

**Conversion and Correction Factors for
Historical Measurements of Iodine-131
in Hanford-Area Vegetation 1948 - 1951**

Hanford Environmental Dose Reconstruction Project

D. H. Denham
E. I. Mart
M. E. Thiede

September 1993

Letter report prepared for
the Technical Steering Panel and
the Centers for Disease Control and Prevention
under Contract 200-92-0503(CDC)/18620(BNW)

Battelle
Pacific Northwest Laboratories
Richland, Washington 99352

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

ds

Preface

In 1987, the U.S. Department of Energy (DOE) directed the Pacific Northwest Laboratory, which is operated by Battelle Memorial Institute, to conduct the Hanford Environmental Dose Reconstruction (HEDR) Project. The DOE directive to begin project work followed a 1986 recommendation by the Hanford Health Effects Review Panel (HHERP). The HHERP was formed to consider the potential health implications of past releases of radioactive materials from the Hanford Site near Richland, Washington.

Members of a Technical Steering Panel (TSP) were selected to direct the HEDR Project work. The TSP consists of experts in the various technical fields relevant to HEDR Project work and representatives from the states of Washington, Oregon, and Idaho; Native American Tribes; and the public. The technical members on the panel were selected by the vice presidents for research at major universities in Washington and Oregon. The state representatives were selected by the respective state governments. The Native American tribes and public representatives were selected by the other panel members.

A December 1990 Memorandum of Understanding between the Secretaries of the DOE and the U.S. Department of Health and Human Services (DHHS) transferred responsibility for managing the DOE's dose reconstruction and exposure assessment studies to the DHHS. This transfer resulted in the current contract between Battelle, Pacific Northwest Laboratories (BNW) and the Centers for Disease Control and Prevention (CDC), an agency of the DHHS.

The purpose of the HEDR Project is to estimate the radiation dose that individuals could have received as a result of radionuclide emissions since 1944 from DOE's Hanford Site. A major objective of the HEDR Project is to estimate doses to the thyroid of individuals who were exposed to iodine-131. A principal pathway for many of these individuals was milk from cows that ate vegetation contaminated by iodine-131 released into the air from Hanford facilities.

The HEDR Project work is conducted under several technical and administrative tasks, among which is the Environmental Monitoring Data Task. Members of the Environmental Monitoring Data Task have developed databases of historical environmental measurements. These databases include iodine-131 concentrations for vegetation samples collected on and around the Hanford Site since 1945, the first full year in which the chemical separation plants were operated and whose effluents led to the release or potential release of radioactive iodine.

A major objective of the Environmental Monitoring Data Task is to provide conversion and correction factors to convert historical measurement data into concentration values that would be determined using current knowledge and equipment. Denham et al. (1993) provided the vegetation data for 1945-1947. Mart et al. (1993) determined the conversion and correction factors for 1945-1947. The vegetation data for 1948-1951 is published in Hanf et al. (1993), which is available as a printed copy in the DOE Richland Operations Office Public Reading Room in Richland, Washington, and as a diskette from the TSP. This report provides the vegetation data conversion and correction factors for 1948-1951, the period when iodine-131 analysis methods changed. These conversion and correction factors will be used as required to adjust original beta counting data and

historically reported radionuclide activity levels in selected vegetation samples to assist in the HEDR model validation studies. With the public availability of the 1948-1951 data (Hanf et al. 1993) and the conversion and correction factors in this report, the HEDR Project Milestone 0502B has been fulfilled.

Summary

Introduction

This report is a part of the Hanford Environmental Dose Reconstruction (HEDR) Project, whose goal is to estimate the radiation dose that individuals could have received from emissions since 1944 at the U.S. Department of Energy's (DOE) Hanford Site near Richland, Washington. The key radionuclide emitted that would affect the radiation dose was iodine-131 (Napier 1992). Because the early methods of measuring iodine-131 were not comparable to later techniques, conversion and correction factors are needed to convert the historical measurement data into concentration values that would be determined using today's knowledge and technologies. This report describes the conversion and correction factors developed for reconstructing historical measurements of iodine-131 in Hanford-area vegetation, which was collected from 1948 through the end of December 1951.

Scope of Work

The scope of work is to quantify the appropriate conversion and correction factors that are necessary to reconstruct true iodine-131 activity levels in vegetation for 1948-1951. In the early operations at the Hanford Site (1945-1947), the method of iodine-131 analysis was to measure gross beta in relatively thick vegetation pellets. By the end of 1948, the method of iodine-131 analysis changed to measuring gross beta in relatively thin precipitates. The precipitates were created by chemically extracting iodine-131 from 5-gram vegetation aliquots as opposed to the 1-gram samples used in the 1945-1947 analyses. Beginning in mid-1951, all parameters had been determined and were being applied to convert net counting rates to activity (Wolff 1951) which provided accuracy comparable to that of today. Hence, no other conversion or correction factors were developed for vegetation data after 1951.

Because two distinctly different methods of iodine-131 analysis on vegetation were used in the early years, the conversion and correction factors for each method are being presented in two separate reports. Mart et al. (1993) provides the conversion and correction factors for the 1945-1947 method. This report provides the conversion and correction factors for the 1948-1951 method. The vegetation data for 1945-1947 are published in Denham et al. (1993). The vegetation data for 1948-1951 are available in Hanf et al. (1993) either as a printed copy in the DOE Richland Operations Office Public Reading Room or as a diskette from the Technical Steering Panel.

Technical Approach

To determine the conversion and correction factors for iodine-131 concentrations in vegetation, Battelle, Pacific Northwest Laboratories (BNW) staff reviewed the historical records and interviewed veteran Hanford employees familiar with the historical vegetation sampling and measurement techniques. The objective was to learn as much as possible about the historical collection and measurement techniques, including radiation-detector systems, sample preparation and counting

techniques, as well as how historical correction factors were developed and applied. This approach allowed BNW staff to evaluate the historical data and correction factors used and to determine their appropriateness for estimating reconstructed iodine-131 concentrations in vegetation.

Conclusions

To convert historical counting data (reported as counts per minute, cpm) to the best estimate of activity in microcuries, four conversion/correction factors are necessary, three of which were previously discussed by Mart et al. (1993): measurement, M ; decay, D_e ; and the iodine-131 assumption factor, I_{cf} . These are included in this report for continuity, and their relative values are compared with those shown in the previous report. The overall detection efficiency, as indicated by the reconstructed measurement conversion factor (M), increased approximately an order of magnitude between the 1945-1947 pellet analysis technique and the 1948-1951 precipitate analysis technique. A new conversion factor, yield (Y), is discussed in greater detail in Section 3.0 of this report. A yield of 63 percent for a yield conversion factor of 1.59 is recommended to best represent the actual yield for vegetation analyzed from December 1948 through 1951. Iodine contributions from weapons test fallout were considered but were found to be insignificant for this time period. These contributions had not been of concern until after the first major atmospheric nuclear weapons test by the Soviet Union in August 1949. (See Section 7.0 for a brief discussion of the iodine contributions from weapons test fallout.)

The conversion and correction factors provided in the 1945-1947 report by Mart et al. (1993) are recommended for the 1948 vegetation pellet counting data through November, the last month in which both pellet and iodine extraction samples were analyzed. One exception to this may be the need to evaluate the iodine-131 assumption correction factor, I_{cf} , on a case-by-case basis because it may have been influenced as early as March 1948 by the addition of filtration systems to the exhaust stacks. This addition could have resulted in less iodine being released compared to other beta emitters and, hence, a value less than 1.0 for I_{cf} .

The reconstructed total conversion factors are $0.0044 \times D_e$ for 1949 and $0.0061 \times D_e$ for 1950-1951. These factors are recommended for the December 1948 through December 1951 iodine-extraction analyses and should be used to interpret, where necessary, the 1948-1951 vegetation data for use in model validation.

Data Quality Objectives

The information generated for this report was prepared before the implementation of formal data quality objectives (Shipler 1993). However, data quality was assessed in the manner described below.

Accuracy and Precision

The results of the research presented in this report are conversion and correction factors with which iodine-131 levels in vegetation can be reconstructed using historical data. The conversion and correction factors presented in this report were determined by comparing the assumptions used in deriving the 1948-1951 data to the processes and procedures that are standard today (EML 1992). The accuracy and precision of these factors should be judged by the uncertainties reported by Gilbert et al. (1992). Because of the improved method of analyzing iodine-131 that was developed and initially implemented in December 1948, the uncertainties reported by Gilbert et al. (1992) for the 1945-1947 conversion and correction factors should decrease for the 1948-1951 period. Therefore, the degree of uncertainty of the reconstructed iodine-131 levels that would be obtained using the conversion and correction factors reported here will not specifically be addressed because the improved method of analysis should drastically reduce the uncertainty associated with the individual analyses.

Completeness

Original Hanford-Site documents on environmental analysis techniques were reviewed as well as documents from non-Hanford-Site sources. The non-Hanford documents confirmed that Hanford had historically (starting in the 1950s) applied appropriate analysis techniques (Baltakmens 1977; Bayhurst and Prestwood 1959; Burt 1949; Gleason et al. 1951; Seliger and Schwebel 1954). Enough information has been uncovered in the documents reviewed and from discussions with veteran Hanford employees to adequately understand the detector systems used and the change in measurement techniques from 1-gram sagebrush pellets to chemical extraction of iodine-131 from 5-gram vegetation aliquots. Hence, any further review of historical documents is not expected to have a major impact on the results of this report.

Representativeness

The factors covered in this report are representative of those reported by Schwendiman (1954) and Wolff (1951) for converting and correcting the historical data.

Comparability

Comparability is not an issue because no previous values other than those historically reported are available for comparison. This report provides a technique of estimating actual values, even if they differ from those historically reported by more than a factor of two.

Conclusions

The uncertainty of the factors (Gilbert et al. 1992) provided in this report and in Mart et al. (1993) could be reduced by additional research. However, additional reviews of documents and notebooks, empirical studies, or mathematical modeling are not expected to have a major impact on the conversion and correction values reported here nor on the final dose estimates of the HEDR Project.

Acknowledgments

BNW staff held three meetings with veteran Hanford employees^(a) (Denham et al. 1988) who provided valuable information by putting in context the methods and intent of the radiation measurements on vegetation during the period of this report. These meetings with the veteran Hanford employees greatly increased the understanding of how to interpret some of the documents. For example, they explained that certain reports were summaries written to management which helped clarify why some of the historical documents were so brief and included little supplementary technical information. In addition, the veteran Hanford employees provided recommendations on the development of conversion and correction factors for historical measurements of iodine-131 in the Hanford-area vegetation. The following veteran Hanford employees were invaluable in establishing what was done and why:

| | | |
|---------------------|-----------------------|----------------|
| Z. E. Carey | H. J. Paas (deceased) | J. K. Soldat |
| C. C. Gamertsfelder | R. W. Perkins | C. W. Thomas |
| J. W. Healy | K. R. Price | R. C. Thorburn |
| P. O. Jackson | D. L. Reid | L. D. Turner |

Along with the veteran Hanford employees, the following BNW staff are greatly appreciated for their assistance with the research for and publication of this report:

R. L. Dirkes - previous Environmental Monitoring Data Task Leader
R. W. Hanf - cataloguing of historical documents, database design and development
J. P. Duncan - preparation of vegetation-measurement databases
K. J. Lessor - preparation of vegetation-measurement databases
R. O. Gilbert - technical guidance and ad hoc reviews
S. D. Cannon - editorial support

(a) Memorandum (HEDR Project Document No. 03930003), "Notes from February 6, 1989 Meeting on Chemical Separation and Early Gamma Spectrometry Methods of Analyzing Iodine-131 Levels on Vegetation," from E. I. Mart, BNW, to Veteran Hanford Employees - P. O. Jackson, K. R. Price, J. K. Soldat, C. W. Thomas and to R. K. Woodruff, BNW, February 21, 1989. Memorandum (HEDR Project Document No. 03930008), "Notes on March 27, 1989 Meeting on Biases and Uncertainties in 1945-1957 Measurements of ¹³¹I Levels on Vegetation," from E. I. Mart, BNW Consultant, to R. K. Woodruff, BNW, September 27, 1989.

