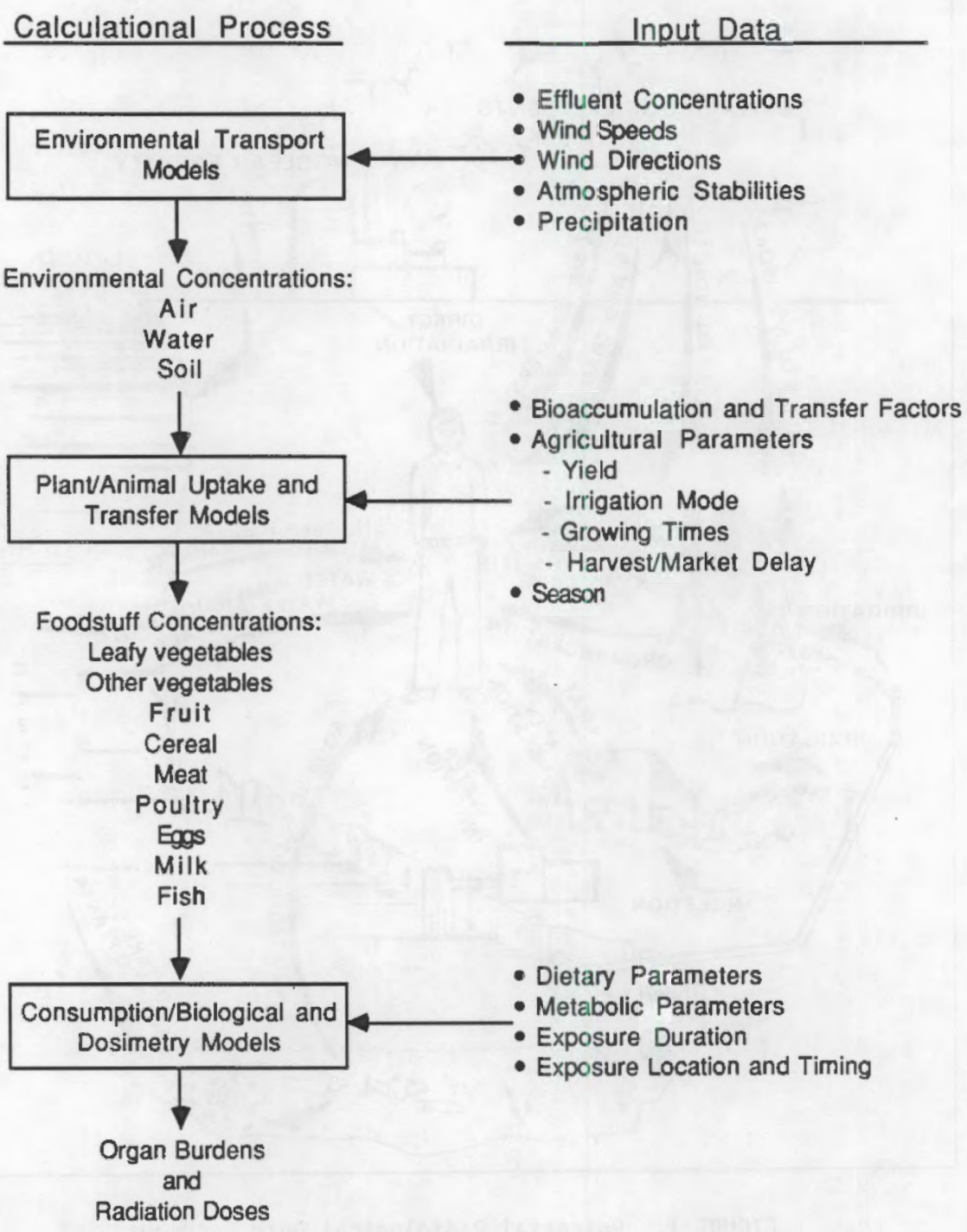

FIGURE 2. Environmental Dose Calculation Process

EVALUATION

Calculations can be performed, under certain circumstances, to estimate an individual's exposure to a radionuclide based on measurements of that radionuclide in organ tissue following exposure. That type of calculation is accomplished using many of the same models, data, and assumptions used in the effluent-based calculations. However, the concentration of a radionuclide in organ tissue changes with time during and following an uptake. As a result, the process of estimating previous intake based on measurements of subsequent radionuclide concentration in tissue requires additional data concerning the timing, nature, and duration of exposure, as well as any other prior and/or ongoing exposures. To use measurements of one radionuclide to predict potential past exposure to another, it is also necessary that the relationship of the two radionuclides at the time of exposure be known.

Two significant issues impact the potential use of ^{129}I measurement data for reconstructing exposure to historic ^{131}I releases:

1. The ratio of ^{131}I to ^{129}I is highly variable as a result of the extreme differences in their half-lives. The ratio of ^{131}I to ^{129}I in the total annual gaseous effluent varies over approximately 4 orders of magnitude during the years 1944 through 1972 (Table 2). Additional fluctuations undoubtedly occurred within each year. The ratio of ^{131}I to ^{129}I will also vary among geographic locations and media in the environment as ^{131}I continues to decay during both environmental transport and the interval between deposition and human exposure.

2. Levels of ^{129}I in the environment are an accumulation of material released to the environment over a relatively long period of time. Because of its relatively short half-life, ^{131}I levels found in the environment are associated with relatively recent releases. Further, following a short-term uptake of ^{131}I in the thyroid, ^{131}I will be removed with an effective half-time of 7.5 days, while the effective half-time of ^{129}I will be 120 days. Therefore, concentrations of ^{129}I observed in human thyroid tissue may be the result of an intake that occurred at any time during the preceding 2 or 3 years, while ^{131}I will be completely removed from the thyroid within 2 or 3 months following an uptake.

