**Conversion and Correction Factors for
Historical Measurements of Iodine-131
in Hanford-Area Vegetation, 1945-1947**

Hanford Environmental Dose Reconstruction Project

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This document has been reviewed and
approved by the Technical Steering Panel.

A handwritten signature in black ink, appearing to read "J. E. Till", is written over a horizontal line.

J. E. Till, Chair
Technical Steering Panel
Hanford Environmental
Dose Reconstruction Project

December 30, 1993
Date

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, 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 (Napier 1992b).

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 separations plants were operated. It was the emissions from the chemical separations plants that led to the release of radioactive iodine.

3,4,5,6 *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. (1993a) provide the vegetation data 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 1945-1947, and Denham et al. (1993b) provide the vegetation data conversion and correction factors for 1948-1951. The uncertainty in the 1945-1947 conversion factors is discussed by Gilbert (1992).*

2,5 *The conversion and correction factors will be used when necessary to adjust original beta counting data and historically reported radionuclide activity levels in selected vegetation samples to assist in the HEDR model validation studies. This report fulfills HEDR Project Milestone 0502A. It is the final report, replacing the previous version dated May 1993. Appendix C is a record of the TSP comments and BNW responses.*

Summary

This report is a result 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 HEDR Project is conducted by Battelle, Pacific Northwest Laboratories (BNW).

One of the radionuclides emitted that would affect the radiation dose was iodine-131. This report describes in detail the reconstructed conversion and correction factors for historical measurements of iodine-131 in Hanford-area vegetation which was collected from the beginning of October 1945 through the end of December 1947.

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 level in vegetation for 1945-1947. This has been accomplished by 1) providing information on historical methods of vegetation sampling and analysis, 2) providing information on the assumed efficiency of the Geiger-Mueller (GM) detector, and 3) developing factors needed to convert original gross beta counting rate data and reported iodine-131 activity levels to estimates of iodine-131 activities as they would be measured today. The potential impacts on the conversion and correction factors that have been addressed are 1) the radioactive decay that potentially occurred between sample collection and measurement, 2) the measurement efficiency due to the geometry of the measurement system, 3) the sample weight and configuration, 4) the radiation absorption and scattering, and 5) the original assumption that all beta activity in vegetation was due to iodine-131.

Although this report addresses measurement techniques from the start of Hanford operations, the conversion and correction factors provided only apply to 1-gram sagebrush samples measured from October 1945 through December 1947. It is limited to one-gram sagebrush samples because these were the samples predominantly collected starting in October 1945.

Technical Approach

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 measuring techniques. The objective was to learn from them as much as possible about how iodine-131 and other radionuclides dispersed once they were released from

the stacks, and about the collection and measurement techniques, radiation-detector systems, original counting data, and historical conversion factors. Although counting data are available from the 1940s, calibration techniques were limited. Therefore, the reconstructed conversion factors were developed by evaluating 1950s research on calibration techniques to determine absolute or near-absolute beta activity. The 1950s research involved the same thin-window (mica) GM counting systems as had been used in the 1940s to measure iodine-131 levels in vegetation. The research done in the 1950s is considered highly reliable for the counting samples and configurations for which they were developed. This reliability is evidenced both by the confidence expressed by the veteran Hanford employees and by several recorded accounts that show close agreement between independent Hanford-Site-activity determinations and those reported by the U.S. Bureau of Standards (WSBS, now National Institute for Standards and Testing).

Results and Conclusions

The results produced by the research and included in this report are conversion factors which may be used to convert original counting data to numerical values that represent the best approximation of the actual (as they would be measured today) amounts of iodine-131 deposited onto vegetation at the Hanford Site and surrounding areas.

Three main factors affected the true iodine-131 activity in a vegetation sample measured from 1945 through 1947. These factors are the measurement conversion factor (M , disintegrations per minute/counts per minute--dpm/cpm), the decay correction factor (D_e , unitless), the iodine-131 assumption correction factor (I_{cf} , unitless). Both D_e and I_{cf} must be individually determined for a given measurement date and location. On the other hand, M is generic for the type of vegetation pellet sample counted. The vegetation sampled was formed into pellets by hand until about mid-1946. Then, a hand-operated press was put into use to help ensure consistency in pellet dimensions. Thus, one value of M can be applied to hand-formed pellets and another value for press-formed pellets. The uncertainty associated with each factor (M , D_e , I_{cf}) is discussed in a companion report (Gilbert et al. 1992).

The factors developed in this analysis are approximations that can be used to derive best estimates of true iodine-131 activities in the historical vegetation samples. These conversion factors were developed by evaluating historical studies that developed parameters for absolute beta counting.

The results of this report must be viewed in light of the limitations in the scope of this report. While the results are necessarily approximations, they do provide a basis for estimating the overall uncertainty in the available vegetation data for 1945-1947. Although additional research would reduce the uncertainty in the conclusions, the uncertainty in the conversion factors (Gilbert et al. 1992) relative to the uncertainty in the models and model parameters being developed to predict radiation doses is small. Therefore, further refinement of these vegetation estimates is not deemed necessary.

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 uncertainty of the reconstructed iodine-131 levels was addressed by Gilbert et al. (1992) when they assessed the uncertainty of the factors now being published in this report. They evaluated all parameters of the factors addressed in this report and developed a possible range of values and a best estimate of the true value for each parameter. They used appropriate models, approximated the uncertainty of model parameters, and propagated those parameter uncertainties through the models to obtain the uncertainty of the reconstructed iodine-131 levels.

Completeness

Any further review of historical documents is not expected to have a major impact on the results of this report. Original Hanford-Site documents were reviewed as well as documents from non-Hanford-Site research. The non-Hanford documents confirmed that Hanford had historically (starting in the 1950s) applied appropriate techniques. 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 measurement techniques employed. In addition, the reconstruction of cross-seasonal 1-gram sagebrush pellets (see Section 2.5) provided empirical information on pellet dimensions.

Representativeness

This report provides reconstructed conversion and correction factors with which iodine-131 levels can be computed from historical data. The factors covered in this report are representative of those necessary for converting and correcting the historical data.

Comparability

Comparability is not an issue because the objective of this research is to provide factors for converting and correcting historical data to our best estimates of actual iodine-131 levels. This report does not provide the reconstructed iodine-131 values in vegetation. Also, no previous values other than those historically reported are available for comparison, and use of historical data for comparison is not valid because the historically reported iodine-131 levels could have been inaccurate by a factor of two or more. The 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 in the factors provided in this report and addressed by Gilbert et al. (1992) could be reduced by additional research. Additional reviews of documents and notebooks, empirical studies, and mathematical modeling could be used to further refine these estimates. However, such additional efforts are not expected to have a major impact on the values reported here nor on the end product of the HEDR Project.

Acknowledgments

BNW staff held three meetings with veteran Hanford employees 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 our understanding of how to interpret some of the documents. For example, they explained that certain reports were just 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:

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- R. O. Gilbert - technical guidance and ad hoc reviews
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- S. D. Cannon - editorial support (succeeding communication specialist)

Definitions of Key Words

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

7,8 **Activity:** The number of nuclear transformations *per unit time* (disintegrations per minute, dpm).

Adjustment Factor, A: The unitless ratio of the reconstructed measurement conversion factor, M (dpm/cpm), to the historical conversion factors assumed (dpm/cpm). The adjustment factors convert the historically reported iodine-131 activity levels on vegetation ($\mu\text{Ci/g}$ or $\mu\text{Ci/kg}$) for biased historical conversion factors.

Backscatter Parameter, F_{bs} : The parameter that compensates for an increase in the counting rate from betas being reflected back towards the GM detector. The backscattering medium is typically the material upon which the source is mounted (e.g., a stainless steel planchet). The ratio of the measured activity in the source without the backing material to that of the same source with the backing material is the backscatter parameter (F_{bs}). However, for the 1945-1947 vegetation pellets, the backscattering medium is the vegetation itself because the mass of the vegetation is large. This is one of the parameters that affects the reconstructed measurement conversion factor (M).

Beta-Detection Parameter, E_β : The parameter that considers those betas that entered the sensitive volume of a GM tube but did not generate counts.

Bias: An assumption, procedure, equipment, or process that skews results either greater than or less than the true value.

Burnup: The power, multiplied by the length of time at that power, that the given fuel has been subjected to in a reactor. The unit for burnup is MWd/ST (megawatt days per short ton). Burnup indicates how much of the fuel is burned up (used) and, conversely, how much is left to sustain criticality. Fuel is used up once it reaches the discharge burnup goal.

Cellophane-Scatter Parameter, F_{cel} : The parameter that accounts for an increase in the counting rate due to the cellophane that was placed directly on top of the vegetation pellets and

deflected additional betas into the GM detector. This is one of the parameters that affects the reconstructed measurement conversion factor (M).

Coincidence and Dead-Time Parameter, C_d : The parameter that considers the counts that were lost because a beta particle entered the GM tube's sensitive volume while it was recovering from a previous pulse (dead-time) or coincidentally with another beta.

Conversion Factors: The factors used to convert the observed net counting rate (cpm) to activity (dpm). These factors are necessary because not all beta particles emitted by vegetation samples were detected by the detector systems. Conversion factors account for parameters such as the sample counting geometry, sample size, absorption, and scatter.

Historical Conversion Factor: The relationship of dpm/cpm that was assumed from 1945 to 1947 in converting background corrected count rate (cpm) to disintegration rate (dpm). Note: From 1945 to 1947, the observed counts per minute registered by a mica-window GM detector for a 1-gram vegetation pellet were divided by the assumed detector geometry (cpm/dpm) to obtain dpm/g. The dpm value was then divided by the standard relationship (2.22×10^6 dpm/ μ Ci) to obtain μ Ci/g.

Reconstructed Measurement Conversion Factor, M: The 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. The measurement conversion factor (M) does not include the iodine-131 assumption correction factor or the iodine-131 decay correction factor. The unit for M is dpm/cpm.

Reconstructed Total Conversion Factor, C: The factor to convert 1945-1947 iodine-131 cpm/g data to the best estimate of μ Ci/kg levels. The unit for C is (μ Ci/kg)/(cpm/g). This factor is determined by evaluating all of the parameters that would have affected the percentage of the iodine-131 disintegrations that were registered as counts by the mica-window GM detectors used in the 1940s. It incorporates the reconstructed measurement conversion factor (M), the decay correction factor (D_e), and the iodine-131 assumption correction factor (I_{cf}).

Correction Factors: Factors used to correct the reported gross beta counting data and iodine-131 activity levels for parameters independent of the detector system and thus not compensated for by conversion factors. Two correction factors developed in this report are decay and iodine-131 assumption.

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

Iodine-131 Assumption Correction, I_{cf} : The 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.

Counter Resolving Time: The period of time during which ion-pairs from a previous ionization event render a GM detector unresponsive to additional radioactive particles.

Counting Rate: The 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 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 1945 through 1947. 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 : The 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).

Detector Efficiency: The inverse of the historical conversion factor. The unit is cpm/dpm. Historically, the term "geometry" was used to denote detector efficiency (i.e., the fraction of emitted beta particles that were recorded as counts). Counting data were divided by the geometry fraction to derive an activity value (dpm).^(a) Historically, the value for "geometry" was typically listed as a percentage. Geometry was the term used because the main parameter affecting detector efficiency was the counting geometry (i.e., the geometrical arrangement between the sample and the detector window). In February 1946, a self-absorption factor was added to account for those betas absorbed by the vegetation pellet. After February 1946, the term "geometry" denoted both the geometry and self-absorption factors.

Disintegration: A 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.

(a) However, in Table 2.5, the "Geometry Factor" is given as a number greater than 1 and hence is a "multiplier." This multiplier is the inverse of the geometry value historically used.

- 9 **Four-Pi (4π) Counting:** Counting techniques in which the geometry approaches *that of a sphere; i.e., 4 steradians*. That is, almost every particle emitted is detected and recorded as a count. This is achieved by preparing extremely thin sources to minimize self-absorption and placing the thin source within a detector system so that every emitted particle has close to a 100 percent chance of being detected.

Fuel-Cooling Time: The length of time elapsed between the cessation of fuel irradiation and the initiation of fuel dissolution.

Gamma-Detection Parameter, E_γ : The parameter that considers those gammas that generated counts. Because the source of the counts collected by a GM detector cannot be identified, counts originating from gammas would erroneously be interpreted as beta counts.

- 10 **Half-Life:** *The time required for an initial number of radioactive atoms to be reduced to half that number by transformations.* Each radionuclide has a unique half-life.

Mean Effective Distance, MED: The distance from within the vegetation pellet to the detector. At this distance, the corresponding point-source geometry parameter (G_p) and sample-diameter parameter (F_d) are representative of the entire pellet. The layers within the pellet, and thus the MED, are weighted by the fact that self-absorption occurred in the vegetation at a rate that increased with increasing pellet depth.

Original Counting Data: The historical data collected directly from the counting laboratory. Data refer to recorded counting rates (background corrected counts per minute). The counts were collected by a mica-window GM detector that was used for beta counting. Although these GM detectors were primarily beta detectors, they may also have detected some gamma rays, particularly those of low energy. The counting rate does not directly indicate activity in a given sample. Conversion and correction factors need to be applied to the counting rate to estimate the activity level in microcuries per gram.

Point-Source Geometry Parameter, G_p : The 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 air, cellophane, or window, and no backing behind the source. This parameter is one of the parameters that affects the reconstructed measurement conversion factor (M).

Radium D, E, F Standards, RaDEF: Sources used historically for deriving parameters for absolute beta counting. RaD is lead-210; RaE is bismuth-210; and RaF is polonium-210.

Release Fraction: The amount of a radionuclide emitted to the environment relative to the initial amount. In this report, it refers to the fraction of a given radionuclide that was released to the environment as a result of the fuel-dissolution process.

Sample-Diameter Parameter, F_d : The parameter that corrects for the change in geometry caused by the spreading out of activity such that the source (vegetation pellet) 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} : The 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).

Self-Scatter: The change in direction of iodine-131 betas during passage through the vegetation pellet. The scattered beta particles would also have been reduced in energy.

Standards: A solution or solid that contains a known amount of radioactivity (disintegrations per minute) at a given radionuclide per unit volume, unit weight, or in the entire source.

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1.0 Introduction

During the early years of plant operations at the Hanford Site, the iodine-131 released to the atmosphere from fuel dissolution was the major contributor of radiation dose to the public (Napier 1992b). Iodine-131 is, therefore, one of the key elements being studied in the Hanford Environmental Dose Reconstruction (HEDR) Project. The period 1945-1947 was when the largest quantity of iodine-131 emissions occurred and, thus, the period of most interest to the HEDR Project. A major pathway by which persons may have received doses of radiation from Hanford emissions was drinking milk from cows that grazed on vegetation contaminated with iodine-131. Consequently, one of the purposes of the Environmental Monitoring Data Task staff is to evaluate the concentrations of iodine-131 in vegetation as indicated by historical measurements taken from 1945 through 1947.

Because beta particles are emitted when iodine-131 decays, the detection of beta particles gives an indication (although only relative) of the presence of iodine-131. Thin mica-window Geiger-Mueller (GM) detectors were available in the 1940s to count beta particles and thereby provide counting data. Although the state-of-the art was on the cutting edge at the time, calibration^(a) techniques were not advanced enough to accurately convert the counting data to iodine-131 radioactivity (activity) levels.^(b) This was because iodine-131 standards were not available, and the understanding of all the parameters that affected detection of beta particles was inadequate.

Research to thoroughly investigate all the parameters needed to accurately convert beta counts to activity levels started in the late 1940s. In 1950, Hanford published a document (Healy et al. 1950) which details these parameters and provides adequate calibration techniques for iodine-131. Because this research is based on the same GM detector systems that were used in the 1940s to assess iodine-131 activity levels in vegetation, the reconstructed conversion factors presented in this report are mainly based on the 1950s and follow-on calibration research. No attempt was made to address the detector systems themselves: their electronics, voltage, or design. As a result, any potential biases resulting from inadequacies in the 1945-1947 detector systems are not considered.

(a) Calibration refers to the development of parameters in the measurement conversion factor (M) which is discussed in detail in Chapter 4.0.

(b) Not all the radiation emitted by iodine-131 reached the detector. Thus, correction factors were needed to determine the amount of radioactivity in the sample. However, accurate correction factors for iodine-131 for Hanford detectors were not available until the 1950s. The correction factors used from 1945-1947 provided biased activity values, typically recorded in units of microcuries per kilogram. Some aspects that affected what fraction of radiation emitted by iodine-131 was recorded by the detectors included distance from the detector, sample spread, and thickness of the vegetation being counted.

Original beta counting data can be found in such laboratory notebooks as those by Bulow (1945) and Dickinson (1946). Original beta counting data are more reliable than the historical reported iodine-131 levels because: 1) the reported activities were typically given as averages and we do not know how the averages were obtained; 2) the reported iodine-131 activities were biased, and determining the historical conversion factors used adds another element of uncertainty; and 3) supplementary information for original counting data, such as dates of sample collection and counting, type of vegetation, pellet size, and background, is typically found in the original laboratory notebooks.

Original beta counting data and reported iodine-131 activity levels for vegetation collected from August 1945 through early August 1946 have been found in historical literature. However, from August 1946 through December 1947 only reported iodine-131 activity levels have been found. When original beta counting data are available, they can be directly converted to our best estimate of iodine-131 activity levels. However, when only iodine-131 activity levels are reported, adjustment factors can be applied. The adjustment factors are a ratio of the reconstructed measurement conversion factor (M) to the historical conversion factors.

This report includes factors that can be used to provide best estimates of iodine-131 concentrations in vegetation, either by 1) application of reconstructed factors to directly convert the original beta counting data when beta counting data were available, or 2) factoring out historical conversion factors and applying reconstructed ones by using adjustment factors. Correction factors can be applied to both historical counting and activity data depending on the assessment data.

The limitations of each parameter are included in this report. The biases in the historical conversion factors and the methods of measuring concentrations in vegetation were identified by a review of historic literature and interviews with veteran Hanford employees. To provide context for the reader, we have included information on how the historical measurements were taken and on the 1950s absolute beta-counting research techniques (see Appendix A).

Because the size and density of the vegetation pellet affects the calibration parameters, the reconstructed conversion factors in this report are applicable only to 1-gram sagebrush pellets collected and measured according to the techniques used from September 1945 through December 1947. The factors do not apply to any other type of vegetation or any other pellet weight.

The reconstructed conversion factors reported here do not take into account any variability that may have resulted from sampling, handling, and transporting the samples. These processes are assumed to have caused no losses of iodine and thus to have caused no bias.

This assumption is supported by veteran Hanford employees who, based on both the difficulty of extracting iodine from vegetation even by chemical means and the fact that they did not recall incidences of glove or car contamination, believed that only negligible iodine losses would have occurred (Denham et al. 1988). We also do not take into account any variability in the initial deposition of iodine that may have occurred. This potential lack of representativeness is pointed out by Norton et al. (1956). It is also discussed by Paas (1949) at various times in a series of monthly reports. Paas points out that iodine-131 deposition patterns can have large variances in a given area. Although the deposition variability is a consideration for the interpretation of results, it is not a concern in the establishment of conversion factors.

Other potential sources of variability that have not been taken into account are the differences that may have resulted from sampling different parts of the sagebrush. An example of that variability is reflected in one limited empirical study done in 1949 on a single medium-sized (8.1-kg/18-lb) sagebrush (Paas 1949). In that study, both iodine-131 and radioactive non-volatile (radionuclides remaining after an ashing process) components were measured. The results of that study suggest that iodine-131 concentrations may have been greater on the top of the plant.

Gilbert et al. (1992) conducted parameter uncertainty and sensitivity analyses to evaluate the uncertainty of reconstructed iodine-131 measurements obtained using the conversion factors discussed in this report.

2.0 Historical Techniques and Background

From 1945 through 1947, beta-counting technology had not yet progressed to a point where accurate conversion factors specific to the analysis of iodine-131 activities in vegetation could be determined (Healy et al. 1950). The historical conversion factors in use at that time did not account for all of the parameters that affected overall efficiency of the beta-counting system. Some parameters were ignored or miscalculated, and accurate iodine-131 standards were not available. As a result, the iodine-131 activities in vegetation reported historically were biased. Indeed, the measurements were recognized as being relative at the time (Healy et al. 1950).

The primary intent of the initial routine environmental monitoring program at Hanford was to evaluate trends.^(a) Thus, initially the monitoring program was less concerned with absolute levels in a given sample than with the pattern exhibited in an area. Results were used more to compare results from location to location and over time than to document the absolute concentrations at given locations.