Definitions of Key Words

Absorption Parameter, F_a : Parameter that corrects for absorption of beta particles after they leave the precipitate samples and pass through air and the mica window of the Geiger Mueller (GM) detector. This correction depends on beta particle energy, the combined mass thicknesses of air and the mica windows, and the iodine-131 beta-absorption coefficient. This is one of the parameters that affects the reconstructed measurement conversion factor (M).

Activity: Number of nuclear transformations per unit of time (disintegrations per minute, dpm).

Backscatter Parameter, F_{bs} : Parameter that compensates for an increase in the counting rate from betas being reflected back towards the GM detector from the material upon which the sample is mounted. This is one of the parameters that affects the reconstructed measurement conversion factor (M).

Bias: Assumptions, procedures, equipment, or processes that skew results either greater than or less than the true value.

Blank Sample: Observed counting rate from an environmental vegetation sample that is not spiked but is prepared and analyzed in the same manner as routine environmental or spiked samples.

Conversion Factors: Needed adjustments to the historical correction factors. These adjustments are needed to account for any parameters that were not historically accounted for in the original correction factors.

Historical Detector Efficiency: Relationship of cpm/dpm that was historically used to convert background corrected counting rates (cpm) to disintegration rates (dpm). **Reconstructed Measurement Conversion Factor (M):** Reconstructed estimate of the ratio of iodine-131 disintegrations per minute to counts per minute (dpm/cpm) for the historical measurement process and detector systems used.

Reconstructed Total Conversion Factor (C): Best estimate of the ratio of dpm/cpm in the 1948 to 1951 measurements. This factor is determined by evaluating all of the parameters that would have affected what percentage of the iodine-131 disintegrations were registered as counts by the mica-window GM detectors. It incorporates the yield conversion factor (Y), the reconstructed measurement conversion factor (M), the decay correction factor (D_d), and the iodine-131 assumption correction factor (I_{cp}).

Yield Conversion Factor (Y): Ratio of the difference between the counting rates of a "spiked sample" and a "blank sample" expressed as a percentage of the "spike check" counting rate. Then a correction is made for the difference in the counting efficiency of the "spiked sample" and "spike check" caused by differing mounting characteristics. In 1949, yield was determined by the ratio of the net counting rates (cpm) of "spike" to "spiked sample" that then underwent the chemical extraction procedure. Historically, these yields were determined on a daily basis to assure that the analytical results remained within statistical control limits. The yield values used in the computational procedures were determined on a monthly frequency by averaging the daily yield measurements.

Correction Factors: Adjustments to the original counting data needed to account for any factors not accounted for historically.

Decay Correction, D_e : Exponential value that compensates for iodine-131 decay that occurred in the time interval between sampling and counting.

Iodine-131 Assumption Correction, I_{cf} : Fraction of activity resulting from iodine-131 decay. This corrects for the historical assumption that all of the net counting rates registered by a mica-window GM detector were generated by iodine-131.

Counting Rate: Number of counts registered by a radiation detector system per unit time, usually given in units of counts per minute. Counting rate is converted to disintegration rate by conversion factors that take into account the solid angle relationship between the source being counted and the detector dimensions, and other parameters such as scatter and absorption. Disintegration rate is converted to curies by the standard 2.22×10^{12} dpm/Ci.

Counts: Pulses that have been registered by a radiation detector system. In this report, counts refer to electronic pulses with each pulse generated by one ion cascade that occurred in those mica-window GM detectors used from 1948-1951. Each pulse was then registered as a count. The total counts were collected and divided by the counting time to derive a counting rate in counts per minute (cpm).

Detection Parameter, E_c : Parameter that accounts for the fraction of betas and gammas counted inside the GM tube, and for the coincidence/dead-time losses. This is one of the parameters that is considered in the reconstructed measurement conversion factor (M).

Disintegration: Spontaneous nuclear transformation (radioactivity) characterized by the emission of energy and/or mass from the nucleus. Disintegration rate is the number of disintegrations occurring per unit of time, which, in this report, is minutes.

Half-Life: Time required for a radioactive substance to lose 50 percent of its activity by decay. Each radionuclide has a unique half-life.

Point-Source Geometry Parameter, G_p : Ratio of the solid angle subtended by the aperture of the sensitive volume of the GM detector tube to the 4π solid angle with point source at the origin. Thus, it is that fraction of beta particles emitted from a point source at a fixed distance from the detector that would reach the sensitive volume of the detector if there were no self-scatter, no self-absorption, no absorption in the air or detector window, and no backing behind the source. This is one of the parameters that affects the reconstructed measurement conversion factor (M).

Sample Diameter Parameter, F_d : Parameter that corrects for the change in geometry caused by the spreading out of activity such that the sample precipitate does not represent a point source. This parameter compensates for horizontal displacement whereas the point-source geometry parameter compensates for vertical displacement. This is one of the parameters that affects the reconstructed measurement conversion factor (M).

Self-Absorption Parameter, F_{sa} : Measure of the absorption of iodine-131 beta particles by the vegetation. This is one of the parameters that affects the reconstructed measurement conversion factor (M).

Spike Check: Observed counting rate from a directly counted aliquot of iodine-131 solution that has been precipitated as silver iodide on a stainless steel planchet by the addition of a small amount of silver nitrate.

Spiked Sample: Vegetation sample, to which a known amount of a radioactive solution (in this case, iodine-131) has been added, that is analyzed concurrently with routine environmental vegetation samples of unknown concentration.

Contents

| | |
|--|-----|
| Preface | iii |
| Summary | v |
| Data Quality Objectives | vii |
| Acknowledgments | ix |
| Definitions of Key Words | xi |
| 1.0 Introduction | 1.1 |
| 1.1 History | 1.2 |
| 1.2 Chemical Extraction Method | 1.3 |
| 1.3 Geiger-Mueller (GM) Detectors | 1.4 |
| 1.4 Limitations | 1.4 |
| 2.0 Equations | 2.1 |
| 3.0 Y: Yield Conversion Factor | 3.1 |
| 3.1 Historical Method of Calculating Yield | 3.1 |
| 3.2 Valence Form of Iodine | 3.3 |
| 3.3 Representativeness of Spike and Blank Vegetation | 3.4 |
| 3.4 Other Aspects Affecting the Yield | 3.4 |
| 3.5 Skill of the Technician | 3.5 |
| 3.6 Reconstructed Yield | 3.5 |
| 4.0 M: Reconstructed Measurement Conversion Factor | 4.1 |
| 4.1 Self-Absorption Parameter, F_{sa} | 4.1 |
| 4.2 Air and Mica-Window Absorption Parameter, F_a | 4.3 |
| 4.3 Backscatter Parameter, F_{bs} | 4.3 |

| | | |
|--|---|-----|
| 4.4 | Point-Source Geometry Parameter, G_p | 4.3 |
| 4.5 | Sample-Diameter Parameter, F_d | 4.4 |
| 4.6 | GM Tube Detection Parameter, E_c | 4.5 |
| 4.7 | Summary of the Reconstructed Measurement Conversion Factor, M | 4.5 |
| 5.0 | D_e : Decay Correction Factor | 5.1 |
| 6.0 | I_{cf} : Iodine-131 Assumption Correction Factor | 6.1 |
| 7.0 | Iodine Contributions from Weapons Test Fallout | 7.1 |
| 7.1 | History | 7.1 |
| 7.2 | Historical Method of Determining Fallout | 7.1 |
| 8.0 | Summary and Conclusions | 8.1 |
| 9.0 | References | 9.1 |
| Appendix - Iodine-131 in Vegetation Collected Near the Hanford Site: Concentration and Count Data for 1948-1951 | | A.1 |

Tables

| | | |
|-----|--|-----|
| 3.1 | Average Yield, January through November 1953 | 3.6 |
| 4.1 | Summary of Parameter Values in M | 4.5 |

1.0 Introduction

During the early years of Hanford Site operations, iodine-131 released to the atmosphere from fuel dissolution was the major contributor of radiation dose to the public (Napier 1991). Iodine-131 is, therefore, the key element being studied in the Hanford Environmental Dose Reconstruction (HEDR) Project. Following the 1945-1947 period, when the largest quantity of iodine-131 emissions occurred (Heeb 1993; Napier 1992), the largest single emission known as the "Green Run" occurred in 1949 (Singlevich 1950). That expanded the period of most interest to the HEDR Project. Consequently, the Environmental Monitoring Data Task staff extended their attention to the evaluation of historical measurements of iodine-131 in vegetation beyond the initial 1945-1947 period to the 1948-1951 period. Not only did the Green Run occur during this time, but a new analysis technique (Singlevich 1948; Singlevich 1949; Leboeuf 1950) was instituted as well as efforts to control effluents by the addition of filters (silver reactors) in the stacks (Parker 1948).

Because the factors used from 1948-1951 to convert raw count data to iodine-131 activity levels (correction factors) and to adjust reported iodine-131 activity levels (conversion factors) did not account for all parameters affecting counting efficiency, other conversion and correction factors need to be applied. This report 1) describes the changes made in the method of analyzing vegetation samples for radioactivity, 2) provides correction factors for components not historically corrected, and 3) provides conversion factors for components that were historically corrected for some but not all parameters. The conversion and correction factors can be used to convert historical counting data per unit mass (cpm/g) into best estimates of iodine-131 concentrations ($\mu\text{Ci/kg}$).

To determine the conversion and correction factors, BNW staff reviewed the historical records and interviewed veteran Hanford employees familiar with the respective vegetation sampling and measurement techniques. The objective was to learn from them as much as possible about the collection and measurement techniques used, including radiation-detector systems, sample preparation and counting techniques, as well as how historical correction factors were developed and used. This approach allowed BNW staff 1) to evaluate the historical data and correction factors used and 2) to ascertain the additional appropriate corrections necessary to estimate reconstructed iodine-131 concentrations in vegetation.

This report is a companion report to Mart et al. (1993) which provides conversion and correction factors for 1945 through 1947. In that earlier period, investigators counted gross activity on relatively thick 1-gram sagebrush pellets. Beginning in 1948, they initiated chemical extraction of iodine-131 from 5-gram vegetation aliquots and then counted the beta particles emitted from the relatively thin resulting precipitate. This became the routine analysis technique starting in December 1948. Beginning in mid-1951, all parameters had been determined and were being applied to convert net counting rates to activity (Wolff 1951) which provided accuracy comparable to that of today. Hence, no other conversion or correction factors were developed for vegetation data after 1951.

An earlier report (Mart et al. 1993) provides the groundwork for the components chosen in this report. Mart et al. (1993) discuss the basic methodology for analyzing vegetation samples in the 1945-1947 period, the parameters and their values that need to be considered in reconstructing vegetation concentrations from net counting rates, and the counting systems used in the 1940s and

early 1950s. This report describes the new methods and discusses the estimation of 1948-1951 conversion and correction factors compared with those developed for the 1945-1947 period (Mart et al. 1993).

The conversion and correction factors provided in the 1945-1947 report by Mart et al. (1993) are recommended for the 1948 vegetation pellet counting data through November 1948. The conversion and correction factors developed in this report are appropriate for vegetation samples analyzed by chemical extraction of the iodine-131 and gross beta counting of the resulting precipitate, the measurement system used between December 1948 and the end of 1951. One exception to the 1948 pellet data may be the need to evaluate the iodine-131 assumption correction factor, I_{cf} , on a case-by-case basis because the addition of filtration systems to the exhaust stacks early in the year could have resulted in less iodine-131 being released compared to other beta emitters and hence a value less than 1.0 for I_{cf} . The conversion and correction factors provided in this report should be used to convert historical background-corrected counting data per unit mass (cpm/g) obtained from December 1948 through December 1951 into best estimates of iodine-131 concentrations ($\mu\text{Ci/kg}$).