As illustrated in Table 2, the relationship of ^{131}I to ^{129}I in Hanford gaseous effluents was highly variable. The relationship of ^{131}I to ^{129}I varies further between the several environmental pathways that may lead to consumption of contaminated foodstuff. Pathways with relatively less time

delay between radioiodine emission to the environment and the consumption of foodstuff (e.g., leafy vegetables and cow's milk) will result in relatively less decay of ^{131}I than pathways with longer time delays (fruit, cereals, potatoes, etc.). The much longer-lived ^{129}I will not be affected by variable pathway time delays. As a result, the ratio of ^{131}I to ^{129}I consumed in cow's milk and leafy vegetables, for example, will be higher than that found in radioiodine associated with consumption of cereal and fruit.

In addition to variations between pathways, the ratio of ^{131}I to ^{129}I in an individual's total radioiodine intake will vary between seasons of the year. During summer, pathways with less time delay will dominate the total intake of radioiodine. As a result, the relative ^{131}I content will be higher than during winter when local fresh vegetables are no longer consumed and cows are on stored feed. Table 3 illustrates the variability of $^{131}I/^{129}I$ ratios in radioiodine intake as a function of pathway and season of release.

Because of its extremely long half-life, ^{129}I from both Hanford and non-Hanford sources (such as fallout from weapons testing) has accumulated in the environment. Chronic exposure to ^{129}I previously deposited in the environment contributes to thyroid burdens of this radionuclide without an associated exposure to ^{131}I. Table 4 illustrates the effect of prior depositions and environmental accumulation of ^{129}I on the predicted total intake of ^{131}I and ^{129}I. The iodine intakes shown in Table 4 are calculated from inhalation and from ingestion of terrestrial foodstuffs and animal products, and are based on the release of 0.035 Ci of ^{129}I and 39,000 Ci of ^{131}I per year for 12 years. That annual release is equivalent to the average of the annual releases that occurred between 1944 and 1955 (Table 2).