With time, more parameters were accounted for, and the accuracy of the reported iodine-131 concentration values improved. Understandably, monitoring staff were slow to adopt these improved factors because they would have affected any analyses of trends. By 1951, highly accurate conversion factors had been established so that near-absolute (rather than relative) beta counting was possible at Hanford (Healy et al. 1950). In addition, the method for analyzing iodine-131 activity in vegetation was improved by a chemical extraction method that was put into routine use in December 1948. The direct method of counting 1-gram pellets was entirely replaced by the chemical extraction method (Paas 1949). The approximate half-year overlap (August 1948 through November 1948) in methods enabled researchers to have continuity and a point of reference while testing the chemical extraction procedure (Singlevich 1948a; Singlevich 1948b).

2.1 Measurement Techniques and Instruments from 1945 through 1947

Late in 1944, a Site-Survey Group was organized to measure radiation exposures caused by Hanford emissions. One of the group's tasks was to develop measurement methods (Healy 1950). In 1945, measurement methods were in a state of flux because no precedent existed for

(a) Memorandum (HEDR Project Document No. 03930008), "Notes on the 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.

environmental monitoring of the type required by irradiated fuel dissolution. In addition, staffing for environmental monitoring was low, resulting in few vegetation measurements in early 1945 (Denham et al. 1988). New techniques were developed as the need for additional environmental monitoring became apparent. Experience and trial and error quickly led to changes, and, by the end of 1945, a program for routine vegetation sampling and monitoring was in place. The measurement method that had been established by then remained basically the same until it was replaced by the chemical extraction method in December 1948.

Table 2.1 summarizes our understanding of measurement methods from 1945 through 1947. Table 2.2 provides a broader historical overview. These summaries are based on information found in various reports and notebooks (referenced in detail in this report) and on discussions with veteran Hanford employees as reported in Denham et al. (1988) and two memoranda.^(a) Little documentation of precise procedures used before mid-1946 when the hand-operated press was put into operation has been found. Very little information is available on the procedures used prior to September 1945. At that time, more data become available. Healy et al. (1951) describe in detail the procedure for forming pellets using a hand-operated press, a method implemented around mid-1946.

In the original routine program, the predominant vegetation was sampled weekly at designated locations (Denham et al. 1988). Samples were placed in paper bags, labeled, and delivered to a counting laboratory (Patterson 1946). The vegetation was not artificially dried. It was counted in its natural condition upon arrival from the field (Denham et al. 1988).

In a historical 1957 overview report, Dozer^(b) discusses the types of vegetation sampled. Dozer states that many experiments and tests had been made on various types of vegetation. Based on these experiments, the consensus was that native sagebrush was the best absorbent of the effluent gases, and so, wherever possible, samples of native sagebrush were collected. However, it is not certain when this decision had been reached. Veteran employees recall simply that the predominant vegetation at a given location was sampled (Denham et al. 1988).

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- (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) Report (HEDR Project Document No. 03930002) on early vegetation experiments and tests from R. C. Dozer (General Electric Company) to B. V. Andersen (General Electric Company), August 13, 1957, Richland, Washington.

Table 2.1. Detailed Summary of Beta-Measurement Techniques (1945-1947)

Estimated Date of Implementation	Probable Technique
Before 7/20/45	Sagebrush was piled around a thin, glass-walled GM detector and the counting rate (in counts per minute) was recorded. Apparently no effort was made to determine activity level from the measured counts per minute. Equal sample sizes were visually regulated. Measurement results were only relative.
7/21/45	About 1.5 g of vegetation was spread over a 5-cm- (2-in.-) diameter hole in a mounting card. The hole was plugged by a watch glass. The sample was counted by a mica-window GM detector. ^(a)
8/45	About 1.5 g of vegetation was secured ^(b) over a 5-cm- (2-in.-) diameter hole in a cardboard mounting card. A holder for the sample was developed to ensure consistent (vertical) geometry. The sample was counted on the second shelf <i>from the top</i> of a mica-window GM detector (Figure 2.1).
9/45 - 10/16/45	Hand-formed ~ 1.5-g pellets of vegetation were secured on a cardboard mounting card and counted on the second shelf <i>from the top</i> of a mica-window GM detector.
10/17/45 - 6/46	Hand-formed 1-g pellets of vegetation were secured on a cardboard mounting card and counted on the second shelf <i>from the top</i> of a mica-window GM detector.
7/46 - 12/47	A 1-g pellet of vegetation was formed using a hand-operated press. The pellet was secured to a cardboard mounting card and counted using a mica-window GM detector.
<p>(a) See Figure 2.1 in Section 2.2 for an illustration of the mica-window GM detector system.</p> <p>(b) Samples were secured by taping (sticky side up) the cardboard mounting hole and then covering the vegetation with cellophane that was taped to the mounting card.</p>	

Table 2.2. Broad Summary of Beta-Measurement Techniques (1945-Present)

Period of Implementation	Technique
9/45 - 11/48	Gross beta counts of vegetation pellets
12/48 - 6/57 ^(a)	Chemical separation of iodine-131 from vegetation and other radionuclides, and gross beta counts of the resultant silver-iodide precipitate. The gross non-volatile component, which was also chemically separated, was beta-counted separately from the iodine-131
7/57 - Present	Gamma spectrometry
(a) Starting in August 1948 (Singley 1948a), both pellets and silver-iodide precipitates were beta-counted. This practice continued during the period that the chemical separation technique was being evaluated before it was implemented as the routine procedure.	

2.1.1 Initial 1945 Technique of Analyzing Vegetation

When operations at Hanford started, the major hazards from the emissions were expected to be direct radiation from the passing cloud or inhalation of iodine (Healy 1950). However, soon after the first fuel dissolving, contamination of the vegetation close to the stacks was noticed. Consequently, measurements of radioactivity in vegetation began in January 1945 (Parker and Gamertsfelder 1945). The first measurements were field surveys using portable GM detectors. These instruments were inadequate because the cold weather caused the amyl acetate quench gas added to the filler gas to precipitate (Denham et al. 1988). To overcome this problem, some vegetation samples were collected and analyzed in a laboratory. Although field surveys continued through 1947, laboratory analyses were conducted as well.

The first description of the laboratory analysis technique in the historical literature is by Cantril and Healy (1945) for beta counts taken in June and July 1945. The technique is described as placing vegetation (usually sagebrush) around a thin-walled glass GM detector in the lab and measuring the counting rate. Chamberlin^(a) described the GM detector as a thin-walled glass counter tube hooked up to a scaler which was useful in detecting small amounts of beta and gamma activity. The first use of this technique is not indicated in the literature surveyed. That this particular technique was the routine method in early 1945 is inferred by correlating information in Parker and Gamertsfelder (1945) and Cantril and Healy (1945).

(a) Memorandum (HEDR Project Document No. 03930001), "A Brief History of the Health Instrument Survey Group in the 300 Area," from R. W. Chamberlin (E. I. Du Pont de Nemours and Company) to W. H. Durum (E. I. Du Pont de Nemours and Company), July 14, 1945, Richland, Washington.

Parker and Gamertsfelder (1945) provide information on the maximum counts on sagebrush collected on the site for the week ending June 1, 1945. These samples are noted as standard samples. The mid- and late-July 1945 activities reported by Parker and Gamertsfelder (1945) agree exactly in both quantity and location with results reported by Cantril and Healy (1945) for the glass-walled detectors.

Because piling vegetation around a detector lacks standardization, no corrections for geometry, self-absorption, or weight can be applied for samples analyzed by this means. Therefore, the limited vegetation data available for the first half of 1945 should be considered only qualitative because actual concentrations cannot be determined reliably.

2.1.2 Hand-Formed Pellets

A table in a report by Cantril and Healy (1945) indicates a change in the counting technique between July 20 and 25, 1945. This change is inferred from a brief statement in a table footnote. The footnote indicates that samples collected from July 20 to August 10, 1945, were not corrected for self-absorption and were from 1.5-g samples of sagebrush spread over a circle 5 cm (2 in.) in diameter. The footnote also indicates these measurements were made with an "MW counter" (probably mica-window). That differs from the treatment of previous samples which was to use a GM tube that was glass walled. A corresponding change in laboratory analytical technique can be inferred from Parker and Gamertsfelder (1945) during the same period, the weeks ending July 20, 1945, and July 27, 1945. Parker and Gamertsfelder report use of a particular technique for the first and only time. This technique was described as spreading 1.5 g of vegetation on a watch glass below a mica-window GM counter with an assumed 8 percent (i.e., 0.08) detector efficiency corresponding to a historical conversion factor of 12.5. Healy believes that watch glasses were used for only a short time to plug the hole in mounting cards and were soon replaced by tape as he does not recall ever using watch glasses in vegetation analysis. The use of tape is consistent with the procedure Healy et al. (1951) discuss, and the only mention of a watch glass in the literature we surveyed is that by Parker and Gamertsfelder (1945). It, therefore, appears that watch glasses were probably not used in the routine vegetation monitoring program begun around September 1945.

The first beta count data (using a mica-window GM detector) found in a laboratory notebook are for samples collected July 13, 1945 (Bulow 1945). The notebook includes notations regarding sample size, geometry correction, and counting shelf, and all samples are noted as being sagebrush. Table 2.3 provides information about analytical methods gleaned from notations in this notebook.

Table 2.3. Notations on Iodine-131 Measurement Procedures (Bulow 1945)

Count Date	Measurement Procedure Notations
7/17/45	Samples were counted on the second shelf <i>from the top</i> . The disintegration rate is given in addition to the counting rate. Two samples were counted over four consecutive days. (This provides decay information useful in identifying the constituents, see Chapter 6.0). The detector efficiency ^(a) assumed for the sample collected on Route 4S, Mile Post #7 is ~8.2 percent. The detector efficiency assumed for the sample collected from Route 4S Mile Post #6 is ~9.1 percent. Why different detector efficiencies were assumed is not known.
7/28/45	Sample weights ranged from 1.46 to 1.6 g. An 8-percent detector efficiency was assumed.
8/17/45	Sample weights ranged from 1.4 to 1.7 g. An 8-percent detector efficiency was assumed.
8/22/45	Approximately 1.5-g samples. An 8-percent detector efficiency was assumed.
9/12/45	Sample weights ranged from 0.436 to 1.555 g. The reason three significant figures were given is unclear. Samples were counted on second shelf <i>from the top</i> .
9/20/45	Activity per gram computed from counts per minute is given for one sample. The historical conversion factor indicates an 8-percent detector efficiency was assumed, and no correction was made for one day of iodine-131 decay between sample collection and counting.
10/4/45	Samples were counted on second shelf <i>from the top</i> .
10/17/45	On the form line that requests "accuracy required," "1-g samples" was written. This may indicate a requirement to only count 1-g samples. This may have become standard from that time on because after this date Bulow makes no other reference to sample weight. Activities are listed next to the count data and indicate that an 8-percent detector efficiency was assumed and that no correction was made for decay between sample collection and counting.
11/16/45 - 12/21/45	Activities are listed next to the count data and again indicate an 8-percent detector efficiency was assumed, and no correction was made for the decay between sample collection and counting.
(a) Detector efficiency values are the inverse of historical conversion factor values.	

Our understanding of the iodine-131 measurement procedure, once the routine vegetation monitoring program started around September 1945, is that it began with collecting the predominant vegetation at a given location. From the sample, a 1-gram pellet measured by a pan balance (a pan-and-weight scale) was made by hand and secured to a mounting card by taping the hole and covering it with cellophane. The mounting cards were 8.26 cm (3.25 in.) by 6.35 cm (2.5 in.) with holes 2.54 cm (1 in.) in diameter in the center (Healy and Pahnke 1946; Healy et al. 1951). The mounting card was slipped into the second slot (*from the top*) of a card holder to ensure consistent vertical geometry and then was counted by a mica-window GM counter (Healy and Eisenacher 1946; Denham et al. 1988; Healy et al. 1951; Bulow 1945). The mounting cards tended to bow or wrinkle so that the distance was not necessarily exact from sample to sample.^(a) In addition, the pellets were not flat on the surface, and their thickness was not uniform.

Nonetheless, unlike the earlier method of placing vegetation around a glass-walled GM detector, this new technique was quantitative because the physical geometry could be reproduced and estimated and because a specified sample weight was counted.

Because pellet weight affects pellet dimensions, the conversion factors provided in this report are valid only for 1-gram vegetation samples. The fact that it became routine to count only 1-gram pellets has been inferred from the historical literature and from discussions with veteran Hanford employees. In this report we assume that after October 17, 1945 (see Table 2.3) only 1-gram samples were counted. We draw this inference because Bulow (1945) gives no specific information on sample weights after October 17, 1945, other than the statement "1-g samples required for accuracy." In addition, the conversion of counts per minute to microcuries per gram indicates no correction for other sample weights. All historical periodic reports that mention the procedure, however briefly, list the weight as 1 gram (e.g., Healy 1945).

2.1.3 Press-Formed Pellets

Use of the basic procedure described in Subsection 2.1.2 continued through 1947, except that a hand-operated press was developed to enhance uniformity of pellets starting in about mid-1946, according to Healy.^(b) The routine monitoring procedure can, therefore, be divided into phases: 1) use of hand-formed pellets before July 1946 (an estimated implementation date) and 2) use of press-formed pellets from July 1946 through 1947. Without any direct statements found in the historical documents on the exact timing of implementation, the date Healy recommended (July 1946) is the assumed transition date. The following information supports this assumption:

(a) Memorandum (HEDR Project Document No. 03930007), "Discussions with Former or Present Employees Who Worked at Hanford in the 1940s and 1950s," from E. I. Mart, BNW, to R. K. Woodruff, BNW, April 3, 1989.

(b) Letter (HEDR Project Document No. 03930004) from J. W. Healy, BNW Consultant, to D. H. Denham, BNW, February 23, 1989.

1. Healy states (Denham et al. 1988, p. 11) that

They made pellets by hand before they developed the hand-operated press. The press was developed to ensure uniformity of pellets, and to compress the material.

This comment indicates that use of the press was implemented sometime after the routine collection of vegetation had begun.

2. Healy interprets a statement in a document he found in his personal files to indicate that the press was implemented around mid-1946. Healy (1950, p. 7) states

Early in 1946 an estimate of the degree of contamination was made by measuring directly the activity of one gram of the vegetation, and the possible hazard to animals noted. This series of measurements indicated possible overexposure to animals even outside of the plant reservation and steps were immediately taken to reduce the amount of I^{131} emitted. A press for mounting one gram of vegetation in a reproducible manner was made and surveys of the area at distances up to 100 miles from the stack instituted.

3. Healy (1948b, p. 2) states

A routine program for measuring the activity deposited on vegetation was started late in 1945. The vegetation most prevalent in the particular region was sampled and one gram aliquots mounted in the form of a small pellet about 1-1/2 inches in diameter by using a hand operated press.

In this paragraph, Healy is summarizing the vegetation monitoring program, not giving a detailed description, again in retrospect from 1948. Although he mentions use of a press and instigation of the routine program in the same paragraph, the press may have been put into operation somewhat later than the routine program. Because veteran Hanford employees recall that pellets were first formed by hand and then a press was put into operation later (Denham et al. 1988), July 1946 is assumed to be the transition date.

The procedure for making press-formed pellets directly onto a sample card is described by Healy et al. (1951) who state the procedure was outdated by later analyses that were specific to iodine-131; i.e., chemical separation of iodine-131 and precipitation as silver iodide. However, the pellet procedure was noted as still being useful when large numbers of samples were measured for considerable amounts of activity. From their statement that "this procedure has been outdated" and the fact that the new procedure was put into place in 1948, the method described in 1951 is inferred to be the same method used for forming pellets by press prior to 1948. Healy et al. (1951, pp. 2.11.1-2.11.2) describe the procedure as follows:

1. Change stainless steel compression sleeve in vegetation press before starting to mount samples. The sleeve that has been used should be placed in the cleaning solution crock for one-half hour or longer.
2. Prepare mounting cards. Use mounting cards with 1" diameter hole. Place a short piece of cellulose tape over the hole on the back side.
3. Check pan balance to be sure it is balancing.
4. Place one piece of 4 x 8 scratch paper on each pan and place gram rider at one (1) gram.
5. Copy all identifying data from vegetation sack onto the mounting card. Carefully note sample numbers, date, sample type and time. All data should be copied on the back of the card (the side with the cellulose tape).
 - A. Mark all cards consecutively, e.g., the first sample mounted #1, the second #2, etc.
6. Cut enough vegetation from the sample to weigh 1 gram.
 - A. When preparing vegetation wear a leather glove on the left hand to keep from being stuck by thistle thorns.
7. Mount sample by the following steps:
 - A. Place mounting card in the press with the cellulose tape next to the iron base of the press.
 - B. Lower stainless steel sleeve.
 - C. Pour in vegetation sample
 - D. Place small cellophane square on top of sleeve.
 - E. Lower plunger.
 - F. Raise sleeve enough to allow cellophane to be flattened onto the card.
 - G. Fasten sample to the mounting card with 4 pieces of cellulose tape--one on each edge of the cellophane sheet.
 - H. Remove sample and repeat.

8. Mount off-area samples before using the stainless steel sleeve for any area samples.
9. Mark procedure, weight of analysis and analysts' initials on request sheets for all samples and send to the counting room for beta measurement.

2.2 Beta Counters

Beta counters were thin mica-window GM detectors made at Hanford by the Instrument Division in the 700 Area Tube Shop (717-A Building). Construction of the shop was completed in October 1944. The Tube Shop contained all of the equipment needed for evacuating, assembling, filling, and sealing mica-window GM detectors.

The methods for making mica-window GM detectors were developed at the University of California, used at the British Metallurgical Laboratory and Clinton Laboratory, and modified mechanically for use at Hanford (Du Pont 1946). Most of the information describing mica-window GM detectors comes from a compilation of semi-annual histories of instrument development by Du Pont (1946).

The mica-window GM detectors (Figure 2.1) (Healy et al. 1950) were made from 2.8-cm copper tubing machined in the 300 Area Instrument Shop in the 3717 Building. The tube case was 6.35 cm long and has a 3.18 cm outside diameter which corresponds to a 2.86-cm-mica-window diameter. The tubes were filled to a pressure of 10 cm of mercury with a 90:10 mixture of argon and ethyl alcohol (Du Pont 1946).

A mica window was cemented to a flange at the lower end of the tubes, and a center wire was supported at the top of the tubes by a suitable insulator. Mica was chosen because it had sufficient elasticity and could conveniently be prepared in sheets of nearly perfectly uniform thickness. In addition, the mass thicknesses of mica sheets were small enough for beta counting. That is, the thickness absorbed few beta (energy-dependent) but most alpha particles. In 1950, the mass thicknesses ranged from 2.0 to 4.5 mg/cm². For 13 counters, the average thickness was 3.7 mg/cm² (Healy et al. 1950). The range of mass thicknesses in 1950 may not reflect the range of mass thicknesses used in the period from 1945 to 1947, given that methods may have improved. However, because no other information is available, the 1950 values are assumed to represent the values for the 1940s in this analysis. Once attached, the mica windows bowed upwards in a slightly concave curve because of a pressure differential between the inside and outside of the tube (Schwendiman 1954).

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Figure 2.1. Mica-Window Geiger-Mueller Detector and Shelf Assembly
Used from the Mid-1940s through Mid-1950s (Healy et al. 1950).

The tubes were mounted at the top of a set of aluminum shelves designed to hold sample mounting cards at a set of five known distances from the counter face. The distance from the flange holding the mica window to the first shelf was 0.485 cm (0.191 in.). The distances from the other shelves to the windows were as follows: second shelf - 2.07 cm (0.85 in.), third shelf - 3.66 cm (1.44 in.), fourth shelf - 5.25 cm (2.07 in.), fifth shelf - 6.84 cm (2.59 in.) (Healy et al. 1950). However, the distance from the shelf to the window may not have been the same as the distance of the mounted sample from the window. For example, the cardboard mounting cards and the window may at times have been bowed or warped. In this analysis, the assumed distance from the second shelf upon which the vegetation pellet was placed to the detector does not account for any potential concavity in the window, bowing of the pellet mounting card, or non-uniformity of the pellet thickness.

The entire assembly was contained in a lead shield with walls 5.08-cm (2-in.) thick to minimize background radiation from the room and environment. The detector was operated as a self-quenched unit feeding into a scale of 64. Coincidence losses were assumed to be around 0.2 percent per 1000 cpm (Healy et al. 1950).

The service life of the tubes increased with time as the technicians' skills and experience increased (Du Pont 1946). Initially, a problem occurred with tube failure, especially at temperatures above 75°F. The failure was primarily due to leakage caused by the failure of the deKhotinsky cement seal used to attach the mica window. The service life of the seal depended on the skill with which the seal was made and the temperature to which it was exposed. During the first half of 1945, it was found that the mounting position of the quenching tube under the mica-window tube affected the temperature the seal could withstand. To prevent the temperature from rising too high, counting-room tubes were changed to self-quenched units instead of units operating with an external Neher-Harper quenching circuit (Du Pont 1946).

During the second half of 1945, performance of the mica-window GM tubes improved. The average service life increased from 18 to 119 days. This improvement came mainly from a better seal between the shell and mica windows, reflecting an increase in the skill and experience of the technicians who assembled the tubes. From January to August 1946, the average service life increased to 155 days.