In addition to providing conversion and correction factors for iodine-131 analyses of vegetation samples from 1948 to 1951, this report also provides a discussion in the appendix of the data sets for these years.

1.1 History

To overcome the problems of a large amount of self absorption (the vegetation absorbing the radiation it was emitting) and to increase the sensitivity of analysis, a procedure was developed to extract iodine-131 from vegetation and then count the gross activity of the resulting precipitate (Leboeuf 1950). In December 1948, this procedure entirely replaced the previous method, which consisted of counting the radiation from a 1-gram vegetation pellet (Singlevich and Paas 1949a). This remained the routine procedure with little modification until July 1957 when gamma spectrometry measurements entirely replaced chemical extraction procedures (McConiga and Soldat 1957).

Much work was done in the late 1940s and early 1950s to develop values for measurement parameters so that absolute (or near absolute) beta counting could be possible. This development resulted in a series of reports, the first one being published in 1950 (Healy et al. 1950). An update to the original work was published in 1954 (Schwendiman 1954), and finally Thomas and Polinsky (1956) published what they considered at that date to be the best compilation of correction parameters for a variety of radionuclides. The first application of this research to routine vegetation measurements started on August 27, 1951 (Wolff 1951). As discussed by Mart et al. (1993), the reports on development of these correction components were relied upon heavily in developing the conversion and correction factors presented here.

1.2 Chemical Extraction Method

A method of chemically separating iodine-131 from vegetation and then counting the resulting precipitate was developed in 1948. The new method increased sensitivity enough to allow detection to proposed new tolerance limits (Leboeuf 1950; Healy 1950). This was possible because 1) the sample was thin enough to allow it to be closer to the GM detector since the mounting card was inserted into the first shelf rather than the second shelf used for vegetation pellets, 2) interfering radionuclides were removed by the extraction process, and 3) the thin precipitate minimized self-absorption (Leboeuf 1950). In addition, the technique permitted large numbers of samples to be run routinely without much laboratory space for storing thick pellets or a lengthy processing time.

Use of the chemical extraction procedure for analyzing vegetation samples began in August 1948 (Singlevich 1948). While the new method was being "field tested," gross beta counting of pellets continued to be the routine method of analysis. In December 1948, chemical extraction became the sole routine method (Singlevich 1949). Thus, from August 1948 through November 1948, data from both types of analyses were included in monthly reports (Singlevich and Paas 1949a).

Veteran employees recalled that there were strict instructions on 1) how to cut samples, 2) what to collect (for example, twigs were not to be collected since they could not be digested by the chemical procedure), and 3) how to prepare the sample.^(a) However, documentation of those instructions and when and to what degree they were applied and followed has not been found in the historical literature reviewed by BNW staff. Veteran employees recalled collecting whatever vegetation was predominant in a given sampling location (Denham et al. 1988). This pattern of collection is also indicated by Singlevich (1950), who states that sage was the type of vegetation collected on the Hanford Site and analyzed because it was the predominant vegetation.

Singlevich (1949) states that the method of analyzing for iodine-131 in vegetation was principally based on research conducted by Leboeuf. The method involved weighing 5-gram aliquots; dissolving/extracting the iodine in a mixture of sodium hydroxide, sodium bisulfite, and sodium iodide; heating, filtering, and gross beta counting the resulting silver-iodide precipitate. The specific steps for the chemical extraction method are documented in Leboeuf (1950), Healy et al. (1951), Mickelson (1951), and Singlevich (1949). Veteran employees who reviewed the procedure used from 1948-1951 felt that it (and the one that replaced it in 1951) were both basically sound.^(a,b)

-
- (a) Memorandum (HEDR Project Document No. 03930003), "Notes from February 6, 1989 Meeting on Chemical Separation and Early Gamma Spectrometry Methods of Analyzing Iodine-131 Levels on Vegetation," from E. I. Mart, BNW, to Veteran Hanford Employees - P. O. Jackson, K. R. Price, J. K. Soldat, C. W. Thomas and to R. K. Woodruff, BNW, February 21, 1989.
- (b) Memorandum (HEDR Project Document No. 07930183), "Early Radiochemical Analysis of Vegetation Samples," from E. I. Mart, BNW, to File, March 29, 1988.

Using the 1948-1951 method, the precipitate adhered tightly to the filter paper because of the filtration process. Thus, no cellophane was needed to secure the precipitate to the mounting card as had been necessary with the vegetation pellets.^(a) The procedure required one hour per sample, and the samples were then counted for 15 minutes (Keene et al. 1960). Leboeuf (1950) indicates that the sensitivity for 5-gram samples was from 0.002 to 0.004 $\mu\text{Ci/kg}$.

The type and size of mounting affects sample spread, self absorption, and backscatter (see subsections 4.2, 4.3, and 4.4). Singlevich (1949) indicated with regard to December 1948 analyses that the precipitate was mounted on a stainless steel counting plate and counted by a thin window (i.e., mica-window) GM detector. The steel plates were 1 in. (2.54 cm) in diameter and were constructed of 0.004-in.- (2.54-cm-) thick high-grade stainless steel (Mickelson 1951). By January 1949, the mounting on the stainless steel plates apparently changed. The silver-iodide was mounted instead on 1-in. (2.54-cm) filter paper (Singlevich and Paas 1949b). A veteran Hanford employee^(b) questioned the use of 1-in. (2.54 cm) filter paper as he only recalled use of 1.5-in. (3.8-cm) filter paper. At least by January 1950, 1.5-in. (3.8-cm) filter paper was used (Leboeuf 1950). The effect of filter paper size on the historical correction components and the assumptions made here are discussed in Section 4.2. In addition to analyses for iodine-131, the longer-lived nonvolatile beta-emitters were analyzed from a separate vegetation sample using a dry ashing procedure (Keene et al. 1960; Wolff 1951; Singlevich 1949; Healy et al. 1951; Mickelson 1951). Hence, the vegetation data for 1948-1951 (Hanf et al. 1993) include concentration data for iodine-131 as well as for the longer-lived nonvolatile beta-emitters.

1.3 Geiger-Mueller (GM) Detectors

After chemical extraction, the gross activity of the silver-iodide precipitate was determined by the same type of beta counting on the mica-window GM detectors previously described by Mart et al. (1993). The mica windows ranged from 2 to 4.5 mg/cm² in thickness and were 1.125 in. (2.86 cm) in diameter. The GM detectors were encased in a cylindrical lead shield with 2-in.- (5-cm-) thick walls (Mickelson 1951; Healy et al. 1950). Aluminum shelves with grooves for sample mounting cards were used to position samples consistently to ensure reproducible geometry as shown in Figure 2.1 of Mart et al. (1993). The silver-iodide precipitate was counted on the first shelf, which was 0.191 in. (0.485 cm) from the mica window (Healy et al. 1950).

1.4 Limitations

Gilbert et al. (1992) developed a method for assessing uncertainty in vegetation samples analyzed for iodine-131 from 1945 through 1947. That report provides information on the uncertainty in direct gross beta counting of 1-gram pellets. While that report is only applicable to the vegetation

(a) Communication Log (HEDR Project Document No. 03930009), "Parameters Developed in 1956 in Thomas, Polinsky and Schwendiman Report," telephone conversation between E. I. Mart, BNW Consultant, and C. W. Thomas, BNW Consultant, December 29, 1992.

(b) Communication Log (HEDR Project Document No. 03930005), "Gamma Ray Contributions to Beta Counts," telephone conversation between E. I. Mart, BNW, and C. W. Thomas, BNW, March 9, 1989.

analyses performed in the 1945-1947 period, the improved analysis method discussed in this report is expected to result in decreased uncertainties for the 1948-1951 period. Hence, there will not be a separate uncertainty analysis for the vegetation data described in this report.

In addition, the following sources of potential biases are also not addressed in this report because they are deemed to be negligible:

1. **Sample Collection and Processing** - Sampling, handling, and transporting losses (except for radioactive decay) are assumed to be negligible and Wolff (1951) reported the "collection efficiency" factor for iodine-131 analyses on vegetation to be 1.0, further support that assumption.
2. **Deposition Variability** - Variability in the initial deposition of the material would depend on which part of a plant was sampled (e.g., top versus middle) (Paas 1949) and on which plant at a given location was sampled. Although this consideration is important in the interpretation of results, it is not significant in reconstructing the activity in a given sample once the sample had been collected.
3. **Part of Plant Sampled** - According to Paas (Denham et al. 1988), investigators chose their samples to identify maximum levels, thereby potentially biasing the results. This is another issue in the interpretation of results, but does not affect reconstructing the activity found on a given sample.
4. **Type of Vegetation Sampled** - Biases associated with different vegetation types would occur if the types of plants may have uniquely affected the extraction chemistry (Leboeuf 1950).
5. **Electronic Anomalies** - Biases may have occurred from the electronics of the Geiger-Mueller (GM) detectors used.
6. **Weapons Testing Fallout** - Small biases may have occurred as a result of fallout, although the fallout from 1949 to 1951 did not increase iodine-131 levels on vegetation except for a few isolated cases (Paas and Singlevich 1950).

2.0 Equations

To convert historical counting data (reported as counts per minute, cpm) to the best estimate of activity in microcuries, four conversion/correction factors are necessary, three of which were previously discussed by Mart et al. (1993): measurement, M ; decay, D_e ; and the iodine-131 assumption factor, I_{cf} . (Because the iodine extraction process effectively eliminates all other emitters, the iodine assumption is no longer a significant factor and, therefore, is assigned the value of 1.0.) These factors are included in this report for continuity, and their relative values are compared with those shown in the previous report. The new conversion factor, yield (Y), is discussed in greater detail in Section 3.0 of this report. Iodine contributions from weapons test fallout were considered but were found to be insignificant for this time period. These contributions were not of concern until after the first major atmospheric nuclear weapons test by the Soviet Union in August 1949. (See Section 7.0 for a brief discussion of the iodine contributions from weapons test fallout.)

The reconstructed components are needed because the values that were used historically in converting counting data to activity did not always account for all of the factors that would have affected the percentage of betas detected by the GM detectors. Nor were the historical measurements always documented in sufficient detail to construct unequivocally their origin and values. Beginning in mid-1951, all parameters had been determined and were being applied to convert net counting rates to activity concentrations which provided accuracy comparable to that of today. This is documented in the counting room logs (Wolff 1951). These logs were the collection of daily counting room data cards, typically filed in loose-leaf notebooks, on which were recorded such information as counter number, gross counting rate, background counting rate, sample number, net counting rate, and detection efficiency, from which activity level determinations could be made. Later this type of information was recorded in bound laboratory notebooks.

Historically, to convert observed net beta counting rates to activity per unit mass, the net background-corrected counts per minute were divided by the mass of the sample (1-gram pellets for the 1945-1947 period and 5-gram samples for the 1948-1951 period) and an empirically derived GM tube detection parameter. This latter parameter corresponds to the inverse of the measurement component as described in Section 4.0 of the Mart et al. (1993) report. To account for these new components and, thereby, convert the historical counting data from 1948 through 1951 to a best estimate of activity, Equation 3.4 of Mart et al. (1993) can be rewritten as

$$V (\mu\text{Ci/kg}) = 0.00045 Y M D_e I_{cf} U \quad (2.1)$$

where V = estimated iodine-131 activity in $\mu\text{Ci/kg}$
 Y = yield from the chemical extraction process; i.e., the fraction of iodine-131 extracted from vegetation and recovered as a silver-iodide precipitate that was then counted for gross activity (see Section 3.0)
 M = reconstructed measurement conversion factor (dpm/cpm) (see Section 4.0)

- D_e = radiological decay correction factor (unitless) which accounts for iodine-131 losses in vegetation due to decay during the interval between sample collection and counting (see Section 5.0)
- I_{cf} = iodine-131 assumption correction factor (unitless) which corrects for the fact that all of the counts registered by the GM detector did not result from iodine-131 disintegrations in the sample (see Section 6.0)
- U = background-corrected (cpm/g) measurement of a precipitate

The quotient of the standard conversion constants, 1000 g/kg and 2.22×10^6 dpm/ μ Ci, is 0.00045 (μ Ci)(g)/(dpm)(g).