The predicted intake of ^{129}I from exposure to the twelfth year's effluents and from exposure to radioiodine deposited in the environment by the prior 11 years' effluents (Table 4, column 2) is approximately twice the intake estimated without accounting for prior deposits. However, the predicted intake of ^{131}I remains unchanged. The predicted intake of ^{129}I, after accounting for ongoing exposure (50 years) to the 12 years of environmental accumulations of ^{129}I (Table 4, column 3), is approximately 40 times higher than the intake estimated on the basis of a single year's release and exposure.

TABLE 3. Ratios of ^{131}I to ^{129}I in Calculated Radioiodine Intakes as a Function of Pathway and Season of Release[a]

Pathway	Winter	Spring	Summer	Autumn
Inhalation	31	31	31	31
Leaf Veg	<0.001	320	320	28
Other Veg	<0.001	0.01	1.5	20
Fruit	<0.001	0.01	1.5	20
Cereals	<0.001	<0.001	<0.001	<0.001
Meat	<0.001	94	96	9
Poultry	<0.001	0.01	2	29
Cow Milk	<0.001	320	330	29
Eggs	<0.001	0.01	2	27
All Pathways	0.2	28	130	25

(a) Based on a hypothetical release of 1000 Ci of ^{131}I and 1 Ci of ^{129}I (ratio of ^{131}I to ^{129}I in effluent = 1000

TABLE 4. Estimated Intakes of Radioiodine (in μCi) from Various Release and Environmental Accumulation Scenarios

Radionuclide	1-yr Release, 1-yr Exposure[a]	1-yr Release, 12-yr Deposition, 1-yr Exposure[b]	1-yr Release, 12-yr Deposition, 50-yr Exposure[c]
^{129}I	2.96×10^{-5}	5.11×10^{-5}	1.15×10^{-3}
^{131}I	1.14×10^{1}	1.14×10^{1}	1.14×10^{1}
Ratio $^{131}I/^{129}I$	3.86×10^{5}	2.23×10^{5}	9.90×10^{3}

(a) Calculation based on release of 3.5×10^{-2} Ci of ^{129}I and 3.9×10^{4} Ci of ^{131}I, with exposures only during the year of release.
(b) Calculation based on a total of 12 years of releases at 3.5×10^{-2} Ci of ^{129}I and 3.9×10^{4} Ci of ^{131}I per year, with exposures only during the 12th year.
(c) Calculation based on a total of 12 years of releases at 3.5×10^{-2} Ci of ^{129}I and 3.9×10^{4} Ci of ^{131}I per year, and exposures during the 12th year and subsequent 49 years.

RECOMMENDATIONS

Based on investigations conducted to date, measured levels of ^{129}I in human thyroid or other tissue cannot be used for reconstruction of historical exposure to ^{131}I. To use ^{129}I for this purpose would require knowledge of the following factors specific to past Hanford Site releases:

- the ^{131}I to ^{129}I ratios at the time of emission
- the environmental transport and pathway delays and their effect on the ratio of ^{131}I to ^{129}I prior to exposure and consumption
- the pathways that are actually associated with an individual's exposure
- the chronic intake of ^{129}I that has accumulated in the environment.

Only cursory knowledge currently exists for most of these items; furthermore, the necessary additional information is not likely to be acquired.

Dr. D. E. Wrenn, University of Utah, is using the above factors, specific to past Nevada Test Site releases of radioiodine, to attempt to reconstruct doses for people exposed to radioiodine released during nuclear weapons testing. A final recommendation on whether ^{129}I data can be used to reconstruct potential radiation doses from ^{131}I will be made upon receipt of results of efforts by Dr. Wrenn.

QUALITY ASSURANCE

The work described in this report was performed in accordance with the requirements of ANSI/ASME NQA-1 1986 Edition, Quality Assurance Program Requirements for Nuclear Facilities, as interpreted by PNL's QA program.

Drafts of this document underwent internal independent technical review. Comments from the reviewers were satisfactorily resolved, and there were no controversial resolutions to the comments.

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