Another reason some tubes were returned to the instrument shop in early 1945 was non-statistical (unpredictable) fluctuations in the counting rate (Du Pont 1946). All tubes had a tendency to count low for a short time after they had been operated at a very high rate. This fatigue factor varied from tube to tube. Some tubes recovered immediately, but others required an overnight rest after operating at even a relatively low counting rate. In the first half of 1945, no satisfactory test had been devised to pre-screen tubes. The tubes were simply

replaced after this kind of weakness was discovered (Du Pont 1946). Nothing more was found in the literature reviewed regarding this problem until instrument development for the second half of 1946 was documented. At that time, a study was conducted in which counting rate and fatigue tests were carried out on thirty mica-window GM detectors to determine the effects of operating a tube above 5000 cpm. Each tube was operated with a 2000-cpm standard to determine its normal counting rate. The tubes were then operated at a high counting rate for a short period and then immediately rechecked with the standard. The observed drop in efficiency varied from 0 to 10 percent for the various tubes (Du Pont 1946).

The only other mention of the fatigue factor is a brief comment in a 1944 memo (Overbeck 1944, p. 3). This memo states that

Mica window tubes have a memory effect which makes it difficult to reproduce counts after increasing and then decreasing the voltage or counting a very hot sample. The normal recovery time is 10 or 15 minutes. The warm up time varies from tube to tube and it is good practice to run the final plateau 24 hours after the high voltage is put on the tube.

Plateaus should be run at 2000 counts per minute with at least 10,000 total counts. The tube should not be run at higher than 10,000 counts per minute under normal conditions.

The mention of 10,000 cpm as the limiting counting rate indicates the fatigue factor was not a concern when counting the relatively low-activity environmental vegetation samples.

During the last quarter of 1949, all of the counting laboratory beta detectors were fitted with test Radium D, E, and F (RaDEF) sources whose activity had been determined by the alpha counters. These subsidiary standards were compared with sources from the U.S. Bureau of Standards (USBS, now National Institute for Standards and Testing). The average beta count was 0.7 percent off the expected standard which was considered adequate (Parker 1950). In this analysis, the beta counters referred to are assumed to be identical to the mica-window GM detectors used for analysis of vegetation pellets, from 1945-1947, although this is not explicitly stated.

2.3 Iodine-131 and RaDEF Standards

Because both iodine-131 standards and RaDEF standards were used in the 1950s to develop some of the parameters for absolute beta-counting and because the reconstructed conversion factors are largely based on this 1950s research, a discussion of these standards is

warranted. No information was found in the historical research on the iodine-131 standards used in the 1950s. However, Jackson recalls that the iodine-131 standards either came from the Oak Ridge Laboratory or the Hanford technical group (Denham et al. 1988). This discussion of iodine-131 standards is, therefore, limited to decay characteristics.

Accurate iodine-131 standards were needed to adequately calibrate the GM detectors for iodine-131 measurements because each beta emitter exhibits a unique beta spectrum. These beta spectra are unique because of their differing maximum and average beta energies, spectral distributions, coincident gamma activities, and probabilities of decay by various means.

The RaDEF standards were lead-210 (RaD), bismuth-210 (RaE), and polonium-210 (RaF). The RaDEF standards apparently came from two sources. They were either made at Hanford or obtained from the USBS. No mention of other sources has been found.

The exact date when Hanford started to obtain USBS RaDEF sources is unknown. However, Schwendiman (1954) lists the USBS RaDEF standards that were available in the Radiological Sciences Department at that time. His list gives USBS numbers, certificate disintegration rates, and calibration dates. The list includes two standards that were calibrated (presumably at USBS) on June 3, 1948. Thus, 1948 may have been the earliest date that Hanford received USBS RaDEF sources. These sources were plated on palladium-coated silver discs that were approximately 1.27 cm (0.5 in.) in diameter (Schwendiman 1954). It is assumed that USBS sources were used in the 1950s research on absolute beta-counting methods, given that Schwendiman (1954) compiled research methods and results for absolute beta counting of various isotopes.

Characteristics of the RaDEF standard and iodine-131 are provided in Table 2.4. Average beta energies were obtained from the International Commission on Radiological Protection (ICRP 1983). The other information was obtained from Shleien and Terpilak (1984).

2.4 Historical Conversion Factors

- 16 From September 1945 to February 1946, apparently only a single historical conversion factor was used to obtain disintegration rates in dpm/g for 1-gram vegetation pellets. The resultant historical conversion factor *of 12.5 is the inverse of the 8 percent assumed detector efficiency (geometry) (i.e., 1 divided by 0.08=12.5)* (Parker and Gamertsfelder 1945; Bulow 1945; Dickinson 1946; Lindvig 1946a). Although an 8-percent detector efficiency was probably assumed most of the time, it was not used exclusively (Lindvig 1946b; Bulow 1946).

Table 2.4. RaDEF and Iodine-131 Decay and Beta Energy Characteristics

Radionuclide	Half-Life	Decay Mode	Max. Energy (MeV)	Avg. Energy (MeV)	Maximum Beta Range (mg/cm ²)
iodine-131	8 days	beta	0.606 ^(a)	0.192	220
lead-210	22.3 years	beta	0.061	0.038	5.5
bismuth-210	5 days	beta	1.161	0.389	450
polonium-210	138 days	alpha	5.305	--	--
(a) This is the maximum beta energy for a beta spectrum emitted 89.4 percent of the time. However, the absolute beta maximum is 0.807 MeV from a beta spectrum emitted only 0.4 percent of the time. The corresponding maximum range for this higher-energy beta is 300 mg/cm ² .					

Starting in February 1946, another parameter was included in the historical conversion factor to compensate for the absorption of beta activity by the vegetation pellets themselves. This self-absorption parameter may also have compensated for sample spread, depending on the experimental method used in deriving the value. No information has been found regarding the experimental method used. Self-absorption was historically evaluated to be 3 (Healy and Eisenacher 1946). Therefore, to compensate for both geometry and self-absorption, the counting data were multiplied by a total historical conversion factor of 37.5 to arrive at the historically reported activity levels.

Table 2.5 provides examples of how to determine the historically assumed conversion factors from documents containing original beta counting data. The periods over which the historical conversion factors were applied were estimated by comparing the reported iodine-131 activities ($\mu\text{Ci/g}$) with the corresponding background corrected counting rates (cpm/g). That is, to determine the historical conversion factors used, the reported activity levels ($\mu\text{Ci/g}$) were divided by the applicable parameters to obtain the corresponding originally reported counting rates (cpm/g). Even though inclusion in the original notebooks of both the beta-counting rate and the corresponding assumed iodine-131 activity is rare, enough data were found to establish the historical conversion factors used and to subsequently develop adjustment factors (Bulow 1945; Lindvig 1946a; Dickinson 1946).

Table 2.5. Examples of 1945-1946 Measurement Data and Corresponding Reported Activity Values

Date	cpm ^(a,b)	Geometry Factor ^(c)	Self-Abs. Factor	$\mu\text{Ci/g}^{(a,d)}$	Reference
9/19/45	39	12.5	NA ^(e)	2.2×10^{-4}	Bulow (1945)
10/18/45	1170	12.5	NA	6.7×10^{-3}	Bulow (1945)
11/6/45	277	12.5	NA	1.6×10^{-3}	Bulow (1945)
12/21/45	761	12.5	NA	4.5×10^{-3}	Bulow (1945)
1/1/46	155	12.5	NA	9.0×10^{-4}	Lindvig (1946)
1/16/46	44	12.5	NA	2.6×10^{-4}	Dickinson (1946)
1/25/46	616	12.5	NA	3.4×10^{-3}	Dickinson (1946)
1/31/46	332	12.5	3	5.6×10^{-3}	Dickinson (1946)
2/12/46	35	12.5	3	5.9×10^{-4}	Dickinson (1946)
<p>(a) During this time period, apparently no decay corrections were made; the cpm and $\mu\text{Ci/g}$ values are exactly as they were recorded in the original notebooks.</p> <p>(b) The counting rate in the historical record had been corrected for instrument plus room background.</p> <p>(c) The geometry factor times the self-absorption factor equals the assumed historical conversion factor. Historically, "geometry" was listed as the inverse of the factor here (i.e., $1/0.08 = 12.5$).</p> <p>(d) The conversion of dpm to microcuries is via the standard relationship: $2.22 \times 10^6 \text{ dpm}/\mu\text{Ci}$.</p> <p>(e) NA = not applicable; no self-absorption factor was used at this time.</p>					

The historically assumed overall conversion factors can be determined by using the following equations, each representing a different time period:

$$\mu\text{Ci/g} = \frac{\text{cpm/g}}{G_h \times (\text{dpm}/\mu\text{Ci})} \quad \text{for 10/45 through 1/46} \quad (2.1)$$

$$\mu\text{Ci/g} = \frac{\text{cpm/g} \times \text{SA}_h}{G_h \times (\text{dpm}/\mu\text{Ci})} \quad \text{for 2/46 through 6/46} \quad (2.2)$$

$$\mu\text{Ci/g} = \frac{\text{cpm/g} \times \text{SA}_h \times D_h}{G_h \times (\text{dpm}/\mu\text{Ci})} \quad \text{for 7/46 through 12/47} \quad (2.3)$$

where cpm = net counting rate registered by the GM detector after correcting for the background counting rate; i.e., cpm with no source beneath the detector.

G_h = historical "geometry" which was assumed to be 0.08 from 1945-1947

SA_h = historical self-absorption factor which was assumed to be 3 when applied

D_h = decay correction factor used historically to account for decay occurring in the interval between sample collection and sample counting. The value of D_h varied depending on the length of the interval between sample collection and counting.

No count data, as opposed to activity data, have been discovered for the period from mid August 1946 through November 1947. It was, therefore, necessary to estimate the historical conversion factors assumed from information other than that in laboratory notebooks. However, one data sheet with counting rate information was found for December 1947. The historical conversion factor assumed at that time corresponds to Equation (2.3). From this, we infer that Equation (2.3) applies to the period from July 1946 through December 1947. In all cases, the mass is assumed to be 1 gram.

2.5 Pellet Dimensions

The historical Hanford documents reviewed contained only limited information about the actual dimensions of the vegetation pellets that were constructed from 1945 through 1947. Veteran Hanford employees recall that pellet diameters were about 3.81 cm (1.5 in.) (Denham et al. 1988). Unfortunately, the original pellets no longer exist.

However, the thickness and average diameter of a pellet are parameters in the expression that defines the reconstructed measurement conversion factor (M). (See Section 4.0.) Therefore, to estimate dimensions of the historic pellets, HEDR Project staff constructed several prototype 1-gram green sagebrush and dried cheatgrass pellets in 1989-1990. These prototypes were evaluated by several veteran Hanford employees who worked in the

environmental sampling program in the 1940s.^(a) Some disagreement arose on the cheatgrass pellets, but the consensus on the sagebrush pellets was that they generally resembled those made in the 1940s, only were too perfectly shaped. One person thought the historical pellets contained more leaves, but another person thought that stems would have been included because removing the leaves from the stems entailed too much handling. Apparently, the historical pellets were more crudely formed because personnel tried to minimize the handling of the samples.

Based on these comments, HEDR Project staff collected green sagebrush samples monthly for about a year (1989-1990) and constructed hand-formed and press-formed pellets using the procedures outlined in the historic documents. The majority of these pellets contained both leaves and small stems. Some pellets also contained flowers or parts of flowers. A total of 55 hand-formed and 55 press-formed 1-gram sagebrush pellets were constructed between September 1989 and July 1990--10 pellets of each type in September and usually 5 pellets of each type in other months.

Calipers were used to measure the thickness and diameter of each pellet. Thickness was taken as a single measurement for each pellet. Diameter was measured at four evenly spaced points ($\sim 45^\circ$ apart around the pellet surface). The average (arithmetic mean) diameter was computed from these four measurements. Surface area was computed by the equation

$$S = \pi r_a^2 \quad (2.4)$$

where S = estimated surface area (in cm^2)

r_a = average radius in cm, calculated from the four evenly spaced diameter measurements by the equation $r_a = [D_1 + D_2 + D_3 + D_4]/8$, where D_i represents the diameter measurements made at each of the evenly spaced ($\sim 45^\circ$) points.

In Appendix B, Table B.1 for hand-formed pellets and Table B.2 for press-formed pellets list dimensions of the reconstructed pellets (thickness, four measured diameters, computed average diameter, and computed surface area).

(a) Communication Log (HEDR Project Document No. 03930006), "Evaluation of Our Hand-Formed Sagebrush and Cheatgrass Counting Pellets (1 g)," telephone conversation between R. W. Hanf, BNW, and R. C. Thorburn, BNW Consultant, August 9, 1989.

3.0 Biases Overview

Two types of historical information on vegetation measurements for iodine-131 have been found. One type consists of original counting rate data sheets and laboratory notebooks (e.g., Herde 1946; Dickinson 1946; Bulow 1945). The other type consists of reported iodine-131 activities (typically as averages or sometimes maximums) found in monthly and quarterly reports (e.g., Singlevich 1948a; Healy and Turner 1946). The reported activities were determined in the 1940s by applying the historical conversion factors to the beta counting rates (i.e., background corrected cpm/g) to obtain disintegration rates (i.e., dpm/g), and then further converted to $\mu\text{Ci/kg}$ by the standard relationships between activity and disintegration rate and between grams and kilograms. Beta counting rate data have been found for the period from July 1945 to August 1946. However, from August 1946 through November 1947, only reported iodine-131 activity values have been found.

This section provides an overview of the equations used to convert the October 1945 through December 1947 data to our best estimates of actual iodine-131 activity levels in vegetation. October 1945 is used as the starting date because, as discussed in Section 2.0, that is the time when 1-gram sagebrush pellets are assumed to have been measured in the routine environmental monitoring program, and the reconstructed conversion factors only apply to 1-gram samples.

Three values are necessary for either converting original beta counting-rate data in cpm/g or adjusting historically reported iodine-131 activities in $\mu\text{Ci/kg}$ to a best estimate of true activity in $\mu\text{Ci/kg}$. These three values are M , the reconstructed measurement conversion factor; D_e , the decay correction factor; and I_{cf} , the iodine-131 assumption correction factor. (Each value is discussed in detail: M in Section 4.0, D_e in Section 5.0, and I_{cf} in Section 6.0).

These reconstructed values are needed because the factors historically assumed to convert count data to activity did not account for all of the parameters that would have affected the percentage of emitted betas registered as counts. Furthermore, the historical conversion factors were not derived using an iodine-131 standard of known activity.

Historically, the following equation was used to convert total counts collected by the GM detector to background-corrected counts per minute per gram (cpm/g):

$$U = (G - B) / W \quad (3.1)$$

where U = background-corrected cpm/g measurement made by a GM detector on a historical vegetation pellet

G = gross beta counts per minute in cpm

B = background counts per minute

W = weight in grams of the vegetation pellet.

The historical background-corrected counts per minute per gram are converted to a best estimate of activity by the equation

$$V = U C \quad (3.2)$$

where V = estimated iodine-131 activity in $\mu\text{Ci/kg}$

U = background-corrected cpm/g measurement made by a GM detector on a historical vegetation pellet

C = total reconstructed conversion factor in units of $(\mu\text{Ci/kg})/(\text{cpm/g})$.

The value of C is determined in turn by the equation

$$\begin{aligned} C &= D_e I_{cf} M \frac{1000 \text{ g}}{\text{kg}} \frac{\mu\text{Ci}}{2.22 \times 10^6 \text{ dpm}} \\ &= 0.00045 M D_e I_{cf} (\mu\text{Ci/kg})/(\text{cpm/g}) \end{aligned} \quad (3.3)$$

where 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 (discussed in 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 (discussed in Section 6.0)

M = reconstructed measurement conversion factor (dpm/cpm) (discussed in Section 4.0)

The factors 2.22×10^6 dpm/ μ Ci and 1000 g/kg are standard conversion constants. By substitution, the value of V for a given pellet is determined by

$$V (\mu\text{Ci/kg}) = 0.00045 M D_e I_{cf} U \quad (3.4)$$

To convert the historical count data to a best estimate of activity, a value for U is needed as well as the conversion factors discussed in Sections 4, 5, and 6. The ratio of M to the historical conversion factors adjusts reported activities. This unitless ratio is referred to as an adjustment factor (A) in this report. Reported activity levels can be multiplied by A to obtain an estimation of activity in the pellet. See Table 7.1 for estimates of A.

4.0 M: Reconstructed Measurement Conversion Factor

This section discusses the method and basis for approximating the reconstructed measurement conversion factor (M). The value of M is estimated by first evaluating all of the parameters that would have affected the overall efficiency of the counting system. The second step is to derive values for those parameters that are commensurate with the counting techniques used to evaluate vegetation pellets from 1945 through 1948. The appendices provide a discussion of the historical methods of determining parameters in M (Appendix A) and a discussion of the reconstructed pellet data and results for M (Appendix B).

For the counting systems in use at Hanford during the period from 1945 to 1947, most of the parameters in M are determined as described by Healy et al. (1950) and by Schwendiman (1954). Their research on absolute beta-counting began in 1948 with an extensive program to fully calibrate the Hanford beta detectors for specific radionuclides (Healy 1950). This calibration program continued and the original work published in 1950 was updated by Schwendiman (1954) and again by Thomas et al. (1956). Thomas et al. (1956, p. 1) state that

This report serves the two-fold purpose of presenting a tabulation of the correction factors used in determining the absolute activity of various isotopes and serving as a guide for all groups within the Radiological Chemical Analysis Operation. The results reported were selected as the most valid after careful scrutinization of all available data and are specific to the measuring methods and equipment employed with the Operation.

Thus, the 1956 results were at that time considered to be the best available parameters for computing M. This was reconfirmed by Thomas in 1992.^(a) However, Thomas et al. (1956) did not discuss the methods used in developing these parameters. Thomas indicated that the methods they had used to develop the parameters in M were consistent with those described by Schwendiman (1954). Other research on absolute beta-counting techniques was also carried out in the late 1940s and 1950s at areas other than Hanford (see Aten 1950; Baker and Katz 1953; Bayhurst and Prestwood 1959; Burt 1949; Gleason, Taylor, and Tabern 1951; Nervik and Stevenson 1952; Seliger and Schwebel 1954; and Yankwich, Norris, and Huston 1947). Just as the early Hanford researchers had done, BNW staff also used the non-Hanford research

(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.

to corroborate the Hanford research. In this particular case, the non-Hanford research has been considered in deriving the parameters in M.

Although the point is not explicitly stated in the early Hanford documentation, in the September 1988 workshop Healy stated, "The same counters used in 1945 were used in 1950" (Denham et al. 1988, p. 13). Hence, the absolute beta-counting research done in the 1950s is assumed to have involved the same mica-window GM detectors used from 1945 through 1947. This assumption is further supported by the fact that the research that culminated in the 1950s began in 1948, and brief descriptions of the GM detectors in use from 1945 through 1947 are consistent with the description of the counting system given by Healy et al. (1950) (see Subsection 2.2). Also, the results reported by Healy et al. (1950), Schwendiman (1954), and Thomas et al. (1956) are sufficiently similar to infer that the same counting systems were used.

The research done in the 1950s resulted in radionuclide-specific historical conversion factors that accounted for all of the parameters affecting overall counting system efficiency. By 1954, 4π counting techniques had been developed with which the activity in standards could be determined with a high degree of accuracy (Schwendiman 1954; Mickelson 1954). One potential source of bias in the absolute beta counting program is that the lead-210 component of the RaDEF sources emits a low-energy gamma ray (47 keV) 4 percent of the time. This low-energy gamma ray would probably have been counted with high efficiency.^(a) The result may have been a bias in the historical conversion factors developed using RaDEF sources. This potential bias in the development of historical conversion factors done in the 1950s, however, is not taken into account in this report. Otherwise, veteran Hanford employees consider the research documented in those 1950 records highly reliable.

Documented historical accounts also demonstrate the reliability of this 1950s research upon which much of the reconstructed conversions factors are based. Some historical accounts include the following:

1. Hanford staff independently evaluated the activity of standards received from the USBS. Schwendiman (1954) reported some results from a comparison of activities determined at Hanford using the absolute beta-counting methods with the quoted USBS values. The activities compared well, within 5 percent on 16 beta counters in 1950,

(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.

within 3 percent in 1952, and within 1 percent in 1953 when using 4π counting techniques. (Denham et al. 1988; Healy et al. 1950; Mickelson 1954).