3.0 Y: Yield Conversion Factor

Yield refers to the fraction of the iodine in the vegetation sample that was recovered by the chemical extraction procedure.

The yield would have varied depending on such parameters as

- method for calculating the yield
- the valence form of the iodine deposited on the vegetation
- how well the aqueous iodine-131 spike and blank vegetation (from which yield was calculated) acted as surrogates for the chemical and physical form of iodine deposited on the type of vegetation being analyzed^(a)
- other aspects affecting vegetation (species, moisture, and season)
- skill of the technician performing the measurement of iodine-131 on vegetation.^(b)

Given these sources of variability, there was a range of probable yield values, rather than a single value that applied universally to all vegetation samples analyzed from December 1948 through 1951. However, Leboeuf (1950) concluded that the yield should have been more or less constant, which was also the consensus of the veteran employees who met at Hanford in 1989.^(c)

In this analysis and consistent with the recommendations of the veteran employees, the assumption is made that since apparently no change in the chemical extraction procedure occurred from December 1948 through 1951, one yield factor provides an estimate of the "most representative" value. However, a range of yields around this representative value is believed to have been the actual case.

3.1 Historical Method of Calculating Yield

Exactly how the yield was determined from December 1948 through 1951, and whether the method for determining the value changed during that period, is not clear from the literature

-
- (a) Memorandum (HEDR Project Document No. 07930183), "Early Radiochemical Analysis of Vegetation Samples," from E. I. Mart, BNW, to File, March 29, 1988.
 - (b) Memorandum (HEDR Project Document No. 03930003), "Notes from February 6, 1989 Meeting on Chemical Separation and Early Gamma Spectrometry Methods of Analyzing Iodine-131 Levels on Vegetation," from E. I. Mart, BNW, to Veteran Hanford Employees - P. O. Jackson, K. R. Price, J. K. Soldat, C. W. Thomas and to R. K. Woodruff, BNW, February 21, 1989.
 - (c) Memorandum (HEDR Project Document No. 03930008), "Notes on March 27, 1989 Meeting on Biases and Uncertainties in 1945-1957 Measurements of ¹³¹I Levels on Vegetation," from E. I. Mart, BNW Consultant, to R. K. Woodruff, BNW, September 1989.

surveyed. According to Singlevich and Healy (1949), yield was determined by a ratio of counts per minute of spike (a known amount of iodine-131 that is analyzed concurrently with vegetation samples to evaluate the unknown concentration of the vegetation) to counts per minute of spiked sample that then underwent the chemical extraction procedure, taking the average of 62 samples. Leboeuf (1950) provides a more detailed description of how the yields were determined:

The yields were evaluated by adding a known amount of I^{131} to the sample before analysis. This amount of I^{131} was measured by placing aliquots of the solution used on a stainless steel counting plate and precipitating silver iodide by the addition of a small amount of silver nitrate, the plates were then dried under an infra-red lamp for a minimum time and counted.

Regarding routine analyses in 1956, Norton et al. (1956, p. 4) described the yield as being determined in the following way:

In some procedures, the analytical yields of individual samples are determined by gravimetric means, but in most analyses the yield is defined by a series of "controls". These "controls" consist of a "spike check" which is a directly counted aliquot of a radioactive solution, a "spiked sample" which is the same amount of activity added to a low level sample that is analyzed concurrently with the routine samples, and a "blank" which is a corresponding low level sample that is not spiked but is analyzed in the same manner. The yield is determined by expressing the difference between the "spiked sample" and "blank" counting rates as a percentage of the "spike check" counting rate and then making a correction for the difference in the counting efficiency of the "spiked sample" and "spike check" caused by differing mounting characteristics. These yields are determined on a daily basis to assure that the analytical results remain within statistical control limits. The yield values used in the computational procedures are determined on a monthly frequency by averaging the daily yield measurements.

It is clear from these descriptions and from counting room logs that the yield was not constant. In fact, in a later report Keene et al. (1960) indicate that the yield for the 1949-1951 procedure was 45 percent with a range from 40 to 60 percent, while Paas (1949), Wolff (1951), and Singlevich (1949) indicate a 70 percent yield was used. To reconstruct the yield conversion factor, BNW staff evaluated historical documents. In some cases, the value that had been used historically was recorded directly on the counting room logs. When the yield was not recorded, the historical value used was determined based on the reported activity that was calculated from the recorded net counting rates and knowing all other correction components used.

Other yield values were reported, some with uncertainties, although their exact meanings were not always clear. For example, Paas (1949) indicates that "control data" between December 28, 1949 and January 24, 1950 show that the yield averaged 73.2 percent with a range of from 50 to 100 percent. In a supplement to the site survey monthly report for March 1949, Paas (1949, p. 1 of supplement) indicates that the average yield was 69.1 percent with an error of 1.3 percent. Leboeuf (1950) and Singlevich (1949) indicate that the average yields were determined to be around 70 percent for routine samples and 83 ± 18 percent while testing the procedure. The exact meaning of 18 percent is not given. Singlevich and Paas (1949b) report that control vegetation samples, to which known quantities of iodine-131 were added, were run simultaneously with the regular vegetation

samples; these data indicated about 65-70 percent yield can be expected for this type of analysis. Mickelson (1951) also lists the yield as being 70 percent. However, in the supplement to the site survey monthly report for October 1949, Singlevich and Healy (1949, p. 21) stated, "Several months ago the I^{131} control data indicated that yields approached the 70% figure used here, whereas the yield this month is 37%." This 37 percent yield was obtained from an average of 20 samples.

This latter value (37 percent) was questioned by the veteran Hanford employees as not representative of the actual yield that had been obtainable.^(a) This doubt reflects discrepancies between the probable source of the 37 percent yield (Singlevich and Healy 1949) and comments made a short time later by Leboeuf (1950) regarding these measurements. Singlevich and Healy (1949) discussed a review of 62 spiked vegetation samples and spike checks that indicated a 0.37 yield for the ratio of counts per minute for the spike check to counts per minute for the spiked sample. Singlevich and Healy provided the same measurement correction parameter values for both the spike and the spiked sample. In discussing 62 measurements, which were probably the same ones discussed by Singlevich and Healy (1949),^(b) Leboeuf (1950) indicated that different sample spread and backscatter correction components were required for the initial spike check and for the analysis of the spiked vegetation sample. Leboeuf further noted that, when proper measurement correction parameters were used, a 67 percent yield was obtained, which he felt was still probably lower than the actual yield because of self-absorption of the residue on the paper.

Norton et al. (1956) also indicated a need for separate correction components to account for differences in the counting efficiency of the spiked sample and the spike check because of their different mounting configurations.

3.2 Valence Form of Iodine

Different valence forms of iodine were released from the Hanford Site and deposited on the vegetation, and these forms did not occur in consistent proportions for all batches. What valence form was released depended on the dissolution process. From 1949 through 1951, the bismuth phosphate process was used. Burger (1991) discusses valence forms of iodine released from separation stacks, noting that the iodine releases consisted of considerable amounts of particulate matter and mists containing iodides and iodates, and an organic form that was primarily methyl iodide. Once released, the iodine compounds were subject to oxidation and photochemical reactions. The final species (which may have included iodine dioxide, iodide and iodates as particulates, and perhaps hypoiodous acid and hydroiodic acid in aqueous aerosols) were then deposited on vegetation either directly or with rainfall. Once deposited, the iodine concentration would decrease from decay and weathering (the removal of iodine from vegetation by natural effects such as rainfall).

-
- (a) Memorandum (HEDR Project Document No. 03930008), "Notes on March 27, 1989 Meeting on Biases and Uncertainties in 1945-1957 Measurements of I^{131} Levels on Vegetation," from E. I. Mart, BNW Consultant, to R. K. Woodruff, BNW, September 27, 1989.
 - (b) Even though Singlevich and Healy (1949) and Leboeuf (1950) probably discuss the same 62 measurements, they note different precipitate sizes. Leboeuf indicated 0.5 in. (1.2 cm) and Singlevich and Healy 1 in. (2.5 cm) for the size.

The chemical form of the iodine may have affected the yield in addition to the deposition velocity. Veteran Hanford employees felt that varying iodine valence forms creates a large problem when attempting to reconstruct a single value for the yield.^(a) For example, the chemistry of inorganic iodine is completely different from that of organic iodine. Consequently, the extraction procedure may have easily reduced the inorganic forms of iodine to iodide but not the organic form. At the same time, the deposition velocity of the different chemical forms of iodine could affect the chemical mix of the different iodine forms on a given sample based on distance from the stack. For example, deposition velocities for organic iodine are two to three orders of magnitude less than for elemental particulate iodine (Denham et al. 1988). Because we do not know the exact chemical mix of iodine on any historical measurement, no compensation can be made for the valence state of iodine.

3.3 Representativeness of Spike and Blank Vegetation

A veteran employee questioned how well an aqueous spike solution could act as a surrogate for the iodine deposited onto dry vegetation.^(b) Because the spike was aqueous rather than dry, it may have been extracted with an efficiency different from that for iodine released from separation stacks which subsequently deposited on the vegetation. In addition, a question remains as to how well the chemical form of the spike solution represented the mix of iodine compounds deposited on vegetation. No information on the chemical form of the iodine spike was found in the historical literature reviewed.

3.4 Other Aspects Affecting the Yield

Leboeuf (1950) mentioned that the principal source of difficulty in extracting the iodine-131 from vegetation was the organic matter that was extracted with it. Although most of the material was removed by the aluminum hydroxide precipitation, any that remained tended to interfere with the silver-iodide precipitation. Different vegetation appeared to vary markedly in their capacity to interfere; green rabbit brush and dry cheat grass appeared to be the worst offenders. In addition to the species, moisture content of vegetation may also have affected the yield. A veteran employee noted that dry and wet vegetation may have had different extraction efficiencies.^(b)

Seasonal effects on vegetation may also have affected the yield as reported by Keene et al. (1960). Keene et al. (1960) indicated that during the winter, the yield ranged from 0.60 to 0.77. During the spring, it ranged from 0.70 to 0.80.

-
- (a) Memorandum (HEDR Project Document No. 03930003), "Notes from February 6, 1989 Meeting on Chemical Separation and Early Gamma Spectrometry Methods of Analyzing Iodine-131 Levels on Vegetation," from E. I. Mart, BNW, to Veteran Hanford Employees - P. O. Jackson, K. R. Price, J. K. Soldat, C. W. Thomas and to R. K. Woodruff, BNW, February 21, 1989.
 - (b) Memorandum (HEDR Project Document No. 07930183), "Early Radiochemical Analysis of Vegetation Samples," from E. I. Mart, BNW, to File, March 29, 1988.

3.5 Skill of the Technician

Carey (1951) indicated that skill impacted results in that the most important source of errors in the extraction process was the washing and filtration steps. Veteran Hanford employees felt that higher yields could be obtained when chemists, rather than laboratory technicians, conducted the extraction process. This would have occurred because presumably chemists would use greater care, and because chemists would work with ideal solutions under the best laboratory conditions.^(a) This conclusion is supported by Keene et al. (1960), who reported that the recovered yield (using the 1954 method) was 0.80 to 0.85 when done by a chemist, but only 0.60 to 0.80 when done by laboratory technicians.

3.6 Reconstructed Yield

In a March 1989 meeting with veteran Hanford employees, the consensus of the group was that the yield used to correct the counting data probably decreased in late 1949 because inappropriate correction parameters had been applied. This is supported by Leboeuf (1950). Thus, the veteran employees agreed that the most representative yield to be assumed for the 1949 through 1951 vegetation analyses would be the average of the monthly yields empirically derived in 1953.^(b) Beginning in January 1953, average yields were determined monthly and applied to all samples counted that month. Presumably knowledge and skill increased with time, so that the 1953 values represent the most reliable information up to that time and, therefore, are used as indicative of the variability that may have occurred earlier. Table 3.1 provides the average monthly yields for 1953.

-
- (a) Memorandum (HEDR Project Document No. 03930003), "Notes from February 6, 1989 Meeting on Chemical Separation and Early Gamma Spectrometry Methods of Analyzing Iodine-131 Levels on Vegetation," from E. I. Mart, BNW, to Veteran Hanford Employees - P. O. Jackson, K. R. Price, J. K. Soldat, C. W. Thomas and to R. K. Woodruff, BNW, February 21, 1989. Memorandum (HEDR Project Document No. 03930008), "Notes on March 27, 1989 Meeting on Biases and Uncertainties in 1945-1957 Measurements of ¹³¹I Levels on Vegetation," from E. I. Mart, BNW Consultant, to R. K. Woodruff, BNW, September 27, 1989.
- (b) Memorandum (HEDR Project Document No. 03930008), "Notes on March 27, 1989 Meeting on Biases and Uncertainties in 1945-1957 Measurements of ¹³¹I Levels on Vegetation," from E. I. Mart, BNW Consultant, to R. K. Woodruff, BNW, September 27, 1989.

Table 3.1. Average Yield, January through November 1953

| <u>Date</u> | <u>Percent</u> |
|----------------------------------|----------------|
| 01/27/53 | 57.9 |
| 02/19/53 | 65.3 |
| 03/26/53 | 64.2 |
| 04/30/53 | 64.3 |
| 05/28/53 | 62.7 |
| 06/25/53 | 63.7 |
| 07/30/53 | 62.4 |
| 09/03/53 | 62.4 |
| 10/01/53 | 63.2 |
| 10/29/53 | 62.8 |
| 11/27/53 | 63.6 |
| <hr/> | |
| MEAN | 63.0 |
| Standard Deviation (1σ) | [± 1.9] |

A 63 percent yield (a yield conversion factor of 1.59, which is the inverse of the yield expressed as a fraction) is assumed to best represent the actual yield for vegetation analyzed from 1949 through 1951. As noted earlier, a range of values around a mean yield would better represent actual processes.

4.0 M: Reconstructed Measurement Conversion Factor

Because Mart et al. (1993) provided a description of the methods used in the 1950s and their application to the 1945-1947 period to derive conversion parameters, this section will be limited to a discussion of the changes needed for the 1948-1951 period and a comparison of the respective values for the two different analysis methods (i.e., pellets versus precipitates). Recalling from Mart et al. (1993), the reconstructed measurement conversion factor, M, is modeled by the equation

$$M = (F_{sa}F_aF_{bs}G_pF_dE_c)^{-1} \quad (4.1)$$

where F_{sa} = self-absorption parameter
 F_a = cellophane, air, and mica-window absorption parameter
 F_{bs} = backscatter parameter
 G_p = point-source geometry parameter
 F_d = sample-diameter parameter
 E_c = GM tube detection parameter

The values provided here for the parameters in M are based both on our current understanding of the measurement techniques and detector systems in use from 1948 through 1951 and on research on absolute beta-counting techniques that was done in the 1950s using those same detector systems.

4.1 Self-Absorption Parameter, F_{sa}

The self-absorption parameter (F_{sa}) corrects for absorption of beta particles by the precipitate in which the iodine-131 is dispersed. As mentioned previously, the Hanford environmental measurements group attempted to minimize self absorption by maximizing the amount of precipitate spread.^(a)

Using the following equation and a mass thickness for the silver-iodide precipitate, the value of F_{sa} can be approximated^(b):

$$F_{sa} = 1 - (e^{-mx})/mx \quad (4.2)$$

-
- (a) Communication Log, HEDR Project Document No. 03930005, "Gamma Ray Contributions to Beta Counts," telephone conversation between E. I. Mart, BNW, and C. W. Thomas, BNW, March 9, 1989.
(b) Equation 4.2 does not account for self scatter.

where F_{sa} = fraction of iodine-131 betas not absorbed by passing through the vegetation precipitate
 m = beta-absorption coefficient, 0.0365 cm²/mg (Baltakmens 1977)
 x = mass thickness of vegetation (mg/cm²).

In reconstructing F_{sa} , two periods are defined based on the size of the filter paper used for the silver-iodide precipitate mount. The first is for the year 1949 when 1-in. (2.54-cm) filter paper is assumed to have been used, and the second is for the years 1950 and 1951 when 1.5-in. (3.8-cm) filter paper is assumed to have been used.

The mass thickness of the silver-iodide precipitate was calculated for 1956^(a) at between 3.85 and 4.41 mg/cm² when the precipitate was mounted on an asbestos filter paper (Norton et al. 1956). These values for mass thickness were obtained by dividing the precipitate weight; i.e., 37 mg mentioned in Norton et al. (1956) and 42.3 mg mentioned in Thomas and Polinsky (1956), by the silver-iodide "source" surface area of 1.5 square inches or 9.6 cm² obtained from Norton et al. (1956). Using Equation (4.2), the corresponding F_{sa} value is calculated as 0.92 to 0.93. However, given that the extraction methods differed in the 1949 to 1951 period (including sample size), the 1956 precipitate may not represent the 1949-1951 precipitate mass thickness.

No information on the actual silver-iodide precipitate weight generated from the extraction process used from 1949 through 1951 was found in the literature surveyed. The 1956 method used 15 grams of vegetation rather than 5 grams. Thus, the earlier process probably produced a smaller precipitate weight.

Wolff (1951) provides an F_{sa} value of 0.91, which is close to the value computed above for the 1950-1951 period. Therefore, the value provided by Wolff will be assumed for the 1950-1951 precipitates in the absence of information regarding silver-iodide precipitate weight. An F_{sa} of 0.87 is obtained for 1949 analyses.^(b) The self-absorption parameter values for the 1949-1951 period are significantly less important (i.e., factors of 2 to 3 greater) than those for the earlier vegetation pellet analyses. Mart et al. (1993) reported the F_{sa} values for the 1945-1947 period to range from 0.24 to 0.46 with mean values of 0.35 and 0.31, for hand- and press-formed pellets, respectively.

As with any analytical procedure, there was undoubtedly a range of mass thicknesses resulting from processing variances. In addition, the type of vegetation being analyzed may also have caused variances in self-absorption, as discussed by Leboeuf (1950).

-
- (a) Note that the chemical extraction method for 1956 was different from that used in 1949 through 1951.
(b) The 1949 value is obtained by dividing an assumed precipitate weight of 37 mg by the estimated silver-iodide surface area when filtered onto 1-in. diameter filter paper. Assuming 95 percent coverage by the precipitate, the surface area is 0.75 square inches (4.81 cm²). This equals a mass thickness of 7.69 mg/cm². This value is then used as an input parameter in Equation 4.2.

4.2 Air and Mica-Window Absorption Parameter, F_a

The absorption parameter, F_a , compensates for absorption of beta particles as they pass through the air and the mica window of the GM detector. The primary difference between the air and window absorption parameter, F_a , and that determined for the vegetation pellets is the shorter distance from the sample to the detector and the lack of a cellophane covering. Hence, the combined absorption parameter for the air and mica window of the GM detector for the vegetation sample precipitates analyzed from December 1948 through 1951 is 0.86, compared to the 0.73 value (Mart et al. 1993) for the 1945-1948 vegetation pellets.

For comparison, the total mass thickness of the 1945-1947 barriers is the sum of the individual mass thicknesses 3.7, 3.1, and 1.2 mg/cm² (respectively for the GM detector window, cellophane, and air) for the vegetation pellet analyses. The total mass thickness of the 1948-1951 barriers, however, is the sum of only 3.7 and 0.59 mg/cm² (respectively for the window and air absorption parameters applicable for this analysis) using the iodine-131 beta absorption coefficient of 0.0365 cm²/mg (Baltakmens 1977). In this analysis, as previously noted in Mart et al. (1993), the effects of scattering from the air and window are considered negligible and, therefore, are ignored.

4.3 Backscatter Parameter, F_{bs}

As previously reported by Mart et al. (1993), the backscatter parameter (F_{bs}) compensates for an increase in the counting rate from betas being reflected back towards the GM detector. The type of sample mount and energy of the beta spectrum affects the amount of backscatter that would have occurred. Singlevich (1949) indicates that in December 1948 the samples were mounted on 0.004-in. (0.01-cm) stainless steel plates with a mass thickness of 87 mg/cm² (Healy et al. 1950). However, as discussed above, filter paper is assumed to have been used from January 1949 through December 1951.

An F_{bs} value of around 1.1 is indicated by several historical sources (Leboeuf 1950; Wolff 1951; Schwendiman 1954; and Thomas and Polinsky 1956) for the filter paper on cardboard mounting arrangement. This is the same value assumed for the vegetation pellets, as reported by Mart et al. (1993).

4.4 Point-Source Geometry Parameter, G_p

The point-source geometry parameter (G_p) corrects for counting losses that occurred because some of the beta particles emitted did not reach the detector's sensitive volume. Because the silver-iodide precipitate was counted on the first shelf of the GM detector assembly (Schwendiman 1954), the distance from the first shelf to the mica window was only 0.485 cm compared to the second shelf distance of 2.07 cm (Mart et al. 1993, p. 2.11) used for the 1945-1947 analysis of vegetation pellets. As reported by Mart et al. (1993, p. 4.19), the value of G_p for the pellet analyses was 0.081-0.082, nearly one-third of the historical G_p values reported by Wolff (1951) and Leboeuf (1950) of 0.23. The value of 0.22 ± 0.01 given by Schwendiman (1954) is the value to be used for the 1948-1951 period.

4.5 Sample-Diameter Parameter, F_d

The sample-diameter parameter (F_d), corrects for the change in geometry caused by the spreading out of activity such that it does not approximate a point source. F_d corrects for horizontal displacement.

Historically, those responsible for measuring radiation attempted to minimize self-absorption by maximizing the amount of precipitate spread by keeping it evenly distributed^(a) (Schwendiman 1954). We, therefore, assume that the precipitate covered a significant proportion of the filter paper mount.

Norton et al. (1956) indicate that the surface area of a silver-iodide precipitate (using the 1956 method) was 1.5 square inches (9.6 cm²) or about 78 percent of the total surface area when mounted on a 1.5-in. (3.8-cm) diameter asbestos filter paper. A 1-in. (2.54-cm) diameter filter paper would have a surface area of about 0.79 square inches (5.1 cm²). Assuming that the precipitate covers a larger percentage of the area of the smaller filter paper (95 percent), the precipitate surface area would be ~0.75 square inches (4.82 cm²) for a diameter of 0.97 in. (2.48 cm).

By the first quarter of 1949, the silver-iodide precipitate was mounted onto filter paper (Singlevich and Paas 1949b). No mention is made of mounting onto a stainless steel mounting plate except for measurements done in December 1948 (Singlevich 1949).^(b) The first quarterly report in 1949 mentions 1-in. (2.54-cm) filter paper was used (rather than a stainless steel counting plate) (Singlevich and Paas 1949b). By at least January 1950, the filter paper mount changed to one that was 1.5-in. (3.8 cm) in diameter (Leboeuf 1950; Wolff 1951). It is not clear from the documents reviewed just when in 1949 a change in filter size occurred. In this analysis, an F_d based on silver-iodide filtered onto 1-in. (2.54-cm) filter paper is assumed for 1949, and an F_d based on 1.5-in. (3.8-cm) filter paper is assumed for 1950 through 1951.