2. Healy et al. (1950) gave an account of close agreement with Argonne National Laboratory in determinations of carbon-14 activity. One carbon-14 standard was obtained from Argonne. This standard had been calibrated carefully because it was to be used in a half-life determination. The Argonne value was $1.15 (\pm 0.014) \times 10^5$ dpm; the value obtained at Hanford was $1.22 (\pm 0.029) \times 10^5$ dpm. This was about a 6-percent difference. Although the methods used at Argonne are not known, this agreement at least indicates that two separate determinations were quite close, especially considering the GM detector systems were particularly hard to calibrate to carbon-14 because of its very low beta energy (0.16 MeV maximum; 0.05 MeV average).
3. In 1950, Hanford assessed the activity of a USBS carbon-14 standard. They found approximately a 5-percent variance from the USBS-determined activity. The USBS was notified about the bias and later confirmed that the Hanford determination was accurate (i.e., that the quoted USBS value was in error by about 5 percent) (Denham et al. 1988). Again, this accuracy was remarkable given the low beta energy of carbon-14.
4. Schwendiman (1954, Section 2.10) discussed a means of assessing the accuracy of disintegration rate determinations in the narrative that follows.

An index to the reliability of any interpretation of counting rates is the agreement among disintegration rates determined using different combinations of source mountings and geometrical arrangements for which counting factors have been determined. Ideally, when the same aliquot of a radioactive material is prepared for counting as a point source, as spread source, etc., and counted on any shelf, an identical disintegration rate should be obtained in each case if factors are exact. Only after satisfactory precision has been shown for sources counted in various configurations should estimates be made of overall accuracy.

The activity density of several solutions of isotopes was determined by preparing (a) point sources mounted by evaporation on flat discs, (b) point sources mounted by evaporation on standard one-inch discs, (c) sources spread with one cc of water and evaporated on one-inch dishes, and (d) sources spread with one cc of water and evaporated on standard 1-1/2 inch dishes. The sources were counted on the first four

shelves and disintegration rates calculated using the factors presented. In most cases a multiple of the aliquot used for the first and second shelf counts was used for the third and fourth shelf sources because of the much lower efficiency of counting. Four samples of each configuration were prepared to increase the reliability of the measurement. The percentage sample standard deviation for the samples measured indicates the reproducibility of the measurement including errors from all sources. Results presented in the following table [Table 4.1]:

Table 4.1. Comparison of Activities Determined for Iodine-131 on Different Shelves (Schwendiman 1954, Section 2.10)

Mounting	dpm/25 Microliters			
	Shelf 1 dpm/25 μ L $\pm\sigma\%$ (a)	Shelf 2 dpm/25 μ L $\pm\sigma$ %	Shelf 3 dpm/25 μ L $\pm\sigma$ %	Shelf 4 dpm/25 μ L $\pm\sigma\%$
Point source on flat disc	13,700 $\pm$ 2	13,600 $\pm$ 2	13,600 $\pm$ 1	13,600 $\pm$ 1
Point source on 1-in. (2.54-cm) dish	13,700 $\pm$ 1	13,700 $\pm$ 3	13,500 $\pm$ 1	13,400 $\pm$ 1
Spread source on 1-in. (2.54-cm) dish	13,500 $\pm$ 1	13,600 $\pm$ 3	13,800 $\pm$ 1	13,800 $\pm$ 1
Spread source on 1.5-in. (3.81-cm) dish	13,500 $\pm$ 4	13,600 $\pm$ 2	13,900 $\pm$ 1	13,300 $\pm$ 1
(a) As indicated by Schwendiman (1954), the percentage standard deviation ($\sigma\%$) indicates the reproducibility of the measurement including all errors. Four samples were measured for each configuration.				

In his document, Schwendiman also provided similar tables for sulfur-35, cobalt-60, and phosphorus-32 solutions. The data in these tables indicate a high degree of accuracy as well.

Schwendiman (1954, Section 2.10) also assessed the reliability of the activity determinations using historical conversion factors:

The accuracy with which disintegration rates can be determined is largely a function of the number of factors required to correct the counting rates, the

uncertainty in each factor contributing to the net error in the quoted rate. Greatest accuracy (and precision) is attained with sources mounted in very simple configurations, preferably as point sources with no residue, and counted at high geometries to minimize errors in the air-window extrapolation. For I^{131} and P^{32} mounted in these configurations agreement with U.S.B.S. I^{131} and P^{32} calibrated solutions has been within 5% using factors presented. It is recommended that all calibrations requiring accuracy be made with point sources on flat discs whenever possible.

For samples containing residues spread on large areas the absolute disintegration rate can seldom be quoted with an error as low as 5% and when soft emitters are counted an error of 10% can easily be introduced through inability to duplicate conditions under which the factors were determined and as a result of the uncertainties in the values of the factors.

From Schwendiman's (1954) narrative, it is inferred that although the reliability of the historical conversion factors was probably quite high for the counting conditions they represented, the uncertainty of the reconstructed conversion factors for vegetation pellets is also quite high. The counting conditions for the vegetation pellets have not been reproduced, the pellets were far from being point sources, and the spread of the vegetation certainly was not evenly distributed. In addition, the pellets were thick, introducing the inherent variability in distance (geometry) from the source and size (sample-diameter) of every pellet, as well as self absorption. Gilbert et al. (1992) discuss in detail the uncertainty surrounding the reconstructed conversion factors for vegetation pellets.

4.1 Equation for Measurement Conversion Factor, M

Because much information was available, the method chosen for approximating M involves the evaluation of 1950s historical research concerning the parameters that affect M for the counting systems used in the 1940s. The parameter values and resulting values for M are given in Appendix B.

The measurement conversion factor, M, as partially derived by Healy et al. (1950), Schwendiman (1954), and Thomas et al. (1956) is modeled by the equation

$$M = (F_{sa} F_a F_{bs} F_{cel} G_p F_d E_c)^{-1} \quad (4.1)$$

where F_{sa} = self-absorption parameter
 F_a = absorption parameter
 F_{bs} = backscatter parameter
 F_{cel} = cellophane-scatter parameter

G_p = point-source geometry parameter
 F_d = sample-diameter parameter
 E_c = GM tube detection parameter.

The self-absorption parameter (F_{sa}) compensates for self-absorption of beta particles by the vegetation in which the iodine-131 was dispersed. This compensates for those beta particles that are absorbed by the vegetation when they strike vegetation particles. This parameter is dependent on both energy and the mass thickness of the vegetation and is discussed in detail in Subsection 4.2.

The absorption parameter (F_a) compensates for absorption of beta particles after they leave the vegetation pellet and pass through cellophane, air, and the mica window. This parameter depends on beta particle energy, the combined mass thicknesses of cellophane, air, and the mica window, and the iodine-131 beta absorption coefficient. The F_a parameter is discussed in detail in Subsection 4.4.

The backscatter parameter (F_{bs}) compensates for an increase in the counting rate from betas being reflected back towards the GM detector. The backscattering medium is typically the material upon which the source is mounted (e.g., the stainless steel planchet). The ratio of the measured activity of a source without backing material to that of the same source with backing material is the backscatter parameter (F_{bs}). The amount of reflection depends on both the atomic mass of the backing material and the beta energy. For the 1945-1947 vegetation pellets, the backscattering medium is the vegetation itself because the mass of the vegetation is large. The F_{bs} is discussed in detail in Subsection 4.3.

The cellophane-scatter parameter (F_{cel}) accounts for an increase in the counting rate from the cellophane that was placed directly on top of the vegetation pellets and deflected additional betas into the GM detector. The F_{cel} is discussed in detail in Subsection 4.5.

The point-source geometry parameter (G_p) compensates for count losses that occurred because some of the beta particles emitted did not reach the GM detector's sensitive volume. The greater the distance from the detector's window, the more correction is required. This parameter is independent of beta energy. The G_p is discussed in detail in Subsection 4.6.1.

The sample-diameter parameter (F_d) compensates for the change in geometry caused by the spreading out of activity, such that the source (vegetation pellet) does not approximate a point source. That is, it compensates for horizontal displacement. The F_d is discussed in detail in Subsection 4.6.2.

The GM tube detection parameter (E_c) accounts for the fraction of betas and gammas counted once inside the GM tube, and for the coincidence/dead-time losses. E_c is discussed in detail in Subsection 4.7.2.

4.2 Self-Absorption Parameter, F_{sa}

The self-absorption parameter (F_{sa}) accounts for absorption of the beta particles in the vegetation pellet itself. It can be approximated by the equation of Schwendiman (1954)

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

where F_{sa} = fraction of iodine-131 betas not absorbed by passing through the vegetation pellet.

m = beta absorption coefficient, $0.0365 \text{ cm}^2/\text{mg}$ (Baltakmens 1977)

x = mass thickness of vegetation which is determined individually for each reconstructed pellet by dividing the weight (1000 mg) by the surface area (S) as computed by Equation (2.4). Note that the surface area also equals density times thickness.

In the vegetation pellet, self-absorption would have occurred along with self-scatter and backscatter (see Section 4.3). Both self-scatter and backscatter processes are highly complex in thick vegetation pellets. Equation (4.2) only accounts for self-absorption. This approximation is acceptable in this analysis because the pellets had large mass thicknesses of approximately $70 \text{ mg}/\text{cm}^2$ based on the reconstructed pellets made in 1989 and 1990, and the organic vegetation matter was of a low atomic number. Self-scatter tends to increase the counting rate by deflecting betas into the solid angle. The amount of increase in counting rate is related to mass thickness, beta particle energy, and atomic mass of the material (Nervik and Stevenson 1952; Schwendiman 1954). The lower the mass thickness and the higher the beta energy and the material's atomic number, the greater the increase in counting rate that can be expected. The effect of self-scatter and self-absorption are illustrated for a silver-iodide precipitate in Figure 4.1 (Schwendiman 1954). Starting in 1948, iodine-131 was chemically extracted from vegetation and precipitated as silver iodide. This technique replaced the routine method of counting beta activity in a vegetation pellet and assuming all counts originated from iodine-131 decay. In Figure 4.1, the dashed curve is the self-absorption parameter, F_{sa} , calculated from Equation (4.2). The dashed curve matches empirical results after the initial rise which occurs from self-scatter. The dark line shows the empirical results.

The silver-iodide precipitate had a much lower mass thickness than vegetation pellets and a greater atomic mass. Because of these differences, the values for self-absorption and self-scatter determined empirically at Hanford in the 1950s did not necessarily represent the values for vegetation pellets. This is true even though F_{sa} was presented as a function of mass thickness and the curve of F_{sa} versus mass thickness does approximate reasonably well the equation used in this analysis for estimating F_{sa} in vegetation pellets. Because the

S9109001.2

Figure 4.1. Self-Absorption, F_{sa} (Calculated from Equation 4.2) versus Mass Thickness (Schwendiman 1954).

empirically derived curve is only drawn to a mass thickness end point of 20 mg/cm², self-absorption for greater mass thicknesses can be extrapolated by extending the curve by using Equation (4.2) but may not be accurate.

Schwendiman (1954) determined empirically that the value of F_{sa} was approximately 0.72 for a mass thickness for AgI of 20 mg/cm². This value is supported by Thomas et al. (1956) who indicate an F_{sa} at 20 mg/cm² of around 0.73. These two reports also indicate only a slight increase in counting rate for an F_{sa} value greater than 1.0 because of self-scatter. From Equation (4.2) a value of 0.71 at 20 mg/cm² using Baltakmen's (1977) coefficient can be calculated. Therefore, it appears that, at least from 8 to 20 mg/cm², a rather close agreement exists between the empirical values for silver-iodide precipitates and calculated values.

The F_{sa} results for the reconstructed pellets are given in Appendix B (Tables B.3 and B.4). The average F_{sa} value for the 55 reconstructed hand-formed pellets is 0.35 with a minimum value of 0.28 and a maximum of 0.46. The average for the 55 reconstructed press-formed pellets is 0.31 with a minimum value of 0.24 and a maximum of 0.38. Interestingly, these values are very close to the historical self-absorption parameter of 3 or 1/0.33 empirically

determined from minimal data in 1946. The exact method for this empirical evaluation is not known (see Appendix A for more information).

4.3 Backscatter Parameter, F_{bs}

The backscatter parameter (F_{bs}) compensates for an increase in the counting rate from betas being reflected back towards the GM detector. The backscattering medium is typically the material upon which the source is mounted (e.g., the stainless steel planchet). The ratio of the measured activity of a source without backing material to that of the same source with backing material is the backscatter parameter (F_{bs}). The amount of reflection depends on both the atomic mass of the backing material and the beta energy. For the 1945-1947 vegetation pellets, the backscattering medium is the vegetation itself because the vegetation mass is large.

The amount of backscatter also depends to some extent on the counting geometry because scatter is not likely to be symmetrical and the probability of small-angle scatter is greater than that of large-angle scatter (Healy et al. 1950). At about one-third of the maximum beta range, a backscattering saturation thickness of the mount is obtained (Friedlander et al. 1981). That is, a further increase in the thickness of the backing material does not result in a corresponding increase in the measured counting rate. The maximum range for iodine-131 (considering only the 0.6-MeV beta activity) is about 220 mg/cm². One-third of this range is about 73 mg/cm² which is within the range of the mass thicknesses of reconstructed 1-gram sagebrush pellets. Therefore, it is assumed that saturation thickness is obtained within the pellet.

Backscatter in a radioactive vegetation pellet is a complex process that in reality should not be considered separately from other scatter components and self-absorption. Among the several kinds of scattering that would have occurred in addition to backscattering are self-scattering, scattering from the shield and shelf assembly, scattering from the cellophane covering, and even scattering out of the solid angle by the mica window. Although most of these forms of scatter are neglected, cellophane scattering is discussed in Subsection 4.5. Studies by Burt (1949) indicate that for iodine-131 the number of beta particles backscattered into the solid angle by air and the aluminum holder walls would be negligible. Therefore, this parameter will be ignored in the present analysis. Backscattering of incident betas off the mica window is likewise assumed to be negligible and is also ignored. In this approximation, the backscatter parameter is considered independently of these other processes.

The simplest backscatter parameter to define is that of the reflection of beta particles from a thin radioactive precipitate on an inert metal plate. However, no historical measurements of backscatter from a radioactive vegetation pellet were found.

If the thickness of the radioactive pellet is conceptualized as being divided into N increments, each increment serves as an absorber of the beta particles being emitted from the increment below, and that increment acts as a backscattering material for the beta particles being emitted from above. With each interaction, some attenuation of beta energy occurs with resultant changes in the probabilities of both absorption and scatter.

Although no historical empirical measurements were found that relate to the amount of backscattering from a thick vegetation pellet, backscatter parameters were evaluated for iodine-131 on filter paper which, like vegetation, has a low atomic mass. The empirical measurements made on filter paper are, therefore, used in this analysis to approximate the increase in activity expected as a result of backscattering of beta particles. It should be noted that the filter paper would not have approached the saturation mass thickness for iodine-131 for 0.608-MeV beta which is approximately 70 mg/cm^2 .

Table 4.2 lists backscatter parameters for iodine-131 with various types of backing, including filter paper. These parameters were reported by Burt (1949); Healy et al. (1950); Schwendiman (1954); and Thomas et al. (1956). Most of the backscatter parameters were determined for the first shelf of the GM detector. Only a few measurements were made on other shelves. Schwendiman (1954, Section 2.03) noted that the "results show a trend toward slightly lower factors with decreased geometry;" i.e., on lower shelves. However, a comparison of parameters indicates that this trend is minimal for shelves 1, 2, and 3. Burt (1949) found that iodine-131 tended to have backscatter parameters very close to or just slightly smaller than those for RaDEF standards. To aid in the evaluation, F_{bs} values for RaDEF are also provided in Table 4.2.

For this approximation, a backscatter parameter of 1.1 is assumed for every pellet, based on the measurements given in Table 4.2 and the low atomic mass of vegetation. That is, the backscatter parameter empirically determined for filter paper for iodine-131 is assumed to represent the backscatter expected from a thick vegetation pellet.

4.4 Cellophane, Air, and Mica-Window Absorption Parameter, F_a

The absorption parameter (F_a) compensates for the absorption of beta particles after they leave the vegetation pellet and pass through the cellophane, air, and mica window. This correction depends on beta particle energy, the combined mass thicknesses of cellophane, air, and the mica window, and the iodine-131 beta-absorption coefficient. As beta particles traverse any material, some will be absorbed or scattered by the material being traversed. The probability of absorption increases with mass thickness (mg/cm^2). In this analysis, the effects of additional scattering by the aluminum source holder, air, or the mica window are ignored. Also, the path length is assumed to be normal (i.e., perpendicular) to the detector. However, those betas emitted at angles other than normal would experience longer path lengths and, therefore, greater probabilities of absorption.

Table 4.2. Backscatter Parameters for Iodine-131 with Various Types of Backing

Radio-nuclides	Backing Material	Backing Depth (cm)	Distance ^(a) (cm)	F _{bs}	Reference
Iodine-131	Cardboard	max.	1.66	1.14	Burt 1949
Iodine-131	Whatman #40 filter paper	NG ^(b)	0.49	1.09	Schwendiman 1954
Iodine-131	Whatman #40 filter paper	NG	2.07	1.09	Schwendiman 1954
Iodine-131	H-70 filter paper	0.046	0.49	1.11	Schwendiman 1954
Iodine-131	H-70 filter paper	0.046	2.07	1.10	Schwendiman 1954
Iodine-131	H-70 filter paper	NG	0.49	1.11	Thomas et al. 1956
Iodine-131	H-70 filter paper	NG	2.07	1.07	Thomas et al. 1956
Iodine-131	Aluminum	max.	1.66	1.26	Burt 1949
Iodine-131	Aluminum	max.	0.49	1.19	Healy et al. 1950
Iodine-131	Aluminum	max.	0.49	1.24	Schwendiman 1954
Iodine-131	Aluminum	max.	2.07	1.26	Schwendiman 1954
RaDEF ^(c)	Filter paper	NG	1.66	1.10	Burt 1949
RaDEF	Cardboard	0.099	1.66	1.19	Burt 1949
RaDEF	Aluminum	0.079	1.66	1.29	Burt 1949
RaDEF	Whatman #40 filter paper	NG	0.49	1.08	Schwendiman 1954
RaDEF	H-70 filter paper	NG	0.49	1.09	Schwendiman 1954
RaDEF	H-70 filter paper	NG	0.49	1.18	Thomas et al. 1956
RaDEF	H-70 filter paper	NG	1.66	1.12	Thomas et al. 1956

(a) Distance is from the source to the counter window.
(b) NG = not given.
(c) RaDEF refers to bismuth-210 (RaE) because lead-210 (RaD) emitted a beta that was too weak to penetrate the cellophane covering, and polonium-210 (RaF) emitted only an alpha particle.

Cellophane, air, and the mica window are barriers that would absorb beta particles from the vegetation pellet. The total mass thickness of these barriers is the sum of their individual mass thicknesses. So the absorption parameter, F_a , is determined by the equation

$$F_a = e^{(-mx)} \quad (4.3)$$

where m = iodine-131 beta absorption coefficient, $0.0365 \text{ cm}^2/\text{mg}$ (Baltakmens 1977)

x = mass thickness (mg/cm^2).

For the 1940s GM detector system, this equation can be represented by

$$F_a = e^{[-m (M_c + M_a + M_w)]}$$

where M_c = mass thickness of cellophane, $\sim 3.1 \text{ mg}/\text{cm}^2$ (Healy et al. 1950; Schwendiman 1954).

M_a = mass thickness of the air; i.e., the distance from the top of the pellet to the detector window multiplied by the density of air ($\sim 1.2 \text{ mg}/\text{cm}^3$ is representative of Hanford air density).

M_w = average mass thickness of mica window, $\sim 3.7 \text{ mg}/\text{cm}^2$ in this analysis. This is based on the average mass thickness of the 1950s GM detector's mica windows (see Table 4.3). The Health Instruments group did have a goal of developing thinner windows, indicating the earlier mica windows may have been thicker.

Table 4.3 indicates the thickness of the mica windows varied among counters from about 2.4 to $4.6 \text{ mg}/\text{cm}^2$ with an average mass thickness of $3.7 \text{ mg}/\text{cm}^2$. The assumption is made in this analysis that the mica-window mass thicknesses for these 1950 detectors represent the mica-window mass thicknesses of detectors used from 1945 through 1947.

Thus, Equation (4.3) can further be represented by

$$F_a = e^{\{-0.0365 [6.78 + 1.21(2.07 - T)]\}}$$

where T = thickness of the pellet in cm

$2.07 - T$ = distance from the top of the pellet to the detector window in cm

Results for F_a for the 55 reconstructed hand-formed and 55 reconstructed press-formed pellets are given in Appendix B (Tables B.3 and B.4). The average F_a for both

Table 4.3. Mass Thickness of Thirteen Geiger-Mueller Detector Mica Windows (Healy et al. 1950)

Set. No.	Tube No.	Window Mass Thickness (mg/cm ²) ^(a)
1	158	4.5
2	164	4.6
3	139	2.9
4	119	2.9
5	136	3.4
6	185	3.0
7	128	4.1
8	117	4.6
9	103	3.2
10	168	4.5
11	155	4.3
12	161	3.4
13	5A-60	2.4
(a) Average = 3.7 mg/cm ² .		

hand-formed pellets (with a minimum of 0.72 and a maximum of 0.74) and for press-formed pellets (with a minimum of 0.73 and a maximum of 0.74) is 0.73.