By using Equation 4.5 of Mart et al. (1993) and the surface areas estimated above, F_d can be reconstructed for precipitation onto both 1-in. and 1.5-in. filter papers. The resultant F_d for the 1.5-in. (3.8-cm) diameter filter paper is 0.62; and the value for the 1-in. (2.54-cm) diameter filter paper is 0.90.

As with any analytical procedure, there was undoubtedly a range of silver-iodide precipitate surface areas resulting from processing variances. There would also have been clumps and other

-
- (a) Memorandum (HEDR Project Document No. 03930008), "Notes on March 27, 1989 Meeting on Biases and Uncertainties in 1945-1957 Measurements of ¹³¹I Levels on Vegetation," from E. I. Mart, BNW Consultant, to R. K. Woodruff, BNW, September 27, 1989.
- (b) A veteran employee contends that the 1-in. (2.54-cm) quoted value is in error because he does not recall using filter paper of that size. However, because the 1-in. (2.54-cm) value is documented in the historical record, we assume this filter size as the mount for samples analyzed in 1949 (Communication Log, HEDR Project Document No. 03930005, "Gamma Ray Contributions to Beta Counts," telephone conversation between E. I. Mart, BNW, and C. W. Thomas, BNW, March 9, 1989).

types of non-uniformity between samples in spite of attempts to evenly distribute the precipitate by using vacuum filtration.^(a) Thus, a range of F_d values would better represent actual conditions.

4.6 GM Tube Detection Parameter, E_c

The GM tube detection parameter (E_c) accounts for coincidence/dead-time losses and for the fraction of betas and gammas counted once inside the GM tube. This parameter (E_c) is discussed in more detail by Mart et al. (1993). In the present analyses, E_c is considered to equal unity.

4.7 Summary of the Reconstructed Measurement Conversion Factor, M

Table 4.1 summarizes the estimated values for the parameters in M . Values are given for two periods based on the assumed change in the size of the filter paper onto which the silver-iodide precipitate was mounted. As mentioned previously, this change in filter paper size would affect F_{sa} and F_d only.

Given the values in Table 4.1 and using Equation (4.1), the value of M (dpm/cpm) for 1949 is estimated to be 6.14 (a detection parameter of 0.16), and the value for 1950-1951 is estimated to be 8.52 (a detection parameter of 0.12). In comparison, these values are approximately an order of magnitude less than the values of M or approximately an order of magnitude greater than the $1/M$ values determined for the 1945-1947 hand- and press-formed pellets (see Appendix B, Tables B.3 and B.4 in Mart et al. 1993).

Table 4.1. Summary of Parameter Values in M

| <u>Period</u> | <u>G_p</u> | <u>F_d</u> | <u>F_{sa}</u> | <u>F_{bs}</u> | <u>F_a</u> |
|---------------|-------------------------|-------------------------|----------------------------|----------------------------|-------------------------|
| 1949 | 0.22 | 0.90 | 0.87 | 1.1 | 0.86 |
| 1950-1951 | 0.22 | 0.62 | 0.91 | 1.1 | 0.86 |

(a) Memorandum (HEDR Project Document No. 03930008), "Notes on March 27, 1989 Meeting on Biases and Uncertainties in 1945-1957 Measurements of ^{131}I Levels on Vegetation," from E. I. Mart, BNW Consultant, to R. K. Woodruff, BNW, September 27, 1989.

5.0 D_e : Decay Correction Factor

While it is important to account for the 8-day half-life of iodine-131 in the interval between sampling and analysis, the documents reviewed indicated that such corrections were accounted for. By 1949, the counting room data cards contained information on the date and time of sampling, as well as the date and time of sample collection, allowing for precise decay corrections to the nearest hour. However, in view of the much greater uncertainty expected from other components, decay correction in increments of days may be adequate, as provided in Table 5.1 of Mart et al. (1993).

6.0 I_{cf} : Iodine-131 Assumption Correction Factor

Chemically extracting iodine-131 from vegetation eliminated virtually all of the bias that occurred from the previous analytical method; i.e., gross beta counting of vegetation pellets and assuming all of the observed activity came from iodine-131. Some minimal but unquantifiable bias would have resulted from carryover of other radionuclides through the extraction process, such as naturally occurring radionuclides, fission products, and plutonium (Mickelson 1951). It is likely that the fraction of other radionuclides extracted by the chemical procedure varied with each batch because it would have been a function of the composition and chemical forms of the radionuclides released in the dissolution process, the sampling location, individual deposition velocities, and characteristics of the vegetation itself.

It is assumed in this analysis that all the activity in the silver-iodide precipitate came from iodine-131. The uncertainty associated with this assumption is less than the uncertainty introduced by assuming a single representative yield or the uncertainty associated with measurement conversion parameters.

The assumption that there is little carryover of other radionuclides is supported by historical decay studies, which indicate the activity consisted mainly of iodine-131 (Leboeuf 1950; Paas 1949). Leboeuf (1950, p. 7) states, "A decay curve measured with the iodine extracted from vegetation collected near the separations areas followed the 8 day half-life of I^{131} over a period of four half-lives indicating that little of the other fission products known to be present in the vegetation have been extracted."

In December 1949, contamination was introduced into the counting laboratory, causing anomalous conditions. This contamination occurred because of the Green Run, during which contamination was pulled into the laboratory through air ducts and tracked in by people.^(a) As a result of these conditions, "blank samples" were contaminated with about $2.0E-02 \mu\text{Ci/kg}$ vegetation. We infer from the wording in Singlevich (1950) that the recorded counts had not been corrected for this contamination. However, his description of this occurrence is brief and somewhat nebulous, leaving some uncertainty as to whether or not the measurements were corrected for the contamination.

The disintegration rate that corresponds to $2.0E-02 \mu\text{Ci/kg}$ is $\sim 44 \text{ dpm/g}$. The components used to convert from background-corrected counts per minute per gram to disintegrations per minute per gram in December 1949 were geometry (0.23), decay, and yield (0.37). Thus, assuming no decay, a disintegration rate of 44 dpm/g would have been recorded as about 3.7 cpm/g .

No correction factor is recommended for samples collected in December 1949 in view of 1) the uncertainty regarding exactly when the contamination occurred; 2) the uncertainty as to what is meant by "blank samples" (i.e., a vegetation blank or background counting rates); 3) the lack of knowledge

(a) Communication Log (HEDR Project Document No. 07930183), "Contamination Resulting from Green Run," telephone conversation between D. H. Denham, BNW, and J. K. Soldat, BNW, July 2, 1993.

about both when the laboratory was decontaminated and the half-life of the contamination (as with weapons test fallout, the activity released from the Green Run may have caused other iodine to be present in the samples analyzed); 4) the relatively low level of contamination; 5) the "background" values (presumably not vegetation background) recorded in December 1949 counting room logs that do not exceed background values noted for other periods (nominally 16 cpm); 6) the laboratory background counting rates that were routinely subtracted from the silver-iodide counting rates; and 7) the fact that although a contaminated vegetation blank would result in an extremely high yield factor, none was noted.

7.0 Iodine Contributions from Weapons Test Fallout

Radioactive fallout from weapons testing occurred only once in 1949, not at all in 1950, and several times in 1951. The fallout from 1949 to 1951 did not increase iodine-131 levels in vegetation (except for a few isolated cases), although increased activity was noted in other types of vegetation measurements. Because iodine-131 levels did not increase significantly, no correction factor is provided for fallout for the period 1949 through 1951. This may result in a small bias (to the high side) for a few samples.

7.1 History

Fallout from weapons testing first became substantial at Hanford in September 1949 (Parker 1954). Before that date, we can assume all activity around Hanford came from either Hanford operations or natural background. Much information on fallout was reported by Parker (1954) and in quarterly "environs" reports (e.g., Paas and Singlevich 1951; Paas 1953). This information included dates when detectable fallout occurred at Hanford, the probable source of detonation, deposition density (mCi/mile^2), and vegetation, air, and particle measurements.

Through December 1951, the dates when fallout was detected at Hanford were September 1949, February, April, May, October, November, and December 1951 (Parker 1954). The amount of fallout deposited at Hanford depended on atmospheric conditions (wind patterns and precipitation), detonation location and placement, and yield (kilotonage). As a result, no single value can be applied universally for contributions from fallout. The contributions to environmental measurements made by fallout in general were significant in some cases where the increase was an order of a magnitude or more.

Fallout did not contribute either equally or consistently to the different types of environmental measurements (air, particle, nonvolatiles, and iodine on vegetation). Typically, the environmental measurements indicate that radioactive particulates in air increased the most and iodine-131 levels on vegetation increased the least. For example, the weapons test in September 1949, increased readings on environmental ionization chambers by a factor of two at many locations (Paas and Singlevich 1950). At the same time, the average beta activity collected on air filters increased by a factor of 10. Even so, airborne iodine levels (as recorded by caustic scrubbers) increased only slightly, and there was no indication of elevated iodine levels on vegetation.

7.2 Historical Method of Determining Fallout

Historically, experienced health physicists determined intuitively whether activities were "normal" for Hanford, given such parameters as previous values, amount being discharged, condition of the silver reactors, and meteorological conditions. Besides professional judgment, fallout was identified in several ways:

1. Decay studies of the activity (Paas and Singlevich 1950; Denham et al. 1988). A primary indicator of fallout was the presence of environmental samples that were radioactive enough to do decay studies. Furthermore, the decay pattern for fallout was distinctive compared to that for radioactivity resulting from Hanford activities. Fallout would characteristically cause a rapid rise in activity followed by a rapid decrease (Denham et al. 1988).
2. Comparisons of similar measurements obtained from widely scattered locations.
3. Comparisons of radioactive particles found on Hanford air filters (via auto-radiographing them) with those in Oregon, Idaho, and Montana as well as with previous values (Paas 1952; Paas 1953).
4. Comparisons of the levels of non-volatile beta emitters in vegetation samples collected off the Hanford Site with both previous background levels and levels around Hanford (Paas 1952).

Analysis of particles was the main method for identifying fallout until it became possible to conduct isotopic analyses rapidly and accurately by gamma spectrometry. In short, there is no single correction factor by which historical vegetation data can be corrected for fallout. In addition, the need for such factors is not apparent as the documentation of Paas and Singlevich (1951), Paas (1953), and Parker (1954) demonstrated that they were well aware of each fallout event and took the necessary steps to interpret any samples collected at those times with that knowledge.

8.0 Summary and Conclusions

To convert historical counting data (reported as counts per minute, cpm) to the best estimate of activity in microcuries, four conversion/correction factors are necessary, three of which were previously discussed by Mart et al. (1993): measurement, M ; decay, D_0 ; and the iodine-131 assumption factor, I_{cf} . These are included in this report for continuity, and their relative values are compared with those shown in the previous report. The overall detection efficiency as indicated by the reconstruction conversion measurement factor (M) increased approximately an order of magnitude between the 1945-1947 pellet analysis technique and the 1948-1951 precipitate analysis technique. A yield of 63 percent for a yield conversion factor of 1.59 is recommended to best represent the actual yield for vegetation analyzed from December 1948 through 1951. Iodine contributions from weapons test fallout were considered but were found to be insignificant for this time period.

As derived in Equation (2.1), the reconstructed total conversion factor, C , equals V/U . By plugging the values for Y (1.59), M (6.14 from December 1948-1949, 8.52 from 1950-1951), and I_{cf} (1) into Equation (2.1), the reconstructed total conversion factor, C , equations become

$$C (\mu\text{Ci/kg})/(\text{cpm/g}) = 0.0044 \times D_0 \text{ for 1949} \quad (8.1)$$

$$C (\mu\text{Ci/kg})/(\text{cpm/g}) = 0.0061 \times D_0 \text{ for 1950-1951} \quad (8.2)$$

The conversion and correction factors provided in the 1945-1947 report by Mart et al. (1993) are recommended for the 1948 vegetation pellet counting data through November, the last month in which both pellet and iodine extraction samples were analyzed. One exception to this may be the need to evaluate the iodine-131 assumption correction factor (I_{cf}) on a case-by-case basis since it may have been influenced as early as March 1948 by the addition of filtration systems to the exhaust stacks. This addition could have resulted in less iodine being released compared to other beta emitters and hence a value less than 1.0 for I_{cf} .