4.5 Cellophane-Scatter Parameter, F_{cel}

- 18 Historically, cellophane was placed directly on top of both hand- and press-formed pellets when counting. *Studies done by Schwendiman (1954) indicate that this physical arrangement increased the counting rate of iodine-131 because the cellophane would have scattered some beta particles that were initially moving away from the detector and deflected them toward the detector (Parker 1952; Schwendiman 1954).* Schwendiman (1954) found that the increase in counting rate, resulting when cellophane was placed directly on a RaDEF source on the first

shelf rather than close to the window, was a factor of 1.06. With the RaDEF source on the second shelf, he found the factor to be about 1.07.

Nervik and Stevenson (1952) found that generally the greater the beta energy, the greater the self-scattering component. Based on their data, for a cellophane mass thickness of 3.1 mg/cm^2 and the maximum beta energy for iodine-131 ($\sim 0.6 \text{ MeV}$), the increase from self-scattering is estimated to be a factor of 1.05. Because the bismuth-210 self-scatter component indicated by Nervik and Stevenson is slightly higher than the increases measured by Schwendiman (1954) for cellophane scattering, the iodine-131 self-scatter component measured by Nervik and Stevenson is presumed to be also higher than the cellophane scattering that would have occurred for iodine-131 beta particles emerging from a vegetation pellet. Therefore, an F_{cel} factor of 1.04 is approximated for iodine-131.

The beta spectrum emerging from the top of a pellet would be attenuated by scattering and other processes occurring within the pellet. Thus, even if the F_{cel} for iodine-131 had been measured directly by Schwendiman, it might not represent the value for iodine-131 beta particles emerging from the top of a thick pellet.

4.6 Point-Source Geometry Parameter, G_p , and Sample-Diameter Parameter, F_d

Both the point-source geometry parameter (G_p) and sample-diameter parameter (F_d) relate to the physical geometry of the vegetation pellet and to the GM detector system. Therefore, both of these parameters are considered in this section. In addition, the calculation of G_p and F_d both require an additional parameter called the mean effective distance (MED) discussed in Section 4.6.3.

The point-source geometry parameter, G_p , accounts for the fraction of beta particles that would reach the sensitive volume of the GM detector, assuming a point source at a given distance, with no self-scatter; no self-absorption; no absorption in air, cellophane, or window; and no backscattering from the sample or mount (Schwendiman 1954). The G_p can be defined as the ratio of the solid angle that is subtended by the aperture of the sensitive volume of the GM detector tube to the total (4π) solid angle (Healy et al. 1950).

The sample-diameter parameter (F_d) accounts for the difference between the counting rate of a spread source and that of a point source of the same activity with both centered directly under the detector. A spread source will always produce a lower count rate than a point source of equal activity. Schwendiman (1954) defined F_d as the ratio of the counting rate of a

uniform circular source of a given diameter to the counting rate of the same quantity of radioactive material mounted as a point source.

Historically, values for the G_p and F_d parameters were determined empirically for thin sources. However, because the pellets were considerably thicker than those thin sources, a problem arises in using the empirically determined values for G_p and F_d . The values for these geometry parameters depend on distance. Even a small change in distance can greatly affect the values. With a thick pellet, individual G_p and F_d parameters are needed for a range of distances. Ideally, a complex integration should be performed over the thickness of the pellet. However, to simplify the calculations and uncertainty analysis, a mean effective distance (MED) is calculated and used as the distance parameter in the equations for G_p and F_d (Equations 4.4 and 4.5) to compensate for the pellet thickness.

4.6.1 Determination of G_p

Historically, G_p was determined for each shelf distance, as reproduced in Table 4.4. These historical determinations are used to derive the reconstructed point source geometry parameter.

Table 4.4. Historical Values of the Point-Source Geometry Parameter, G_p , Empirically Determined for Each Shelf Distance^(a)

Shelf	Distance (cm)	G_p
1	0.49	0.22
2	2.07	0.07
3	3.66	0.03
4	5.25	0.02
5	6.84	0.01
(a) Schwendiman (1954) and Norton et al. (1956)—both documents contain identical G_p values.		

To approximate the values in Table 4.4 so G_p could be rapidly calculated, Gilbert et al. (1992, p.3.1) developed the equation

$$G_p = 10^{[0.647 \times \log(1/MED - 0.185) - 0.8381]} \quad (4.4)$$

where MED = 2.07 - fT
 f = 0.70 for hand-formed pellets
 = 0.72 for press-formed pellets
 T = pellet thickness

The fractions 0.70 and 0.72 were determined using the method in Section 4.6.3.

Gilbert et al. (1992) discuss the basis for Equation (4.4) and the uncertainties in the approximation of G_p and F_d .

Because G_p was determined empirically, it actually accounts for the sensitive volume of the detector. The physical dimensions of the detector do not necessarily correspond to the sensitive volume because of dead space behind the mica window.

4.6.2 Determination of F_d

The value of F_d depends on the distance from the sample source to the detector, the source (pellet) diameter, and the detector diameter.

Historically, F_d was determined for diameters of 2.54 cm (1 in.), 3.81 cm (1.5 in.), and 5.08 cm (2 in.) and for the distances between each of the five shelves and the mica window. The historical values are reproduced in Table 4.5. These F_d values were determined empirically for sources with maximum beta energies greater than 0.3 MeV and for the GM detector described in Subsection 2.2.

A curve-fit equation was developed by Gilbert et al. (1992) to fit the empirically determined data given in Table 4.5 for distances less than 2.07 cm (0.81 in.):

$$F_d = 1 - (-0.4136 + 1.565 \log \text{DIAM}) 10^{-0.1311 \times \text{MED}} \quad (4.5)$$

which is appropriate

where DIAM = average diameter of the vegetation pellets
 MED = (see definition following Equation 4.4)

Table 4.5. Historical Values of the Sample-Diameter Parameter, F_d

Shelf	Distance ^(a) (cm)	Pellet Diameter			Reference ^(b)
		2.54 cm (1.0 in.)	3.81 cm (1.5 in.)	5.08 cm (2.0 in.)	
1	0.485	0.83	0.60	0.39	Healy et al. 1950
1	0.485	0.84	0.60	0.39	Schwendiman 1954
1	0.485	0.81	0.56	--	Thomas et al. 1956
2	2.07	0.87	0.82	0.67	Healy et al. 1950
2	2.07	0.87	0.75	0.62	Schwendiman 1954
2	2.07	0.88	0.73	--	Thomas et al. 1956
3	3.66	0.91	0.85	0.77	Healy et al. 1950
3	3.66	0.94	0.86	0.76	Schwendiman 1954
3	3.66	0.93	0.83	--	Thomas et al. 1956
4	5.25	0.95	0.91	0.86	Schwendiman 1954
4	5.25	--	--	--	Thomas et al. 1956
5	6.84	0.96	0.92	0.89	Healy et al. 1950
5	6.84	0.96	0.94	0.90	Schwendiman 1954
5	6.84	--	--	--	Thomas et al. 1956

(a) Distance refers to the distance from the shelf to the mica window.
(b) The 1950 values were specifically determined for iodine-131 sources. The 1954 and 1956 values are listed for beta-emitters with maximum beta energies greater than 0.3 MeV. The values in Thomas et al. (1956) are considered the least reliable because they are for the application of a silver-iodide precipitate distributed on a backing of the diameter listed in the table. Undoubtedly, the silver-iodide precipitate would have a smaller diameter than that of its mount.

4.6.3 Determination of Mean Effective Distance, MED

The geometry parameters (G_p and F_d) generated in the 1950s were for thin precipitates rather than thick pellets. Because the geometry parameters depend on distance from the detector, the values for these parameters actually vary throughout the thickness of the pellets. To compensate for the effect of pellet thickness, a mean effective distance (MED) from the detector is calculated, taking into account the thickness of the pellet as well as the distance from the top of the pellet to the detector window.

The MED should be calculated such that the G_p and F_d values are representative of the entire vegetation pellet, taking into consideration the decreasing proportion of iodine-131 beta particles being emitted from each pellet layer because of varying amounts of absorption within the pellet. Otherwise, the midpoint of the pellet would be the best estimate of MED, whereas the MED is at some point between the midpoint of the pellet and the top. The changing proportion of absorption that occurs with increasing pellet thickness weights the distance from which G_p and F_d are interpolated.

The value of MED was determined using a three-step procedure. First, the measurement conversion factor (M) was approximated by dividing each pellet into 10 layers, calculating M for each of the 10 incremental pellet thicknesses, and taking the average of the 10 values of M :

$$\bar{M} = \frac{1}{10} \sum_{i=1}^{10} \left[F_a F_{bs} F_{cel} E_c G_{pi} F_{di} \exp(-mx_i) \right]^{-1}$$

All of the parameters, except the exponential, are as defined immediately following Equation (4.1). The parameters G_{pi} , F_{di} , and $\exp(-mx_i)$ are the values of G_p , F_d , and F_{sa} , respectively, determined for the i th incremental pellet thickness. The factor $\exp(-mx_i)$ is the weight that is applied to G_{pi} and F_{di} for the i th incremental mass thickness x_i , where

m = beta-absorption coefficient, $0.0365 \text{ cm}^2/\text{mg}$ (Baltakmens 1977)

x_i = mass thickness of the i^{th} incremental thickness of the vegetation pellet.

The factor $\exp(-mx_i)$ is calculated at the center of the i th incremental pellet thickness to approximate the mean absorption for that thickness.

The second step in determining MED is to calculate M using Equation (4.1) for different values of MED in Equations (4.4) and (4.5). The third and last step is to compare $\bar{M}$ computed using the above equation with the values of M computed in Step 2. The best estimate of MED is that value of MED in Step 2 that gave a value of M closest to $\bar{M}$.

Using the model $MED = 2.07 - fT$, where T is known (see Tables B.1 and B.2 in Appendix B) and MED is the best estimate of MED, the value of f was determined to be 0.70 for hand-formed pellets and 0.72 for press-formed pellets.

The approximation of M could be improved by dividing the pellet into a greater number of layers and by considering self-scatter and variation in path length with changing solid angles.

4.6.4 G_p and F_d Results

The G_p and F_d values for the 55 hand-formed and 55 press-formed pellets computed by the method in Subsection 4.6.3 are found in Appendix B (Tables B.3 and B.4). The average G_p value for the 55 reconstructed hand-formed pellets is 0.082 and the F_d is 0.66; the average G_p value for the 55 reconstructed press-formed pellets is 0.081 and the F_d is 0.70.

4.7 Parameters with Negligible or No Impact

This subsection discusses two potential areas of bias that were evaluated and found to have negligible or no impact. Consequently, these parameters do not affect the value for the reconstructed M . The two areas of bias are 1) handling/processing losses, and 2) the GM tube detection parameter, E_c .

4.7.1 Handling/Processing Losses

This potential source of bias represents iodine-131 losses that might have occurred as a result of the collection, transportation, and handling of the vegetation samples. It could also include losses from compressing vegetation into a pellet.

19 Veteran Hanford employees believe that any losses from this source were probably minimal (Denham et al. 1988) because of the difficulty personnel had in finding a successful method to even chemically extract iodine-131 from vegetation. Several attempts were made to remove iodine from vegetation by harsh chemical-extraction procedures before a successful method was discovered in 1948. The early, harsh attempts produced only low yields. One reason for this may have been that iodine-131 entered the stomata of *the leaves by deposition on the plant*, precluding easy chemical separation.^(a) Also, even after severe rainfalls, the iodine-131 was found still tightly bound to the vegetation.

The veteran Hanford employees also felt that the lack of contamination on leather gloves used for collecting data and the lack of incidence of contamination on vehicles and in the laboratory provided more evidence that the iodine-131 had sorbed tenaciously to vegetation (Denham et al. 1988). In addition, Wolff (1951) assigns a value of 1.0 for "vegetation collection efficiency" which supports the above conclusion. Presumably, if removal of iodine-131 had been found from handling losses, the value would have been less than 1.0 to compensate for such losses.

(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.

Because of the evidence cited above, no loss of iodine-131 from handling or processing is assumed.

4.7.2 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 and other instrument effects are assumed not to impact the value for the reconstructed measurement conversion factor, M .

The GM tube detection parameter, E_c , is defined by the equation

$$E_c = C_d (E_\beta + E_\gamma) \quad (4.6)$$

where C_d = coincidence and dead-time parameter

E_β = beta-detection parameter

E_γ = gamma-detection parameter

4.7.2.1 Coincidence and Dead-Time Parameter, C_d

This parameter considers counts that were lost because a beta particle entered the GM tube's sensitive volume while it was recovering from a previous pulse (dead-time) or coincidentally with another beta.

Because nothing has been found in the literature surveyed that suggests corrections were made for coincidence/dead-time losses, it is assumed that prior to 1950 such corrections were not made. However, at least by 1950, any sample with a count rate of 5000 cpm (50,000 dpm, or 0.02 $\mu\text{Ci/g}$, assuming 10 percent efficiency) was moved to a lower shelf of the GM counter to reduce the count rate. By 1950, no specific determination for coincidence losses had been made for the mica-window GM detectors used at Hanford, although in general a 0.2-percent loss per thousand cpm was assumed (Healy et al. 1950). In 1951, coincidence losses in five systems were studied and found to vary from 0.5 percent per 1000 cpm to 1.5 percent per 1000 cpm with an average of 1.0 percent per 1000 cpm (Parker 1951). Schwendiman (1954) supplies an equation that approximates the effect:

$$\text{True cpm} = \frac{\text{cpm observed}}{1 - (\tau \times \text{cpm observed})} \quad (4.7)$$

where τ = counter-resolving time.

Schwendiman (1954) also supplies a table that lists the ratio of observed counts per minute to true counts per minute as a function of counting rate. The table is reproduced in Table 4.6 with corresponding coincidence/dead-time losses.

20 *Few environmental samples during this time period had counts high enough to result in significant coincidence losses. While the releases during this early time period were considered high by today's standards, the observed activities on off-site vegetation were generally less than a few hundred cpm. The highest year (1945) had counts near 1000 cpm/g, but values were generally much less than 100 cpm for 1946 and 1947. The bias from neglecting coincidence/dead-time losses is, therefore, assumed to have been minimal, and no count rate losses from this phenomenon are assumed. C_d is assigned a value of 1 for inclusion in Equation (4.6).*

4.7.2.2 Beta-Detection Parameter, E_β

This parameter considers those betas that entered the sensitive volume of a GM tube but did not generate counts. For any charged particle that enters the sensitive volume of a GM tube, the counting effectiveness is essentially 100 percent (Knoll 1979). However, a small, dead space often occurs just behind the entrance to the mica window in which the local electric field may be too small to prevent recombination or electron loss. In addition, a small amount of beta activity is not counted because the particles interact too far into the walls of the GM counter to liberate any electrons in the GM tube's sensitive volume.

In this analysis, the inference is made that the dead space behind the mica window was compensated for by empirical determinations of G_p . Because the beta-counting effectiveness once inside the sensitive volume is essentially 100 percent, this parameter is assigned a value of 1 for inclusion in Equation (4.6).

Table 4.6. Ratio of Observed Counts per Minute to True Counts per Minute as a Function of Count Rate (Schwendiman 1954)

Observed Count Rate (cpm)	Ratio of Observed to True Activity	Coincidence/Dead-Time Losses
0-1000	1.00	0.00
1000-2000	0.99	0.01
2000-3000	0.98	0.02
3000-4000	0.97	0.03
4000-5000	0.96	0.04
5000-6000	0.95	0.05

4.7.2.3 Gamma-Detection Parameter, E_γ

Iodine-131 emits betas and gammas simultaneously, both of which can be detected (i.e., result in counts) by the GM detector. Because the source of counts (whether from betas or gammas) collected by a GM detector cannot be identified, counts originating from gammas would erroneously be interpreted as beta counts. While gamma detection by the GM detectors cannot be ruled out, Healy et al. (1950, p. 36) indicate that, in general, gamma contributions in the small mica-window GM detectors were minimal. They state that

For most of the routine work no correction is made for the contribution to the counting rate caused by gamma radiation, because of the low efficiency of the particular type of counters used. This in general means that the rate for the routine samples will be higher than the true rate and will therefore serve as a safety factor in health measurements. For accurate work however, it is necessary to obtain a value for the beta radiation only. This is generally accomplished by the use of an aluminum absorber of such thickness as to allow complete absorption of the beta particles but minimum absorption of the gamma photons.

Therefore, because no quantitative indications of the gamma contributions from the decay of iodine-131 to the beta analyses of vegetation pellets using GM detectors have been found in the historical literature surveyed, E_γ is assigned a value of zero in Equation (4.6).

4.8 Limitations

The parameters in the equation for M must be considered only approximations and interpreted with an understanding of the limitations of the methods by which they were determined. (See Appendix A for a discussion of the historical methods used to derive the parameters in M .)

One broad method of refining the estimates of M would be to build a mathematical model in which all of the processes of beta interactions would be incorporated along with the pellet and detector geometries. Such a model would require a complex series of equations and would also have limitations. Some items that would have to be considered in building the model include:

1. probable dimensions of the GM detector's sensitive volume

2. probability of backscatter, self-scatter, and absorption based on beta energy (considering the beta spectrum, incident angle, and location of emission with the pellet)
3. effects occurring in each pellet layer or increment and corresponding effects in adjoining layers
4. solid-angle relationships in each pellet layer.

In addition, not all of the notebooks or historical documents have been comprehensively reviewed with respect to parameters that affect M . A comprehensive review of all available historical documents could yield more information to be used in refining M .

4.8.1 Limitations in F_{sa} and F_{bs}

Ideally, self-absorption (F_{sa}) and scatter within the pellet (such as F_{bs}) would not be considered independently because the interrelationship between these processes is very complicated, particularly in a thick vegetation pellet (Aten 1950). Scattering in air or from the aluminum source holder was not evaluated.

4.8.2 Limitations in F_a and F_{cel}

The beta absorption coefficient used in computing F_a assumes an unattenuated iodine-131 beta spectrum. However, the actual iodine-131 beta spectrum that emerges from a pellet would be of lower energy overall because of interactions with the vegetation pellet. Therefore, a consideration of the lower-energy iodine-131 spectrum in the computation of F_a could help refine this estimate.

Because path lengths differ depending on the angle of the particle incident to the detector window, the actual mass thickness is slightly greater than the mass thickness for a beta particle traveling perpendicular to the GM counter. However, given the low density of both the air and the mica window, this difference would be negligible and is ignored in this analysis.

The cellophane-scattering parameter (F_{cel}) for iodine-131 was never directly measured for the detector systems used in the 1940s. However, the measurements of F_{cel} made with a USBS RaDEF standard by Schwendiman (1954) and other information provided by Nervik and Stevenson (1952) indicate the expected upper limit. An iodine-131 beta spectrum that had already traversed the pellet would be more attenuated than an iodine-131 spectrum from a sample with low mass thickness. Consequently, the actual energy of the beta particles subject to cellophane scattering would be lower.

4.8.3 Limitations in G_p and F_d

Ideally, M would be computed by dividing the pellet into a very large number of increments for the determination of G_p , F_d , and F_{sa} . In addition, variations in path length that would occur as the solid angle changed with distance were not considered.

4.8.4 Limitation in E_c

The assumption that the E_γ component of E_c equals 0 is only an approximation. Some gammas, albeit few, would have been detected.

5.0 D_e : Decay Correction Factor

Iodine-131 has a half-life of 8.05 days. As a result, iodine-131 activity in vegetation samples would decrease steadily in the interval between sampling and counting. After about 8 days, the activity would be half the original. This section discusses the need to correct data for losses in activity caused by iodine-131 decay that would have occurred during the time interval (t) between sample collection and counting.

All original counting rate data (cpm/g) for which the specific time interval (t) is known should be corrected individually. From July 1946 on, all reported activities ($\mu\text{Ci/kg}$) are assumed to have been decay-corrected. However, if reported activity data for the period before July 1946 is used, it should be corrected for decay assuming an average decay time. It should be noted that the uncertainty in the decay correction factor for reported activities prior to July 1946 will be greater than for the following period.

Herde (1946) indicates that the method for correcting counts for decay was known in 1945. However, the conversions of counts per minute (cpm) to microcuries per gram ($\mu\text{Ci/g}$) shown on 1945 and 1946 count sheets indicate that decay corrections were not made for measurements before February 1946. After February 1946, the data sheets found do not contain both cpm/g and corresponding $\mu\text{Ci/kg}$. The first mention of the use of iodine-131 decay corrections in the historical literature is by Turner (1946) in reference to vegetation measurements for July 2 through August 31, 1946. Subsequent periodic reports clearly indicate that decay corrections were conducted routinely after that time. The average activities ($\mu\text{Ci/kg}$) reported after June 1946 are, therefore, assumed to have already been corrected for decay.

The decay correction factor D_e can be estimated by the equation

$$D_e = e^{\lambda t} \quad (5.1)$$

where D_e = ratio of the amount of activity in the sample when collected to the activity in the sample when counted

λ = decay constant; for iodine-131, $\lambda = (\ln 2)/8.05$ days or 0.086/day

t = time interval in days between sample collection and counting.

In addition to iodine-131, the vegetation pellets also contained longer-lived radionuclides, both from naturally occurring sources and from other fission products. With longer intervals between collection and counting, the percentage of these other, longer-lived radionuclides would increase. The estimated iodine-131 correction factor (see Section 6.0) compensates for other radionuclides and eliminates the bias (i.e., the overestimation of the amount of activity) caused by decay-correcting the total activity by the iodine-131 half-life. Such a bias would occur because the decay constant for longer-lived fission products is much smaller than that of iodine-131.

If the decay interval for a given sample is not known, a best estimate of the interval can be calculated by evaluating the interval for other measurements made the same day or the day before or after. In some cases, the average decay interval for the given week may be the most appropriate estimate. For specific cases, the method of obtaining a best estimate of the decay interval must be determined individually based on the information available.

Decay correction factors are given in one-day increments in Table 5.1. These are factors by which the observed data must be multiplied to obtain the activity level at the time of sampling.

Table 5.1. Decay Correction Factors for Iodine-131

Decay Interval (t) (days)	Decay Correction Factor (D_t)
1.0	1.09
2.0	1.19
3.0	1.29
4.0	1.41
5.0	1.54
6.0	1.68
7.0	1.83

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

This section provides estimates of the I_{cf} correction factors that compensate for the bias that occurred by assuming the total beta counts on vegetation reflected the presence of iodine-131 only. The goal is to approximate I_{cf} as a function of fuel-cooling time. BNW staff approached evaluating I_{cf} as a function of fuel-cooling time by reviewing empirical monitoring data and computer-generated fuel-inventory data. Both of these data sets contain information on the fraction of iodine-131 activity to total activity on a sample of vegetation. Both modeled and empirical data are then correlated with fuel-cooling times. The parameter I_{cf} is defined to be:

$$I_{cf} = 1 - (FP/T) \quad (6.1)$$

where I_{cf} = Fraction of activity resulting from iodine-131 decay
FP = activity of fission products other than iodine-131
T = total activity (iodine-131 + FP).

The quantity FP/T is approximated on the basis of a comparison of historical counting data and computer-modeled fuel-inventory data correlated with fuel-cooling time.

From 1945 through 1947, the observed gross beta activity on vegetation pellets was assumed to reflect the presence of iodine-131 only (Healy 1948a), even though beta radiation from other fission products and from naturally occurring radionuclides could also have contributed to the observed activity levels. This assumption was acceptable initially for samples where the observed counting rates were at least two standard deviations above the naturally occurring background counting rate, given that iodine-131 was by far the most predominant radionuclide deposited on vegetation. This is indicated by decay measurements of onsite samples (see Subsection 6.1.1 for details). Later, when iodine-131 was first chemically separated from vegetation before beta counting, the bias caused by the assumption that all beta counts represented iodine-131 was nearly eliminated. This separation technique became the sole routine analytical method beginning in December 1948.

Although the levels of naturally occurring radionuclides in vegetation varied only slightly (seasonally and with plant species), I_{cf} as defined by Equation (6.1) is believed to have varied widely according to where the depositions occurred and the time interval between deposition and collection as well as fuel-cooling time prior to processing. The contribution from iodine-131 to the total activity on vegetation could have changed with a number of variables such as

fuel-cooling time before dissolution, fuel-specific power history,^(a) dissolution process, meteorology, and time elapsed after deposition. In addition, the activity measured in a given sample may have resulted from deposition from multiple dissolution campaigns. The number of these variables makes it difficult to estimate a generic correction factor for the iodine-131 assumption for a specified time period or location. Because of weather variations, weathering (i.e., removal of iodine-131 from vegetation by natural weathering processes such as wind or rain), and the short half-life of iodine-131, the true fraction of iodine-131 on vegetation on any given date and location could approach zero and, correspondingly, I_{cf} could approach zero as well. In addition to the large number of variables affecting I_{cf} , there are few historical vegetation-monitoring data, particularly for samples collected offsite, from which I_{cf} can be estimated. For this reason, computer-modeled data were also used to estimate I_{cf} (see Subsection 6.3 for a description of the computer-modeled data).

The application of I_{cf} to a given vegetation sample requires knowledge of the probable fuel-cooling times for the multiple production campaigns that would have resulted in deposition on a given vegetation sample. This requires a correlation of dissolution with meteorology and knowledge of decay times since deposition. Thus, accurate application is not straightforward. The uncertainty analyses conducted by Gilbert et al. (1992) investigate the effect of this uncertainty in I_{cf} on the uncertainty of the reconstructed total conversion factor (C).

Subsection 6.1 discusses radionuclides other than iodine-131 that may have contributed to the observed gross beta activity on vegetation. These other radionuclides include naturally occurring potassium-40 and uranium decay-chain emitters, and longer-lived fission products, including the particulates that were released because of a stack ductwork corrosion problem that occurred in 1947. Subsection 6.2 covers the historical data from which contributions from other fission products can be estimated. Subsection 6.3 describes the method used to approximate the correction factors for the iodine-131 assumption.

6.1 Other Radionuclides

The vegetation present in the Hanford area from 1945 through 1947 contained naturally occurring radionuclides as well as any iodine-131 and other longer-lived fission products that resulted from fuel dissolution at Hanford. Some of these fission products were radioactive particulates that came from corroded stack ductwork that flaked off in 1947. This singular occurrence was corrected shortly after discovery. In the 1950s, above-ground testing of nuclear weapons at times also contributed to the activity detected on vegetation.

(a) Fuel burnup in units of megawatt days per short ton (MWd/ST).

6.1.1 Natural Background Radioactive Materials

The only important radionuclide occurring naturally in vegetation in this analysis is potassium-40. The potassium-40 content of vegetation was analyzed extensively in the late 1940s (South 1949). In one document, a value as high as 50 pCi/g was given (Singlevich 1948b). The concentration levels were found to vary with plant moisture and the season, peaking in late April and May (Paas and Singlevich 1951). More recent data on potassium-40 levels are found in the 1973 and 1974 Hanford Environmental Monitoring Reports (Nees and Corley 1974; Fix 1975). These reports indicate that 10 pCi/g is a good estimate of average potassium-40 levels in vegetation native to the Hanford area. Singlevich (1948b) summarizes research on potassium-40 content in sagebrush. The average concentration of potassium-40 was about 3.5 cpm/g.

In their report for the week ending December 13, 1945, Parker and Gamertsfelder (1945) indicate that noncontaminated vegetation obtained from Portland, Oregon, and from Berkeley, California, also produced counting rates above instrument background. Therefore, even noncontaminated Hanford vegetation would have produced a certain count rate (although an insignificant one) with the counting systems in use in the 1940s, and this count rate presumably would have come from potassium-40. The count rate from naturally occurring potassium-40 is not high enough to warrant a special correction factor. The error resulting from failure to compensate for potassium-40 is minimal and causes a bias in the conservative direction; i.e., an overestimate of true activity, albeit a minimal one.

Other radionuclides occurring naturally in vegetation include radium-224 (<0.04 pCi/g), radium-226 (<0.11 pCi/g), and total uranium (<0.55 pCi/g) (Fix 1975). The average activities from these components are even lower than those from potassium-40. Therefore, they do not warrant special correction factors.

6.1.2 Longer-Lived Fission Products

From the start of operations, in addition to iodine-131, longer-lived fission products were also emitted from the exhaust stacks of the Hanford fuel-dissolution plants, mostly as aerosols. Trace amounts of fission products other than iodine-131 were detected on desert vegetation at an early date (Parker 1948). Although each element would have a unique dispersion pattern based on chemical interactions in the environment, in this analysis the longer-lived fission products (both droplets and aerosols) are assumed to have dispersed similarly to iodine-131. The dispersion and deposition of these droplets and aerosols were casually tracked by analyses of vegetation and air (Parker 1948). Parker observed that the percentage of longer-lived activity to iodine-131 found in the samples was increasing steadily with time.

The historical record contains few measurements from which levels of other fission products can be compared with iodine-131 levels on vegetation in the period from 1945 through 1947. Parker (1948) remarks on the meagerness of this type of data in a review of longer-lived fission product activity. As a result, only limited historical information is available to estimate FP/T.

The fission products that would have been released from the stacks were somewhat dependent on the actual dissolution chemistry. The principal longer-lived fission products that would have been counted in the measurement of pellets include isotopes of ruthenium, cerium, yttrium, cobalt, strontium, and cesium (Napier 1991). Most of the other fission products were generated in minimal quantities or were of limited volatility which precluded significant release through the stacks. Noble gases are highly volatile and were released, but because of their inert chemical characteristics, they would not have been deposited or retained on vegetation.

6.1.3 Radioactive Particulates from Corroded Ductwork

In 1947, a unique problem occurred with fission products that had adhered to corroded stack ductwork and subsequently flaked off (Parker 1948). Replacement of the corroded ductwork in 1948 eliminated contamination by the larger particulates but had little influence on the emission of smaller particulates (droplets), confirming that the primary source of small particulates was directly from fuel dissolution.

The radioactive particulate problem resulting from corroded iron ductwork was discovered in September 1947 because of recurrent contamination, presumably of personnel (Parker 1948). It was estimated that the problem started around April 1947 about six months prior to detection. However, a review of contamination data indicated the problem had rapidly increased in magnitude just prior to its discovery (Mickelson 1947; Parker 1948). By mid-October 1947, the general distribution pattern around T and B Plant stacks had been defined sufficiently to indicate these stacks were the source. By late November 1947, the particulates were found to have come from effluent fission products that had adhered to black iron in the stack fan ductwork and then started to flake off.

The visible particulates derived from the corroded ductwork ranged from about 0.02 to 1.5 mm (linear dimensions), and their activities ranged from about 0.1 to 3 μ Ci. The actual sizes ranged all the way down to ultra-fine particulates that were effectively aerosols. The particulates from the ductwork contained only long-lived emitters. The effective half-life was about 300 days. The activity was identified as representing cerium-144, yttrium-90, strontium-90, and some alpha activity from plutonium (Burger 1991).

Deposition velocities would have varied, depending on particulate size and shape, with larger particles being deposited closer to the stacks. The corroded duct particulates were mainly deposited close to the 200 Areas in which they were produced, trailing off in an ellipsoid pointing toward the southeast (Rees 1947; Zuerner 1947). Once deposited, some particulates may have been re-suspended and transported farther by wind. Analyses of variations in the samples, taken at 1-mile intervals about 2 to 5 miles from the stacks as well as within the 200 Areas, indicated that the deposition of long-lived fission products was more like a continuous distribution than a particulate one. General Electric's Health Instruments Department concluded that the main source of this continuously distributed long-lived activity was not stack corrosion particles and that the 1947 particulate releases represented only a fraction of the total long-lived fission product emissions from the stacks (Parker 1948). This conclusion was supported by the observation that numerous smaller particulates continued to be emitted even after the replacement of fans and ductwork in January 1948, although the replacement did eliminate the emissions of large corrosion particles. The smaller particulates were identified as coming from mists produced by boiling solutions and by the transfer of liquids using steam jetting (Burger 1991).

Because the duct particulates were generally large, they are assumed to have been only deposited close to the 200 Area stacks and, therefore, did not contribute to the gross beta activity of off-site vegetation collected in 1947. This assumption is supported by Parker (1948) where he concludes the lack of wide variation in counts of 1-gram vegetation pellets (even those from the plant environs) indicates minimal contributions. For these reasons, separate correction factors for samples with potential particulate depositions from corroded ductwork are not provided in this analysis.

6.2 Historical Data

With time, radionuclides other than iodine-131 began to contribute increasingly to the total observed activity on vegetation. Longer fuel-cooling times, accumulation of long-lived fission products both in the fuel and on vegetation, and iodine-131 emission control devices placed in the stacks in 1948 contributed to this trend. The best historical data from which to assess I_{cf} come from 1945 and early 1946 because decay data have been found in a laboratory notebook (Bulow 1945). Also, other documents discuss conclusions based on decay curves or an estimate of the ratio of iodine-131 to other fission products on vegetation (Parker 1948). The contributions from other radionuclides were rarely measured before chemical separation procedures were adopted in mid-1948, and the few activity or count data found for other radionuclides are from samples collected on the Hanford Site.

Basing I_{cf} on historical information has limitations. For instance, the number of dissolution campaigns that resulted in deposition on the collected samples and the time elapsed between deposition and sample counting is not known. The length of time elapsed would have affected the amount of activity resulting from iodine-131 relative to the activity of other fission products. With a long elapsed time, the overall activity would be expected to be relatively low, and with a long enough time, I_{cf} would approach zero. However, for samples collected from 1945 through 1947, a sample with undetectable iodine-131 levels would likewise be expected to have minimal or undetectable quantities of longer-lived fission products.

The value of FP/T indicated by historical measurements of samples collected on the Hanford Site may not correspond to the contribution at more distant off-site locations. Such a lack of correspondence could result from differences in how the components dispersed in the atmosphere, in the rates at which they were deposited on vegetation, in the rates at which the radionuclides were retained, and in the length of time elapsed from deposition to sample counting. The rate of deposition of iodine-131 relative to other species is complex. Burger (1991) discusses iodine behavior in the atmosphere and deposition; thus, this topic will not be addressed further here.

Another limitation is that BNW staff have not discovered the analytical methods used prior to 1948 to measure iodine-131 and other fission products separately. The historical record contains only limited information on these measurements: date of collection and counting and sometimes the sampling location. Consequently, the historical measurements must be interpreted in the light of these limitations.

Plotting the data obtained by periodically counting a given vegetation sample over several days or weeks can be used in conjunction with radionuclide half-life information to indicate which radionuclide predominates in a sample. Decay data on samples collected in 1945 indicate that iodine-131 was the predominant radionuclide (Bulow 1945; Parker and Gamertsfelder 1945; Healy and Turner 1946). Bulow (1945) reported counting data for two on-site samples collected approximately 4 and 5 miles southeast of the 200 East Area in July 1945 (see Table 6.1). The background-corrected counts per minute (cpm) given by Bulow (1945) for each day are compared with the cpm that would be expected if iodine-131 were the only radionuclide present. The expected cpm values were obtained by decay correcting the observed cpm taken on July 17, 1945 (i.e., Day 0) by the iodine-131 decay factor for each day thereafter. These results, given in Table 6.1, indicate that the observed activity from both samples was primarily from iodine-131.

Table 6.1. Decay Data for Two Samples, July 1945 (Bulow 1945)

Location ^(a)	Day	Observed cpm	Expected cpm from ¹³¹ I ^(b)
R4S-MP6	0	317	317
R4S-MP6	1	281	291
R4S-MP6	2	268	267
R4S-MP6	3	246	245
R4S-MP6	4	227	225
R4S-MP7	0	287	287
R4S-MP7	1	253	263
R4S-MP7	2	242	242
R4S-MP7	3	222	222
R4S-MP7	4	204	203

(a) Location: R4S refers to the main Hanford-Site highway diagonally southeast from the 200 East Area and designated as Route 4 South. MP6 and MP7 refer to Mileposts 6 and 7, respectively, from the 200 East Area.

(b) The expected cpm, assuming all of the counts per minute were from iodine-131, was computed by multiplying the observed "zero-day" cpm by $e^{-0.086t}$ for iodine-131.

Other reports also indicate that the activity was mainly iodine-131, again based on decay measurements. Parker and Gamertsfelder (1945) in their report for the week ending February 16, 1945, state that activity on a sagebrush sample decayed with a half-life of 8 days. Healy and Eisenacher (1946) indicate that a sagebrush sample collected November 26, 1945, and counted in January 1946 gave a value indicative of iodine-131 with its characteristic 8-day half-life. In addition, both Richland air and fuel-dissolution stack measurements made in March 1945 indicate that iodine-131 levels in the air were two to four orders of magnitude greater than the levels of long-lived fission products (Parker 1948). These results support the assumption that off-site iodine levels on vegetation in early 1945 were significantly higher than levels of long-lived fission products. Thus, the activity on vegetation in 1945 is assumed to have been predominantly from iodine-131.

In the historical literature surveyed, few documents provided actual measurements on activities of both long-lived fission products and iodine-131 for the same samples. One of

these documents (Healy 1948b) contained activity data as provided below in Table 6.2. The ratio of fission products to total activity was computed from these data and is provided in Table 6.2.

All of the data in Table 6.2 were for samples collected on the Hanford Site. The ratio of FP/T for off-site samples could have been different because of differences in the deposition patterns for iodine-131 and the other fission products. As discussed previously, no particulates from corroded ductwork are assumed in the relatively low activity for 1947, given that these particulates were so highly radioactive the count data would have been extremely high, and the problem did not escalate until about September 1947 (Parker 1948).

Table 6.2. Iodine-131 and Other Fission Product Activity Levels (Healy 1948b) and Computed Fission Products to Total Activity (FP/T)

Sampling Date	Location ^(a)	Average Fuel-Cooling Time ^(b) (Days)	Iodine-131 (pCi/g)	Other FP (pCi/g)	FP/T ^(c)
11/04/46	R4S-MP4	65	800	100	0.11
11/25/46	R4S-MP7	65	14000	700	0.05
11/26/46	R4S-MP2	65	13000	1900	0.13
4/29/47	Met	68	600	300	0.33
6/20/47	200-E	77	2600	2200	0.46
7/22/47	200-W	78	1100	700	0.39
7/14/47	200-W	78	3300	5000	0.60

(a) The respective locations are at Route 4 South, Mileposts 4, 7, and 2; the meteorological tower located between the 200 West and 200 East Areas; and within the 200 East and 200 West Areas, respectively.

(b) The average fuel-cooling time is calculated from the cooling times for all dissolution campaigns that occurred during the given month (see Table 6.4).

(c) The relationship between FP/T in this table with average fuel-cooling time for the given month is imprecise. This is because the actual fuel-cooling time for the campaign(s) that resulted in deposition on the collected samples is unknown. Also, the decay interval after deposition(s) and sample counting is unknown. As discussed previously, iodine-131 has a short half-life so the interval between deposition and counting affects the amount of iodine-131 on a sample.

Parker (1948) estimated the ratio of other fission products to total activity as given in Table 6.3. For location, he merely refers to "desert vegetation" which is presumably on site. He does not, however, describe the analytical process used.

6.3 I_{cf} Method

This section includes discussions of the approach (6.3.1), fuel-cooling times (6.3.2), and estimated FP/T values (6.3.3) based on computer-generated fuel burnup to estimate the FP/T values on vegetation in the Hanford environs as a function of fuel cooling time.

6.3.1 Approach

I_{cf} as defined by Equation (6.1) is believed to have varied over time and area. The contribution from iodine-131 to the total activity on vegetation could have changed with a number of variables such as the fuel-cooling time before dissolution, burnup, fuel-dissolution process, multiple fuel-dissolution campaigns, meteorology, and time elapsed after deposition. These variables make it difficult to estimate specific values for a given time period. In addition to the large number of variables affecting this ratio, there are few historical data from which FP/T values can be estimated, particularly for samples collected off site. For this reason, additional information is needed to derive estimated I_{cf} values.

Table 6.3. Estimated Fraction of Fission Products to Total Activity (FP/T) on Vegetation (Parker 1948)

Period Stated in Parker (1948)	Assumed Date	FP/T
After one year of operation	12/1945 ^(a)	0.00005
After two years of operation	12/1946 ^(b)	0.10
March 1948	3/1948	0.50 ^(c)
<p>(a) This date is the month/year assumed to be the one Parker is referring to by his description, "after one year of operation," which would have been about December 1945. Correlation of an average fuel-cooling time with his data is even more difficult as Parker only provided his conclusions for a period of time rather than on a specific date.</p> <p>(b) Again, as for the first date, Parker did not define operation, but provided the listed FP/T value "after two years of operation." However, because it is mentioned in the context of fuel-cooling time, it is, therefore, assumed to mean fuel-dissolution operations.</p> <p>(c) Parker notes that at the time of writing (March 1948), the longer-lived activity in the context of vegetation was comparable to the levels of iodine-131 activity. Hence, we have chosen a 50 percent value, although his statement may also be interpreted to mean only that the two levels were of the same order of magnitude.</p>		

This additional information has been provided by ORIGEN2 (Croff 1980),^(a) a computer code that calculates the fission product inventory in fuel based on burnup and length of time after removal of fuel from the reactor. This computer code provides the inventories of light activation products and fission products, and inventories of actinides at a fuel burnup of 200 MWd/ST, the target discharge burnup in the years prior to 1950. ORIGEN2 does not compute the expected fission product activity at the point of stack discharge resulting from fuel dissolution. To derive this latter value, release fractions must be applied to the inventories.