The conversion and correction factors provided in this report are recommended for the December 1948 through December 1951 iodine-extraction analyses. These conversion and correction factors should be used to interpret, where necessary, the 1948-1951 vegetation data for use in model validation.

9.0 References

- Baltakmens, T. 1977. "Accuracy of Absorption Methods in the Identification of Beta Emitters." *Nuclear Instruments and Methods*, 142(3):535-538.
- Bayhurst, B. P., and R. J. Prestwood. 1959. "A Method for Estimating Beta-Counting Efficiencies." *Nucleonics*, 17(3):82-85.
- Burger, L. L. 1991. *Fission Product Iodine during Early Hanford-Site Operations: Its Production and Behavior during Fuel Processing, Off-Gas Treatment and Release to the Atmosphere*. PNL-7210 HEDR, Pacific Northwest Laboratory, Richland, Washington.
- Burt, B. P. 1949. "Absolute Beta Counting." *Nucleonics*, 5(2):28-43.
- Carey, Z. E. 1951. *Laboratory Notebook, Radiological Sciences Division*. HEW-4446-T, General Electric Company, Hanford Works, Richland, Washington.
- Denham, D. H., R. L. Dirkes, R. W. Hanf, T. M. Poston, M. E. Thiede, and R. K. Woodruff. 1993. *Phase I Summaries of Radionuclide Concentration Data for Vegetation, River Water, Drinking Water, and Fish*. PNWD-2145 HEDR, Battelle, Pacific Northwest Laboratories, Richland, Washington.
- Denham, D. H., E. I. Mart, and R. K. Woodruff. 1988. *Notes from Key Former Hanford Employees Workshop on Vegetation Data Biases and Uncertainties 1944 to 1948, September 19-21, 1988*. PNL-SA-16497, Pacific Northwest Laboratory, Richland, Washington.
- EML - Environmental Measurements Laboratory. 1992. *Environmental Measurements Laboratory Procedures Manual*. EML Report No. HASL-300, New York.
- Gilbert, R. O., E. I. Mart, D. L. Strenge, and T. B. Miley. 1992. *Uncertainty and Sensitivity Analysis of Historical Measurements of Iodine-131 for Vegetation in 1945-1947*. PNWD-1978 HEDR, Battelle, Pacific Northwest Laboratories, Richland, Washington.
- Gleason, G. E., J. D. Taylor, and D. L. Tabern. 1951. "Absolute Beta Counting at Defined Geometries." *Nucleonics*, 8(5):12-21.
- Gydesen, S. P. 1992. *Status of Document Search and Data Quality Objectives Efforts*. PNWD-2047 HEDR, Battelle, Pacific Northwest Laboratories, Richland, Washington.
- Hanf, R. W., J. P. Duncan, and M. E. Thiede. 1993. *Iodine-131 in Vegetation Collected near the Hanford Site: Concentration and Count Data for 1948-1951*. PNWD-2177 HEDR, Battelle, Pacific Northwest Laboratories, Richland, Washington.
- Healy, J. W. 1950. *H. I. Methods Development and History*. HW-89078, General Electric Company, Hanford Works, Richland, Washington.

Healy, J. W., L. C. Schwendiman, and R. C. Thorburn. 1950. *Counter Calibrations in the Health Instrument Methods Group*. HW-18258, General Electric Company, Hanford Works, Richland, Washington.

Healy, J. W., R. C. Thorburn, and Z. E. Carey. 1951. *H. I. Control Laboratory Routine Chemical Procedures*. HW-20136, General Electric Company, Hanford Works, Richland, Washington.

Heeb, C. M. 1993. *Iodine-131 Releases from the Hanford Site, 1944 Through 1947, Vol. 1: Text, Vol. 2: Data*. PNWD-2033 HEDR, Battelle, Pacific Northwest Laboratories, Richland, Washington.

Keene, A. R., D. L. Reid, H. M. Parker, E. W. Christopherson, V. R. Cooper, J. W. Healy, B. V. Anderson, J. M. Nielson, J. K. Soldat, and G. E. Backman. 1960. *Collection of Notes, Memos and Monthly Reports Regarding: Chemical Separation and Early Gamma Spectrometry Techniques for Determining ¹³¹I Levels on Vegetation; Comparison of Results by the Two Methods; and ¹³¹I Reporting Limits 6-23-54 to 12-6-60*. HW-89069, General Electric Company, Hanford Atomic Products Operation, Richland, Washington.

Leboeuf, M. B. 1950. *Analysis of Vegetation for I¹³¹*. HW-15743, General Electric Company, Hanford Works, Richland, Washington.

Mart, E. I., D. H. Denham, and M. E. Thiede. 1993. *Conversion and Correction Factors for Historical Measurements of Iodine-131 in Hanford-Area Vegetation, 1945-1947*. PNWD-2133 HEDR, Battelle, Pacific Northwest Laboratories, Richland, Washington.

McConiga, M. W., and J. K. Soldat. 1957. *Radioactive Contamination in the Hanford Environs for the Period April, May, June 1957*. HW-52803, General Electric Company, Hanford Atomic Products Operation, Richland, Washington.

Mickelson, M. L. 1951. *Radiation Background Survey*. HW-20246, General Electric Company, Hanford Works, Richland, Washington.

Napier, B. A. 1991. *Selection of Dominant Radionuclides for Phase I of the Hanford Environmental Dose Reconstruction Project*. PNL-7231, Pacific Northwest Laboratory, Richland, Washington.

Napier, B. A. 1992. *Determination of Radionuclides and Pathways Contributing to Cumulative Dose*. BN-SA-3673 HEDR, Battelle, Pacific Northwest Laboratories, Richland, Washington.

Norton, H. T., P. O. Jackson, and D. L. Reid. 1956. *Standard Practices Radiochemical Calculation Manual*. HW-46436, General Electric Company, Hanford Atomic Products Operation, Richland, Washington.

Paas, H. J. 1949. *Regional Survey Monthlies 1949*. HW-89075, General Electric Company, Hanford Works, Richland, Washington.

- Paas, H. J. 1952. *Radioactive Contamination in the Environs of the Hanford Works for the Period October, November, December 1951*. HW-24203, General Electric Company, Hanford Works, Richland, Washington.
- Paas, H. J. 1953. *Radioactive Contamination in the Environs of the Hanford Works for the Period July, August, September 1953*. HW-30174, General Electric Company, Hanford Atomic Products Operation, Richland, Washington.
- Paas, H. J., and W. Singlevich. 1950. *Radioactive Contamination in the Environs of the Hanford Works for the Period July, August, September 1949*. HW-18615, General Electric Company, Hanford Works, Richland, Washington.
- Paas, H. J., and W. Singlevich. 1951. *Radioactive Contamination in the Environs of the Hanford Works for the Period January, February, March 1951*. HW-21214, General Electric Company, Hanford Works, Richland, Washington.
- Parker, H. M. 1948. *Review of the Stack Discharge Active Particle Contamination Problem*. HW-9259, General Electric Company, Hanford Works, Richland, Washington.
- Parker, H. M. 1954. *Fallout Comparisons*. HW-33754, General Electric Company, Hanford Atomic Products Operation, Richland, Washington.
- Schwendiman, L. C. 1954. *Standard Practices Counting Manual*. HW-30492, General Electric Company, Hanford Atomic Products Operation, Richland, Washington.
- Seliger, H. H., and A. Schwebel. 1954. "Standardization of Beta-Emitting Nuclides." *Nucleonics*, 12(7):54-63.
- Shipler, D. B. 1993. *Integrated Task Plans for the Hanford Environmental Dose Reconstruction Project, FY 1992 through May 1994*. PNWD-2020 HEDR, Battelle, Pacific Northwest Laboratories, Richland, Washington.
- Singlevich, W. 1948. *H. I. "Environs" Report for the Month of August 1948*. HW-10998, General Electric Company, Hanford Works, Richland, Washington.
- Singlevich, W. 1949. *H. I. "Environs" Report for the Month of December 1948*. HW-12131, General Electric Company, Hanford Works, Richland, Washington.
- Singlevich, W. 1950. *H. I. "Environs" Report for the Month of December, 1949*. HW-15593, General Electric Company, Hanford Works, Richland, Washington.
- Singlevich, W., and J. W. Healy. 1949. *Vegetation Corrections for I¹³¹*. HW-89093, General Electric Company, Hanford Works, Richland, Washington.

Singlevich, W., and H. J. Paas. 1949a. *Radioactive Contamination in the Environs of the Hanford Works for the Period October, November, December 1948*. HW-13743, General Electric Company, Hanford Works, Richland, Washington.

Singlevich, W., and H. J. Paas. 1949b. *Radioactive Contamination in the Environs of the Hanford Works for the Period January, February, March 1949*. HW-14243, General Electric Company, Hanford Works, Richland, Washington.

Thomas, C. W., and D. M. Polinsky. 1956. *A Tabulation of the Isotopic Counting Correction Factors and Decay Schemes*. HW-18258-APP, General Electric Company, Hanford Atomic Products Operation, Richland, Washington.

Wolff, J. H. 1951. *Calculation Constants Used by Regional Survey*. HW-22682, General Electric Company, Hanford Works, Richland, Washington.

Appendix

Iodine-131 in Vegetation Collected Near the Hanford Site: Concentration and Count Data for 1948-1951

Appendix

Iodine-131 in Vegetation Collected Near the Hanford Site: Concentration and Count Data for 1948-1951

Introduction

This appendix provides a description of the databases of the 1948-1951 measurements of iodine-131 in vegetation and a description of the data from those databases that is being published. Rather than attach numerous pages of these vegetation data to the report for the few readers interested in them, the HEDR Project Office staff decided that a more economical and efficient method would be to make the data available on a computer diskette to anyone requesting it. The 1948-1951 vegetation data diskettes (Hanf et al. 1993) can be obtained from the Technical Steering Panel (c/o K. CharLee, Office of Nuclear Waste Management, Department of Ecology, Technical Support and Publication Information Section, P.O. Box 47651, Olympia, Washington 98504-7651). In addition, one printed copy of the data presented on the diskettes is provided in the U.S. Department of Energy Richland Operations Office Public Reading Room, located on the campus of Washington State University in Richland, Washington.

1948-1951 Vegetation Databases

With dBASE III PLUS, data in the 1948, 1949, 1950, and 1951 vegetation databases were compiled from published and unpublished (draft) reports and from unpublished historic counting room data cards. The original counting room data cards were located in the Records Holding Area of the Federal Records Center in Richland, Washington. Data cards had either been sorted by location into individual file folders or compiled in notebooks and identified by sampling trip name. Some of the notebooks contained the original field collection data sheets and maps of the area sampled. The field collection data sheets were useful for translating the sample numbers on the counting room data cards to location names and for identifying the type of vegetation sampled.

As indicated by Gydesen (1992), all information needed to date to reconstruct the radiation doses has been identified, sought, retrieved, evaluated, declassified, and made available. The amount of vegetation data available in the documents for 1948-1951 varied. The least amount of data was found for 1948 and the most for 1951. Data for 1949 and 1950 were also plentiful. Published average values represent a significant share of the 1948 database, but the bulk of the data for 1949, 1950, and 1951 were for individual samples and were obtained from the counting room data cards. Not all recovered data were entered in the 1948-1951 vegetation databases because of problems identifying specific sampling locations, including some data for the north Pasco/Ringold area. However, the databases include a large percentage (80-90 percent) of the recovered data and are deemed to accurately represent concentration values and net counting rates for samples collected at most locations during these years.

Reported data were usually average and/or maximum values expressed in terms of concentration per unit mass ($\mu\text{Ci}/\text{kg}$). The counting room data consisted of information for samples collected at individual locations on specific dates. These counting room data included gross background and net counting rates, counting times, shelf numbers, and, sometimes, correction factors for each sample as well as sample sizes and collection and analysis times.

Two significant releases of iodine-131 occurred during the 1948 through 1951 time period. An experiment in early December 1949 (the "Green Run") and equipment failures in the spring of 1951 (the silver reactor failures) resulted in larger than normal releases of iodine-131 into the atmosphere. Subsequent vegetation sampling showed elevated levels of iodine-131 on samples collected off site. Special vegetation samples were collected at many nearby and distant offsite locations following the Green Run to help define the area and intensity of impact. Additional sampling was also conducted following the discovery of the silver reactor failures in early spring 1951, but a fairly extensive routine sampling operation was already in place. All of the available data collected at offsite locations following these two events have been included in the 1949 and 1951 vegetation databases.

The data reported for samples collected during the first seven months of 1948 were labeled as iodine-131 but were actually a measure of total beta. A chemical extraction procedure to separate iodine-131 from other longer lived beta emitters was under development in 1948, and the first data for iodine-131 "in" vegetation started appearing in the published reports around August 1948. From August through November 1948, while the extraction technique was being perfected, both iodine-131 "in" vegetation data and "on" vegetation data were reported. The "on" and "in" vegetation terminology was used for a few months in the published reports to differentiate between the old analytical method and the new chemical extraction procedure, respectively. "On" vegetation data were not reported in the published reports after November 1948. From December 1948 through the end of 1951, most reported vegetation data included both iodine-131 concentrations in vegetation and concentrations for longer half-life beta emitters. However, a noteworthy exception to this occurred following the Green Run in early December 1949. Samples collected and analyzed during the period immediately following the Green Run were only analyzed for iodine-131.

All database entries have been verified for accuracy by comparing them to the information in the original document. When possible, verification was done by an individual who did not participate in the initial data entry. A hard copy of each database, signed and dated by the person or persons verifying the data, has been retained in the HEDR Project Office files.

1948-1951 Vegetation Data Published

The vegetation data diskettes (Hanf et al. 1993) have been converted from a dBASE format to a text file format to make the diskettes as user friendly as possible. Therefore, the data on the diskettes cannot be manipulated as would be possible with a dBASE file. The databases have been edited and shortened to make them easier to view and print. Each complete database consists of as many as 25 individual data fields (columns). The fields (columns) provided in this report contain the data being used by HEDR Project staff and of greatest interest to both the Technical Steering Panel and the

general public. A reader interested in any of the other data available (data for longer half-life beta emitters, cross references, time the sample was collected, etc.) should contact the HEDR Project Office to make arrangements.

The data presented in Hanf et al. (1993) only include iodine-131 concentration and counting rate data for vegetation samples collected off the Hanford Site, the area of greatest interest to the HEDR Project. Each entry (database record) is sorted or indexed according to 1) the collection location and 2) the collection date. The other data fields provide information on sample type (various types of vegetation), the number of samples collected, concentrations of iodine-131 in vegetation samples, sample counts and associated counting uncertainties, a reference for each record, and notes. Specifically, the data fields provided are:

Collection Site - The specific location or general area where the sample was collected.

Collection Date - The date the sample was collected.

Sample Type - The type of vegetation collected. The general term "vegetation" or "veg" was used when a more specific identification was either unknown or uncertain. The monthly reports generally did not provide information on vegetation type. The field collection data sheets almost always listed either the plant name or a plant specific letter code. This code has been partially deciphered because some of the field collection data sheets listed both the plant names and corresponding code letters. Some of the code, however, still remains unsolved. Several telephone calls were made to former field collection personnel, but no one could clearly remember to what the code letters corresponded. All available code letters have been included in the database. Counting room data cards also contained letter codes to identify the plant type being analyzed, but the sample code letter on the counting room card did not always match the letter code on the corresponding field collection data sheet. When a discrepancy of this kind arose, the field collection data sheets were assumed to contain the correct identification. This may or may not be true because many of the field sampling personnel were probably not trained in specific plant identifications.

| | |
|--|-----------------------------------|
| A - alfalfa | L - unknown |
| B - unknown | O - wheat |
| C - cheat grass (?) | P - unknown (pine?) |
| DC - unknown (dead/dried cheat grass?) | R - rabbitbrush |
| DG - unknown (dead/dried grass?) | S - sagebrush |
| DT - unknown (dead/dried thistle?) | T - tumbleweed or Russian thistle |
| DW - unknown (dead/dried weed?) | TH - unknown (thistle?) |
| E - evergreen | W - weed |
| G - grass | X - unknown |

of Samples - The number of samples collected. This is important for the published average data only. A zero in this column means the number of samples is unknown.

I-131 Avg ($\mu\text{Ci}/\text{kg}$) - The individual iodine-131 sample concentrations from counting room data cards and unpublished monthly reports or average iodine-131 values reported in published reports. For the 1948 database, "on" denotes the early measurements of total beta and "in" denotes the later measurements using a chemical extraction procedure.

I-131 Corr. Count - The corrected net sample counting rates of iodine-131 obtained from the counting room data cards. Again, "on" and "in" distinguish between the two measurement methods used in 1948.

I-131 +/- - The uncertainties reported with the corrected net sample counting rates of iodine-131. Uncertainties were generally, but not always, reported. They express 95 percent confidence interval on the true counting rate (1.645 x standard deviation). Again, "on" and "in" distinguish between the two measurement methods used in 1948.

Reference - The document number of the published documents or the Federal Records Center box number where the data for a particular sample were located. "UMR" was used to indicate data obtained from unpublished (draft) monthly reports. The draft reports sometimes contained specific data that were summarized or averaged in the published versions. A "Resources" section in this report provides the reader with the complete reference information for each document.

Notes - Several letters and symbols have been used in this category to further explain the data:

A - Indicates that the iodine-131 $\mu\text{Ci}/\text{kg}$ value was reported with a less than (<) sign.

\$ - Indicates that the sample was counted and that the reported concentration was actually zero (0.000). The dBASE III PLUS software automatically inserts zeroes into all empty spaces, making it nearly impossible to tell an actual zero from a database zero.

RI - Indicates some of the samples were counted more than once. This was usually done if the initial count was suspect and again if the recount value was questionable. In the databases, a recount usually follows the initial count (in other words, both are listed in the database, one right after the other). An RI in the notes column for the 1948 database indicates a recount of an individual "in" vegetation iodine-131 sample. For the 1949-1951 databases, an RI indicates a recount of an iodine-131 sample. Sometimes recounts were averaged with the original data, and a single concentration was given. Sometimes several recounts were averaged and a concentration was given, but a concentration for the original sample data was not given. Concentration data for recounted samples were only included when they could be related to a specific sample.

RB - Indicates a recount of an "on" vegetation sample, specific to 1948. Both concentration and counts values are affected.

Distribution

**No. of
Copies**

**No. of
Copies**

Offsite

Technical Steering Panel

D. S. Barth
University of Nevada
4505 Maryland Parkway
Las Vegas, NV 89154

W. A. Bishop
P.O. Box 47651
Olympia, WA 98504-7651

M. L. Blazek
Oregon Department of Energy
625 Marion Street N.E.
Salem, OR 97310

G. G. Caldwell
Tulsa City-County Health Dept.
4616 East 15th Street
Tulsa, OK 74112

S. N. Davis
Dept. of Hydrology and Water Resources
Building 11
University of Arizona
Tucson, AZ 85721

N. J. Germond
224 Iron Mountain Blvd.
Lake Oswego, OR 97034

P. C. Klingeman
Civil Engineering Dept.
Apperson Hall 202
Oregon State University
Corvallis, OR 97331-2302

K. J. Kopecky
Fred Hutchinson Cancer Research Center
1124 Columbia Street
Seattle, WA 98104

P. D. McGavran
Dept. of Health and Welfare
450 W. State Street, 4th Floor
Boise, ID 83720-5450

R. L. Morrill
Dept. of Geography, DP-10
University of Washington
Seattle, WA 98195

A. H. Murphy
3115 NW McKinley Drive
Corvallis, OR 97330

D. W. Price
Agricultural Economics
Hulbert Hall Room 211
Washington State University
Pullman, WA 99164-6210

M. A. Robkin
Radiological Sciences, SB-75
University of Washington
Seattle, WA 98195

G. S. Roessler
Route 1, Box 139H
Elysian, MN 56028

B. Shleien
2421 Homestead Drive
Silver Springs, MD 20902

**No. of
Copies**

**No. of
Copies**

A. P. Slickpoo, Sr.
809 Nez Perce Lane
P.O. Box 311
Kamiah, ID 83536

J. E. Till
Route 2 Box 122
Neeses, SC 29107

D. E. Walker, Jr.
P.O. Box 4147
Boulder, CO 80306

Other

2 DOE Office of Scientific and
Technical Information
Technical Information Center
P.O. Box 62
Oak Ridge, TN 37830

B. G. Brooks, EH-421
Room J-112
U.S. Department of Energy
Germantown, MD 20545

Z. E. Carey
32 Vista Court
Richland, WA 99352

18 K. CharLee
Technical Support and
Public Information
P.O. Box 47651
Olympia, WA 98504-7651

L. E. Denham
Centers for Disease Control
and Prevention
4770 Buford Highway N.E., F-35
Atlanta, GA 30341-3724

M. R. Donnelly
Public Health Service
Centers for Disease Control
and Prevention
2201 Sixth Avenue, RX-22
Seattle, WA 98121

C. C. Gamertsfelder
4728 Santala Drive
Knoxville, TN 37909

H. A. Haerer
Golder Associates, Inc.
4104 148th N.E.
Redmond, WA 98052

J. W. Healy
51 Grand Cannon Drive
Los Alamos, NM 87544

E. I. Mart
20509 N. Woodside Drive
Bend, OR

R. W. Perkins
1413 Sunset
Richland, WA 99352

K. R. Price
1329 Goethals, #3
Richland, WA 99352

D. L. Reid
1910 Harris Street
Richland, WA 99352

J. P. Thomas
HEAL
1720 N. Ash
Spokane, WA 99205

**No. of
Copies**

**No. of
Copies**

| | | | | |
|-----------|---|--|---|---|
| | R. C. Thorburn 132 Sandhill Circle Menlo Park, CA 94025 | | W. T. Farris M. D. Freshley R. O. Gilbert W. A. Glass (3) R. H. Gray S. P. Gydesen G. L. Harvey C. M. Heeb T. A. Ikenberry P. O. Jackson B. A. Napier J. V. Ramsdell D. B. Shipler S. A. Stage J. K. Soldat C. D. Taylor W. L. Templeton M. E. Thiede C. W. Thomas W. H. Walters Publishing Coordination Records Center (2) Technical Library (2) | K3-54 K6-77 K7-34 K3-53 K1-33 P8-55 K1-77 K8-34 K3-54 P7-07 K3-54 K6-55 K1-25 K6-55 K3-54 B1-40 K1-30 K6-52 P8-08 K6-60 K1-06 K3-70 P8-55 |
| | L. D. Turner 303 Comstock Richland, WA 99352 | | | |
| | Onsite | | | |
| 6 | DOE Richland Operations Office | | | |
| | R. F. Brich, TSD Public Reading Room (5) | A5-55 A1-65 | | |
| 61 | Battelle, Pacific Northwest Laboratories | | | |
| | D. M. Anderson G. L. Black (25) S. D. Cannon D. H. Denham (5) D. E. Deonigi P. W. Eslinger | K8-15 K1-25 K1-25 K3-56 K8-11 K6-52 | | |

END

**DATE
FILMED**

11/01/93