The ORIGEN2 code prints activities per short ton for three radionuclide categories: 1) light-element activation products, 2) actinides and their decay products, and 3) fission products. It also includes total activities in each category. The values generated for total activities for all light activation products and fission products, iodine-131, and the noble gases were used to derive an estimate of FP/T.

In this approximation, the approach used in estimating FP/T from ORIGEN2 and historical data as a function of fuel cooling time is to assume that the contribution that fission products other than iodine-131 made to the total activity (determined from measurements of on-site samples and determined from the ORIGEN2 run at the point of stack discharge) represents the contribution at all sampling locations.

6.3.2 Fuel-Cooling Times

Table 6.4 lists average monthly fuel-cooling times from Napier (1991). Longer fuel-cooling times resulted in more iodine-131 decay before emission, increasing FP/T and consequently decreasing I_{cf} . For example, after 30 days of cooling, fuel would contain approximately 8 percent of the original iodine-131 activity at reactor discharge. After 60 days it would contain 0.6 percent, and after 90 days it would contain only 0.04 percent. Longer-lived radionuclides would not be depleted as rapidly.

6.3.3 Estimated FP/T Based on ORIGEN2 Run

The data generated by the ORIGEN2 computer code include activities for total fission products as a function of fuel-cooling time and for a burnup of 200 MWd/ST. The ORIGEN2 data list activities per short ton (ST) for each radionuclide after cooling times ranging from 10 to 90 days in 20-day increments, at 150 days, and also at discharge. The ORIGEN2 output also includes total activities at each cooling time for each category. The

(a) ORIGEN2 data transferred from C. M. Heeb, BNW, to E. I. Mart, BNW Consultant, January 13, 1993.

Table 6.4. Average Number of Days Fuel Cooled before Dissolving (Napier 1991, p. 2.2)

Month/Year	Average Fuel-Cooling Time (Days)	Month/Year	Average Fuel-Cooling Time (Days)	Month/Year	Average Fuel-Cooling Time (Days)
12/44	49	1/46	62	1/47	62
1/45	39	2/46	67	2/47	69
2/45	53	3/46	70	3/47	69
3/45	52	4/46	62	4/47	68
4/45	42	5/46	62	5/47	68
5/45	36	6/46	73	6/47	77
6/45	35	7/46	68	7/47	78
7/45	40	8/46	63	8/47	82
8/45	39	9/46	59	9/47	84
9/45	38	10/46	65	10/47	90
10/45	44	11/46	65	11/47	95
11/45	52	12/46	62	12/47	95
12/45	47				

ORIGEN2 data only compute the inventory in the fuel, not at the point of discharge from the stacks. Release fractions must be applied to each element in the inventory to derive fission-product activities at point of stack discharge.

Release fractions resulting from fuel dissolution were obtained from Heeb (1993) for iodine-131 and from Napier (1992a) for all other fission products. A release fraction of 0.91 for iodine-131 was provided by Heeb (1993, p. 4.28). Napier (1992a, p. 5, 6) provides the following estimated release fractions for other fission products: 1.00 for noble gases, and 0.00001 for all other nonvolatile radionuclides and for ruthenium isotopes.^(a)

(a) Napier (1992a) revised the release fraction for ruthenium isotopes from Phase I estimates of 0.01 (Napier 1991). The Phase I estimates were given to compensate for an initial impression that large amounts of ruthenium were emitted in periodic (particulate) events. Because continued research indicated that ruthenium and other fission products were continuously emitted as mists, and the bulk of emissions were from these mists rather than particulates (see Section 6.1.3 for a discussion on the particulate releases), Napier reduced the estimated release fraction. Napier based the release fraction for all other fission products on an analysis of historical data for the 1945 through 1947 period found in Healy (1948a, 1948b), Parker (1948), Greager and MacReady (1948), and on information in documents covering later years.

The following equation is used to compute fission products other than iodine-131 at the point of stack discharge^(a):

$$FP = (Total - Kr - Xe - {}^{131}I) 0.00001 \quad (6.2)$$

where FP = fission and activation products other than actinides, iodine-131, and the noble gases at the point of stack discharge.

Total = total activity for all fission products as given in the ORIGEN2 run

Kr = activity for significant krypton isotopes^(b) as given in the ORIGEN2 run (Kr-85)

Xe = activity for significant xenon isotopes^(c) as given in the ORIGEN2 run (Xe-131m, Xe-133, Xe-133m)

${}^{131}I$ = activity for iodine-131 as given in the ORIGEN2 run.

For simplicity, all fission products were used because ORIGEN2 output represents total activity. Decay modes and beta energies were not considered, so that even those fission products that would not have been detected by the counters used in the 1940s were included in FP. Even though some fission products may have been present on vegetation, they may not have emitted beta particles at all or at least not those with sufficient energy to have been readily detected. Conversely, other fission products may have emitted much more energetic beta particles and, consequently, may have been detected much more readily. In this analysis, the simplifying assumption is made that all fission-product activity would have been detected equally by the mica-window GM counters. The krypton and xenon radionuclides were not accounted for in the total fission-product activity because they are noble gases which do not react chemically and, thus, do not deposit onto vegetation. Actinides were excluded because insignificant quantities would have been emitted. The ORIGEN2 run provided actinide values separately from the fission products.

The estimated activities of iodine-131 and other fission products as calculated by ORIGEN2 and computed by Equation (6.2) at the stack-discharge point are listed in Table 6.5. As given in Table 6.5, FP/T approximates the fraction of activity not resulting from

(a) Note that actinides are not included in the total fission products. Actinides are not fission products and would not be expected to be released from the dissolution process except in minute quantities. Xenon and krypton isotopes are subtracted out of the total amount because they are noble (inert) gases and would not have deposited onto vegetation. They could not have contributed to the gross beta count registered by the GM detectors.

(b) Significant krypton isotopes are those present in an appreciable quantity and, thus, affect the ratio FP/T if included in the total amount.

(c) Significant xenon isotopes are those present in an appreciable quantity and, thus, affect the ratio FP/T if included in the total amount.

iodine-131 decay relative to the total activity at the stack-discharge point in fuel-cooling time increments ranging from 10 to 90 days. Table 6.6 shows FP/T values computed from data in the ORIGEN2 run, those inferred from interpretation of historical vegetation data, and averages of those two data sets which BNW staff assume to be the best estimates of FP/T.

Table 6.5. Iodine-131, Fission Product (FP), and Total Activity (Curies) at Stack Discharge Computed from ORIGEN2 Data

Fuel-Cooling Time (Days)	$^{131}\text{I}^{(a)}$ at Stack Discharge	FP ^(b) at Stack Discharge	$^{131}\text{I} + \text{FP} = \text{Total (T)}$	FP/T ^(c)
10	10064	4	10068	<0.001
30	1796	3	1799	0.001
50	320	2	322	0.006
70	57	2	59	0.03
90	10	1	11	0.12

(a) Iodine-131 activity at stack discharge is estimated as the total iodine-131 activity given by the ORIGEN2 run multiplied by a release fraction of 0.91.
(b) FP at stack discharge is estimated by Equation (6.2).
(c) FP/T approximates the fraction of the total activity coming from fission products other than iodine-131. Conversely, the quantity $(1 - \text{FP/T})$ is the fraction of the activity that came from iodine-131: $\text{FP/T} = [\text{FP}(0.00001)]/[\text{FP}(0.00001) + \text{iodine-131}(0.905)]$

Table 6.6. Average Fuel-Cooling Times and Correlation of Fission Product to Total Activity (FP/T) from ORIGEN2 and Historical Measurements^(a)

Fuel-Cooling Time (Days)	FP/T Computed from ORIGEN2 Data	FP/T Inferred from Vegetation Data ^(b)	FP/T Averages of ORIGEN2 and Vegetation Data ^(c)
10	<0.001	<0.001	<0.001
30	0.001	0.001	0.001
50	0.006	0.003	0.005
70	0.03	0.23	0.13
90	0.12	0.50	0.31

(a) This table was compiled from information in Tables 6.2 through 6.5.
(b) The FP/T was inferred from vegetation data by considering the average fuel-cooling times and information on FP/T at certain dates given in Bulow (1945), Healy (1948b), and Parker (1948).
(c) Average FP/T values are assumed to be the best estimate from which to compute I_{FP} .

Values of FP/T generated from ORIGEN2 data must be considered only approximations. Computer-modeled fuel inventories have inherent limitations, especially for interpreting historical conditions. For instance, the actual burnup for any campaign would have varied somewhat even though they had the target burnup of 200 MWd/ST. It is difficult for any simulation to represent true reactor core conditions, given non-uniform flux and neutron energies, other variations and unknowns. In addition, the true release fraction from the fuel-dissolution process would be unique for each fission product. Nonetheless, the approximation of FP/T computed from ORIGEN2 data in this analysis is useful in quantifying the decrease in iodine-131 activity associated with longer fuel-cooling times because the vegetation data also have limitations (see Section 6.0).

The values of FP/T estimated from the ORIGEN2 data support the conclusions drawn from the historical record for fuel-cooling times from 10 to 50 days. However, for fuel-cooling times for 70 to 90 days, the estimates of FP/T from these two sources diverge. While the values at FP/T obtained using ORIGEN2 continue to increase fairly proportionally, the values obtained from the vegetation data increase sharply. This casts doubt on the reliability of the vegetation data for fuel-cooling times from 70 to 90 days, particularly for samples collected immediately after deposition. Because the time between deposition and counting is difficult to assess and because both estimation methods have limitations, it was decided to average the values of FP/T obtained using the two methods. I_{cf} was estimated using Equation (6.1) and the averages given in Table 6.6.

Table 6.7 provides best estimates of I_{cf} for cooling times. These approximate values of I_{cf} can be applied to historical pellet measurements by applying information on the fuel-cooling time from the reactor discharge to sample counting for the particular production campaigns that resulted in deposition on the collected sample. As mentioned previously, multiple campaigns could have resulted in deposition or not on a given vegetation sample depending on wind patterns and varying lengths of decay time since deposition. Thus, the application of I_{cf} is complex, and the I_{cf} value itself must be understood to be only an approximation, as are any measurements calculated using I_{cf} . In addition, I_{cf} can only be applied to measurements where detectable activity was observed; i.e., above 40 pCi/g (Leboeuf 1950).

Table 6.7. Best Estimate of I_{cf} Based on the Average of Historical Vegetation Data and Fission Products to Total Activity (FP/T) Generated from ORIGEN2 Data

Fuel-Cooling Time ^(a) (Days)	Best Estimate of I_{cf} ^(b)
10	> 0.99
30	0.99
50	0.99
70	0.87
90	0.69

(a) See Table 6.4 for information on average fuel-cooling times from December 1944 through December 1947.

(b) $I_{cf} = 1 - \text{FP/T}$. Application of I_{cf} to any given sample requires an estimate of the fuel-cooling time from the reactor discharge to the sample counting and interpolation of I_{cf} from the values in this table.

7.0 Results and Conclusions

In the years from 1945 through 1947, beta-counting technology had not yet progressed to the point where detector efficiency specific to the analysis of iodine-131 activities in vegetation could be determined (Healy et al. 1950). Historical conversion factors did not account for all of the parameters that affected overall efficiency of the beta-counting system. Some parameters were ignored or were miscalculated, and accurate iodine-131 standards were not available. As a result, the activities reported for vegetation were biased. However, as time went on, more factors were accounted for, so that the accuracy of the reported values improved.

Therefore, to adequately interpret the historical measurements, this analysis was conducted with the goal of providing reconstructed conversion factors for converting historical count data to an estimate of actual activity.

7.1 Results

Three primary factors affected what percentage of the beta particles emitted by iodine-131 disintegrations in a vegetation pellet resulted in counts registered by the GM counters used in the 1940s. These factors are the measurement conversion factor (M), the decay correction factor (D_e), and the iodine-131 assumption correction factor (I_{cf}).

7.1.1 Measurement Conversion Factor, M

The measurement conversion factor (M) converts original counting data into an approximation of disintegration rate from which activity in curies can be obtained by the standard relationship of 2.22×10^6 dpm/microcurie. The measurement conversion factor approximates the historical relationship between disintegrations in a 1-gram sagebrush pellet and the count rate registered by a GM detector.

M is also used to derive an adjustment factor for reported activities. The adjustment factor (A) is obtained by dividing the reconstructed M by the historical conversion factor assumed. Then, the reported activity can be multiplied by A , I_{cf} , and/or D_e (if appropriate) to obtain an approximation of the true activity in the pellet.

Table 7.1 provides results for A for two time periods, October 1945 through June 1946 and July 1946 through December 1947, and results for three time periods. The time periods are divided according to when changes occurred in the historical conversion factors assumed.

Table 7.1. Historical and Reconstructed Measurement Conversion Factors and Adjustment Factors for Measurements of Iodine-131 in Vegetation Pellets

Date ^(a)	Reconstructed M ^(b) (dpm/cpm)	Historical Conversion Factor ^(c) (dpm/cpm)	Adjustment Factor (A) ^(d)
10/45 - 01/46	{ 63.2	12.5	5.06
02/46 - 06/46		38.5	1.64
07/46 - 12/47	68.9	38.5	1.79
<p>(a) The time periods are based on historical procedure and historical conversion factors used. The pellets were formed by hand until about 7/46. The assumed historical conversion factor changed in February 1946 because of an additional factor of 3 to compensate for self-absorption within the pellet.</p> <p>(b) M is the factor discussed in Section 4.0.</p> <p>(c) The historical conversion factors are discussed in Section 2.0.</p> <p>(d) These unitless values are the ratio of M/(historical conversion factor).</p>			

7.1.2 Decay Correction Factor, D_e

The decay correction factor, D_e , corrects for the loss of iodine-131 in the interval between sample collection and counting. Because most raw count data in notebooks included information on the length of this time interval, a decay correction can be made. The uncertainty in this correction is based on a lack of knowledge about the time of day the sample collection and counting occurred (see Gilbert et al. 1992).

No decay correction is required for activities reported after June 1946 because this is when decay corrections started being routinely made.

The time of deposition to time of counting is not reflected in D_e . However, the interval between deposition and counting is an issue in interpreting and using the historical vegetation data. It is not an issue in the determination of D_e .

7.1.3 Iodine-131 Assumption Correction Factor, I_{cf}

The iodine-131 assumption correction factor, I_{cf} , corrects for the bias that occurred in historical measurements from assuming that all of the registered counts reflected iodine-131 decay. I_{cf} is based on an estimation of the ratio of iodine-131 activity to total activity as a

function of fuel-cooling time. The values in this report are based on limited historical information averaged together with the computer modeling data.

As discussed in Section 6.0, the application of I_{cf} requires an analysis of a number of variables that determine which fuel campaigns resulted in deposition on the given sample and the decay time. The value of I_{cf} to use for any given pellet depends on the probable fuel-cooling time for the campaign(s) that resulted in the deposition at that particular sampling location and on the time interval between deposition and sample collection.

7.2 Conclusions

Gilbert et al. (1992) provide a corollary analysis of the uncertainty in the total conversion factors. Gilbert et al. evaluate each parameter and develop a possible range of values and a best estimate of the true value for a given parameter based on the range. The results given in this report should be interpreted with an understanding of the uncertainty as provided in Gilbert's corollary report.

While these results are necessarily estimates, they provide a basis for estimating the overall uncertainty in the available vegetation data for the 1945-1947 period. Because the uncertainty in the total conversion factors (Gilbert et al. 1992) relative to the uncertainty in the models and model parameters being developed to predict radiation doses is small, further refinement of these vegetation estimates is not deemed appropriate nor planned for the balance of this project.

8.0 References

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Appendix A

Historical Techniques of Determining Parameters in M

Appendix A

Historical Techniques of Determining Parameters in M

This appendix describes the methods used in the 1940s and 1950s to derive parameters to calibrate mica-window GM counters to enable absolute beta counting.

A.1 Historical Techniques of Determining F_{sa}

A.1.1 Technique Used from February 1946 through November 1948

In 1945, the Health Instruments Group recognized the limitations of their method of counting iodine-131 activity on vegetation. They realized that the measurements were not a determination of absolute beta levels in vegetation but were relative. They attributed the relative nature of counting mainly to absorption of beta particles in the vegetation that had not been accounted for. Nevertheless, they felt that the relative values were good (Healy and Pahnke 1946). To reduce the amount of self-absorption, attempts were made in 1945 and early 1946 to develop chemical methods for extracting iodine-131 from vegetation (Healy and Eisenacher 1946; Healy and Pahnke 1946; Lindvig 1946a; Parker and Gamertsfelder 1945). These initial efforts failed, and no viable method of separating iodine-131 was achieved until 1948 (Singlevich 1948a). In the absence of such a method, it was decided that the most appropriate procedure would be to continue counting 1-gram pellets but to determine an additional parameter that would compensate for self-absorption. A factor of 3 (corresponding to an F_{sa} parameter of 0.33) was determined early in 1946 and applied to all results starting in February 1946 (Parker and Gamertsfelder 1945).

Although the literature surveyed does not provide an experimental method, Healy has surmised that the 1946 self-absorption parameter was probably determined by first counting an aliquot of iodine-131 solution on a planchet, then dropping an equivalent aliquot evenly onto the standard vegetation sample and counting again (Denham et al. 1988). The ratio between the two measurements was the self-absorption parameter. Self-absorption measurements were done five times each for four types of vegetation (sagebrush, seeds, twigs, and straw grass). Of these four types of vegetation, only sagebrush and straw grass were routinely collected (Bulow 1945; Dickinson 1946). Seeds and twigs were not typically counted. For sagebrush the average self-absorption parameter was 3 (Healy and Eisenacher 1946). Table A.1 lists the averages for the five determinations on the four vegetation types.

Table A.1. Historical Averages for Self-Absorption in Vegetation (Healy and Eisenacher 1946)

Vegetation Type	Average Empirical Parameter	Inverse Parameter (F_{sa})
Sagebrush	3.0	0.33
Seeds	2.5	0.40
Twigs	3.4	0.29
Straw Grass	2.4	0.42

Interpretation of the results is limited because the exact experimental method is not known. Thus, these values cannot be used to reconstruct F_{sa} as described in Section 4.2. For example, if the initial iodine-131 aliquot dropped onto the pellet approximated a point source, the empirical determination could also have compensated for sample spread. If the initial aliquot spread to approximate a pellet diameter, the result would not have compensated for that effect. In addition, the empirical determination could have accounted for any self-scatter and backscatter in the vegetation depending on how the aliquot was distributed. Another consideration is that if the initial aliquot was placed on stainless steel, the initial count would have been elevated because of the high level of backscatter expected from stainless steel.

A.1.2 Technique Used in the 1950s

In the 1950s, precise determinations were made of the self-absorption parameter for the types of analyses being done. At that time, counts of vegetation pellets had been replaced by counts of a chemical extraction of iodine-131 from the vegetation. The self-absorption measurements were, therefore, made for the type of carrier/residue obtained from the extraction process. The values for the self-absorption parameters, F_{sa} , generated from the 1950s studies are approximated by Equation (4.2) for silver iodide.

Schwendiman (1954) has detailed how the self-absorption parameter was determined for silver-iodide precipitates. A known volume of iodine-131 was associated with set quantities of silver-iodide precipitates. The silver iodide was precipitated according to the standard procedure and transferred in varying quantities to a standard mount. After drying, the plates were re-weighed and counted. Knowing the weight of precipitate made it possible to calculate the apparent specific activity (cpm/mg). This value was then divided by the actual specific activity to obtain the self-absorption/self-scatter parameter. From these data, curves of F_{sa} versus mass thickness (mg/cm^2) were generated.

A.2 Historical Techniques of Determining F_{bs}

In 1950, backscatter parameters were obtained by preparing sources of several isotopes of interest on Formvar film, the mass thickness of which was 0.3 to 0.5 mg/cm² (Healy et al. 1950). The film was supported on an open aluminum ring to aid in accurate positioning of the backing plates. An aliquot of carrier-free isotope was evaporated on the film. After drying, the source was counted on a holder which ensured that the ring position was reproducible. Backing material was then put into position just under the film and a second count was taken. The ratio of the second count to the first was the backscatter parameter. Because the backscatter parameter was expected to vary by 3 percent for a single type of measurement, several sources were prepared so that an average could be obtained (Schwendiman 1954).

A second method of determining the backscatter parameter was also mentioned by Schwendiman (1954). In this method, which was used less commonly, the counting rate of an aliquot evaporated directly on the backing material was then compared with the counting rate of an identical aliquot dried on the thin Formvar film. Backscatter parameters determined by this less common method were about the same as those determined by the first method.

From a figure in Thomas et al. (1956, p. 9) for a maximum beta energy of ~ 0.6 MeV, the backscatter parameter for shelf 1 is about 1.10 for H-70 filter paper.

A.3 Historical Technique of Determining F_a

Historically, absorption parameters were determined using Equation (4.3). The 1950s calibration research involved determining individual beta absorption coefficients for the isotopes being analyzed. Beta absorption coefficients for iodine-131 were determined in 1950, 1954, and 1956 and differed only slightly. Healy et al. (1950) and Schwendiman (1954) reported their methods for determining the iodine-131 beta absorption coefficient. No technique was reported by Thomas et al. (1956).

Beta absorption coefficients were determined by placing various mass thicknesses in mg/cm² of aluminum between the source and the mica-window GM counter (Healy et al. 1950; Schwendiman 1954). For thin absorbers, the relationship between absorption and counting rate followed an exponential curve. Thus, plotting the counting rate versus mass thickness on semi-logarithmic paper resulted in a straight line. The slope of this line was defined as the beta-absorption coefficient for the radionuclide being measured. However, the slope of the line was dependent on the relative geometries of the source, detector, and absorber, meaning the measurement technique could affect the beta-absorption coefficient. If the absorber were placed directly on the source, the counting rate would have been higher because of backscatter from parts of the absorber outside the solid angle. This effect was

noted in both the 1950 and the 1954 reports. To counteract this problem, the 1950 and 1954 determinations were made with the absorber as close as possible to the detector rather than the source.

In addition to the relative geometries, inert material associated with the source could also affect the beta-absorption coefficient (Schwendiman 1954). Schwendiman (1954) observed that when inert material was associated with the source, the beta absorption coefficient was higher. It was, therefore, critical that sources contain as little residue as possible and preferably none.

A.4 Historical Technique of Determining F_{cel}

Schwendiman (1954) observed that counting rate increased when the cellophane covering used to block the weak RaD (lead-210) beta and RaF (polonium-210) alpha particles was placed directly above the RaDEF standard as opposed to just below the window. He measured the ratio of counts per minute with cellophane at 0.0, 0.06, 0.12, and 0.18 inches from the source relative to counts per minute with the cellophane at the window. These measurements were made for all five shelves. Schwendiman (1954) concluded that the counting rate increased because beta particles that had initially been moving at angles far outside the geometry cone between the source and counter window were deflected by the cellophane. The measurements made by Schwendiman (1954) are relevant to the present analysis because historically cellophane was placed directly on top of vegetation pellets to aid in securing them.

A.5 Historical Technique of Determining G_p

Schwendiman (1954) has described in detail how G_p was determined for shelves 1 through 5. Both RaDEF and iodine-131 standards were used, and all the other parameters that affected counting rate were factored out. The USBS RaDEF standards have been cited as having an accuracy of ± 5 percent. However, by 1954, the activity of all standards was always verified at Hanford by using 4- π counting techniques. Four- π counting techniques were much more accurate because the standard was placed inside the detector's sensitive volume and, therefore, no losses would have occurred as a result of geometry or absorption outside of the sample itself. Thus the 4- π counting techniques should have provided an accurate determination of activity.

In addition, iodine-131 standards were used to determine the iodine-131 mass absorption coefficient (used in F_a) and to empirically determine F_{sa} . In the present analysis, both of these parameters are based on equations using a value for the iodine-131 beta absorption coefficient determined in 1977 (Baltakmens 1977).

Historically, G_p was calculated using the equation (Schwendiman 1954)

$$G_p = \text{cpm}/(\text{dpm} \times F_{bs} \times F_d \times F_a \times F_{sa} \times F_{cel} \times F_{decay}) \quad (\text{A.1})$$

where dpm refers to the disintegration rate for the RaE standard (bismuth -210), not iodine-131. The historical parameters F_{bs} , F_d , F_a , F_{cel} , and F_{sa} are discussed in Section 4.0 of this report. Historically, F_{sa} included a factor of 1.05 to account for scattering from the lead oxide carrier in the USBS RaDEF sources. The F_{decay} factor was the decay correction for the 22.2-year half-life of RaD (lead-210). The beta particles emitted by RaD (lead-210) were too low in energy to penetrate the cellophane covering, and RaF (polonium-210) emitted only an alpha particle which could not penetrate the cellophane, air, and mica window. The error in F_{decay} was < 1 percent if the original activity was decay-corrected every 3 months. All of the parameters in Equation (A.1) had been determined empirically. The result was the G_p for a given shelf (Healy et al. 1950).

The average G_p calculated for shelf 1 for all mica-window beta counters was 0.22 with a standard deviation of 0.01. Schwendiman (1954) found that the error introduced by assuming this average for all beta counters was less than the error arising from other sources and, therefore, felt it was justifiable to use these average G_p values for every mica-window GM detector in the counting laboratory.

A.6 Historical Technique of Determining F_d

An empirical method of determining F_d was reported by Healy et al. (1950). Schwendiman (1954) and Thomas et al. (1956) also used this same method. In 1950 and 1954 (Healy et al. 1950; Schwendiman 1954), F_d was determined empirically by first measuring the counting rate of a point source at various distances along a radius perpendicular to the axis of the detector. This measurement was repeated at each shelf distance. The relative counting rates (counts per minute at the radial distance to counts per minute at the origin where radial distance refers to the distance from the center to the circumference of the ring) were then integrated graphically after the counting rates at the radial distances had been adjusted to the area represented by an annular ring at that radius. Then F_d was the ratio of the graphically integrated counting rate of a uniform circular source of a given diameter (equalling two times the radial distance) to the counting rate of a point source of the same activity centered directly under the detector.

Healy et al. (1950) presented a figure with the vertical axis as relative counting rate (the ratio of the rate at a given distance to the rate at the center) using the RaDEF source and the horizontal axis as distance from the center in inches. The curve included both the measured points (the relative counting rate at radial distance versus distance) and values calculated using an equation that considered the difference in solid angles between a point source centered directly under the detector and a point source located at given radial distances (given the shelf

distances and diameter of the beta detector). They found excellent agreement for the RaDEF source between the empirical results and the values derived using their equation.

Kalmon (1953) confirmed the F_d data reported by Schwendiman (1954) for flat circular sources with maximum beta energies that were >0.3 MeV. Schwendiman's data include F_d values for diameters at increments of 1.27 cm (0.5 in.), 2.54 cm (1.0 in.), 3.81 cm (1.5 in.), and 5.08 cm (2.0 in.) for shelves 1 to 5. In addition, Schwendiman (1954) compared the 1950 method (called the point source traverse method) with a method in which sources were carefully prepared to be uniformly spread. The source used was ruthenium-rhodium-106, and these two methods also compared favorably.

Appendix B

Reconstructed Pellet Data and Results for M

Appendix B

Reconstructed Pellet Data and Results for M

This appendix contains tables of the dimensions of reconstructed hand-formed and press-formed sagebrush pellets and the values for parameters in M calculated for these pellets.

Table B.1. Dimensions for 1-Gram Hand-Formed Pellets

Pellet	Surface				
	Date Collected	Thickness (cm)	Diameter (cm)	Area ^(a) (cm ²)	Density (mg/cm ³)
1-H	9/11/89	0.64	4.06 4.37 3.86 3.96	13.0	121
2-H	9/11/89	0.66	4.22 4.01 3.76 3.86	12.3	123
3-H	9/11/89	0.51	3.91 3.81 5.18 3.56	13.3	148
4-H	9/11/89	0.64	3.96 3.35 4.06 3.66	11.1	142
5-H	9/11/89	0.61	4.67 4.98 4.37 4.57	17.0	97
6-H	9/11/89	0.66	4.52 3.45 4.83 4.52	14.7	103
7-H	9/11/89	0.64	4.01 4.52 4.01 3.61	12.8	123

(a) Surface area of the top of the pellet only.

Table B.1. (contd)

Pellet	Surface				
	Date Collected	Thickness (cm)	Diameter (cm)	Area ^(a) (cm ²)	Density (mg/cm ³)
8-H	9/11/89	0.61	4.11 4.22 3.81 5.08	14.6	113
9-H	9/11/89	0.64	3.86 3.20 3.76 4.67	11.8	134
10-H	9/11/89	0.58	3.66 4.42 4.62 4.27	14.1	121
11-H	10/16/89	0.58	3.56 4.06 4.06 4.32	12.6	137
12-H	10/16/89	0.62	3.30 4.32 4.57 3.05	11.4	142
13-H	10/16/89	0.67	3.81 4.06 3.30 4.06	11.4	132
14-H	10/16/89		4.06 4.06 3.81 4.57	13.4	110
15-H	10/16/89	0.63	4.06 3.56 4.06 4.06	12.2	130
16-H	11/30/89	0.51	3.30 3.56 4.32 4.57	12.2	162

(a) Surface area of the top of the pellet only.

Table B.1. (contd)

Pellet	Surface				
	Date Collected	Thickness (cm)	Diameter (cm)	Area ^(a) (cm ²)	Density (mg/cm ³)
17-H	11/30/89	0.56	4.06 5.33 5.08 5.59	19.8	91
18-H	11/30/89	0.51	4.06 4.57 4.06 4.57	14.6	134
19-H	11/30/89	0.46	4.57 5.08 3.81 4.83	16.4	133
20-H	11/30/89	0.51	3.81 4.83 4.57 4.06	14.6	134
21-H	12/28/89	0.33	6.05 4.14 3.66 5.08	17.6	172
22-H	12/28/89	0.41	4.14 4.45 4.14 4.45	14.5	170
23-H	12/28/89	0.56	4.45 4.78 4.78 4.14	16.1	111
24-H	12/28/89	0.48	3.66 4.78 5.08 4.45	15.8	131
25-H	12/28/89	0.48	4.14 4.93 5.41 4.45	17.6	118
(a) Surface area of the top of the pellet only.					

Table B.1. (contd)

Pellet	Surface				
	Date Collected	Thickness (cm)	Diameter (cm)	Area ^(a) (cm ²)	Density (mg/cm ³)
26-H	1/24/90	0.61	3.81 3.30 3.81 4.06	11.0	149
27-H	1/24/90	0.61	3.81 4.32 3.81 3.81	12.2	135
28-H	1/24/90	0.58	3.56 4.32 3.56 3.81	11.4	150
29-H	1/24/90	0.58	3.81 3.56 3.81 3.56	10.7	161
30-H	1/24/90	0.56	3.30 4.32 4.32 3.30	11.4	157
31-H	2/26/90	0.64	3.81 4.57 3.81 4.06	13.0	121
32-H	2/26/90	0.66	4.83 4.32 3.81 3.81	13.8	110
33-H	2/26/90	0.61	4.06 4.32 3.56 4.32	15.1	126
34-H	2/26/90	0.71	4.32 4.06 4.06 5.08	15.1	93
(a) Surface area of the top of the pellet only.					

Table B.1. (contd)

Pellet	Surface				
	Date Collected	Thickness (cm)	Diameter (cm)	Area ^(a) (cm ²)	Density (mg/cm ³)
35-H	2/26/90	0.56	3.81 4.57 4.06 4.83	14.6	122
36-H	4/12/90	0.48	4.57 3.56 4.32 4.83	14.6	142
37-H	4/12/90	0.56	4.06 5.33 4.32 4.32	16.0	112
38-H	4/12/90	0.66	3.81 4.06 3.30 4.06	11.4	133
39-H	4/12/90	0.51	3.81 5.08 4.57 4.06	15.1	131
40-H	4/12/90	0.56	4.32 4.57 4.06 5.08	16.0	112
41-H	5/24/90	0.58	4.32 4.57 4.57 3.56	14.2	120
42-H	5/24/90	0.56	4.06 3.56 4.57 4.57	13.8	130
43-H	5/24/90	0.53	4.32 4.57 4.06 4.06 4.57	14.2	132
(a) Surface area of the top of the pellet only.					

Table B.1. (contd)

Pellet	Surface				
	Date Collected	Thickness (cm)	Diameter (cm)	Area ^(a) (cm ²)	Density (mg/cm ³)
44-H	5/24/90	0.64	4.57	15.1	104
			3.56		
			5.33		
			4.06		
45-H	5/24/90	0.56	4.32	16.4	109
			5.08		
			4.32		
			4.57		
46-H	6/18/90	0.58	4.06	14.6	117
			4.57		
			4.57		
			4.06		
47-H	6/18/90	0.66	3.30	12.2	124
			4.57		
			3.56		
			4.32		
48-H	6/18/90	0.53	4.83	14.2	132
			3.56		
			4.06		
			4.57		
49-H	6/18/90	0.56	4.32	15.1	119
			4.32		
			4.32		
			4.57		
50-H	6/18/90	0.56	4.57	15.5	115
			5.08		
			3.81		
			4.32		
51-H	7/20/90	0.61	3.56	11.8	139
			3.81		
			4.57		
			3.56		
52-H	7/20/90	0.64	4.32	13.4	118
			3.81		
			3.81		
			4.57		

(a) Surface area of the top of the pellet only.

Table B.1. (contd)

Pellet	Surface				
	Date Collected	Thickness (cm)	Diameter (cm)	Area ^(a) (cm ²)	Density (mg/cm ³)
53-H	7/20/90	0.58	3.30 4.06 4.32 4.32	12.6	136
54-H	7/20/90	0.56	3.56 3.81 4.32 3.81	11.8	152
55-H	7/20/90	0.61	3.30 4.57 4.32 4.83	14.2	115
Average		0.58	4.19	13.9	128
(a) Surface area of the top of the pellet only.					

Table B.2. Dimensions for 1-Gram Press-Formed Pellets

Pellet	Surface				
	Date Collected	Thickness (cm)	Diameter (cm)	Area ^(a) (cm ²)	Density (mg/cm ³)
1-P	9/11/89	0.58	3.71 3.40 3.96 4.22	11.5	150
2-P	9/11/89	0.56	3.96 3.96 4.42 4.01	13.1	136
3-P	9/11/89	0.58	3.76 3.76 3.25 4.06	10.8	160
4-P	9/11/89	0.53	3.61 3.30 3.05 3.51	8.9	212
5-P	9/11/89	0.56	3.56 4.27 3.35 3.91	11.2	160
6-P	9/11/89	0.51	3.81 3.76 3.35 3.76	10.6	185
7-P	9/11/89	0.53	3.76 3.96 3.71 3.76	11.3	167
8-P	9/11/89	0.51	3.81 3.71 3.51 3.56	10.5	188
9-P	9/11/89	0.51	3.91 3.76 3.66 3.66	11.0	178
(a) Surface area of the top of the pellet only.					

Table B.2. (contd)

Pellet	Surface				
	Date Collected	Thickness (cm)	Diameter (cm)	Area ^(a) (cm ²)	Density (mg/cm ³)
10-P	9/11/89	0.53	3.61 3.76 3.66	11.3	168
11-P	10/16/89	0.70	3.56 3.81 3.81 3.30	10.3	139
12-P	10/16/89	0.63	3.30 3.81 3.81 3.56	10.3	154
13-P	10/16/89	0.48	4.06 3.81 4.06 3.81	12.2	171
14-P	10/16/89	0.52	4.06 3.81 3.81 3.81	11.8	163
15-P	10/19/89	0.62	3.81 3.30 3.56 3.81	10.3	157
16-P	11/30/89	0.51	3.81 4.06 3.81 3.81	11.8	166
17-P	11/30/89	0.51	4.32 4.06 4.32 4.83	15.1	130
18-P	11/30/89	0.58	3.56 4.06 3.81 4.32	12.2	142
(a) Surface area of the top of the pellet only.					

Table B.2. (contd)

Pellet	Surface				
	Date Collected	Thickness (cm)	Diameter (cm)	Area ^(a) (cm ²)	Density (mg/cm ³)
19-P	11/30/89	0.53	4.06	12.6	150
			4.06		
			3.81		
			4.06		
20-P	11/30/89	0.53	4.06	12.2	179
			4.06		
			3.81		
			3.81		
21-P	12/28/89	0.56	3.51	10.7	166
			4.14		
			3.81		
			3.33		
22-P	12/28/89	0.56	3.81	11.0	163
			3.66		
			3.81		
			3.66		
23-P	12/28/89	0.56	4.45	12.9	138
			4.14		
			3.66		
			3.96		
24-P	12/28/89	0.41	4.78	14.5	168
			4.78		
			3.81		
			3.81		
25-P	12/28/89	0.41	3.96	10.7	228
			3.81		
			3.66		
			3.33		
26-P	1/9/90	0.61	3.56	10.7	154
			3.81		
			3.56		
			3.81		
27-P	1/9/90	0.53	3.81	11.0	171
			3.81		
			3.30		
			4.06		

(a) Surface area of the top of the pellet only.

Table B.2. (contd)

Pellet	Surface				
	Date Collected	Thickness (cm)	Diameter (cm)	Area ^(a) (cm ²)	Density (mg/cm ³)
28-P	1/9/90	0.56	4.06 4.06 3.30 3.81	11.4	157
29-P	1/24/90	0.56	3.81 4.32 4.06 4.06	13.0	138
30-P	1/24/90	0.66	3.81 3.81 4.83 4.06	13.4	113
31-P	2/26/90	0.53	3.81 3.81 4.06 4.06	12.2	155
32-P	2/26/90	0.56	3.56 3.81 3.56 4.06	11.0	162
33-P	2/26/90	0.56	4.06 3.81 3.56 4.06	11.8	152
34-P	2/26/90	0.53	3.81 3.81 3.56 3.56	10.7	177
35-P	2/26/90	0.58	4.32 4.32 4.32 3.56	13.4	129
36-P	4/12/90	0.48	3.81 3.81 3.81 4.06	11.8	177

(a) Surface area of the top of the pellet only.

Table B.2. (contd)

Pellet	Surface				
	Date Collected	Thickness (cm)	Diameter (cm)	Area ^(a) (cm ²)	Density (mg/cm ³)
37-P	4/12/90	0.48	3.81	13.0	161
			4.32		
			4.06		
			4.06		
38-P	4/12/90	0.53	3.81	11.8	160
			4.06		
			3.81		
			3.81		
39-P	4/12/90	0.48	3.81	12.2	171
			3.81		
			4.32		
			3.81		
40-P	4/12/90	0.48	4.57	14.2	147
			4.32		
			4.06		
			4.06		
41-P	5/24/90	0.58	4.06	12.2	142
			3.81		
			3.81		
			4.06		
42-P	5/24/90	0.53	4.06	12.6	150
			3.81		
			4.06		
			4.06		
43-P	5/24/90	0.56	4.06	13.4	134
			4.32		
			4.06		
			4.06		
44-P	5/24/90	0.48	4.06	11.8	177
			4.06		
			3.81		
			3.56		
45-P	5/24/90	0.51	4.06	12.6	156
			4.06		
			4.06		
			3.81		

(a) Surface area of the top of the pellet only.

Table B.2. (contd)

Pellet	Surface				
	Date Collected	Thickness (cm)	Diameter (cm)	Area ^(a) (cm ²)	Density (mg/cm ³)
46-P	6/18/90	0.53	3.56 4.06 3.81 4.06	11.8	160
47-P	6/18/90	0.53	4.06 4.06 3.81 4.06	12.6	150
48-P	6/18/90	0.56	3.81 3.81 3.81 3.56	11.0	162
49-P	6/18/90	0.53	3.81 3.81 3.81 4.06	11.8	160
50-P	6/18/90	0.56	4.06 4.06 3.81 3.81	12.2	147
51-P	7/20/90	0.58	4.06 3.30 3.56 3.81	10.7	162
52-P	7/20/90	0.58	3.81 3.30 4.06 3.81	11.0	157
53-P	7/20/90	0.53	4.06 3.56 4.32 3.56	11.8	160
54-P	7/20/90	0.58	3.56 3.81 4.06 4.32	12.2	142
(a) Surface area of the top of the pellet only.					

Table B.2. (contd)

Pellet	Surface				
	Date Collected	Thickness (cm)	Diameter (cm)	Area ^(a) (cm ²)	Density (mg/cm ³)
55-P	7/20/90	0.61	3.81	11.8	139
			3.81		
			3.81		
			4.06		
Average		0.54	3.87	11.8	159
(a) Surface area of the top of the pellet only.					

Table B.3. Parameters in M for 1-Gram Reconstructed Hand-Formed Pellets

Pellet	G_p	F_d	F_{ba}	F_{col}	F_m	F_s	E_o	M (dpm/ cpm)	1/M (cpm/ dpm)
1-H	0.084	0.67	1.10	1.04	0.33	0.73	1.00	63.5	0.016
2-H	0.085	0.68	1.10	1.04	0.32	0.73	1.00	64.6	0.015
3-H	0.080	0.67	1.10	1.04	0.34	0.73	1.00	65.4	0.015
4-H	0.084	0.70	1.10	1.04	0.29	0.73	1.00	69.1	0.014
5-H	0.083	0.62	1.10	1.04	0.41	0.73	1.00	56.7	0.018
6-H	0.085	0.64	1.10	1.04	0.37	0.73	1.00	59.2	0.017
7-H	0.084	0.67	1.10	1.04	0.33	0.73	1.00	63.9	0.016
8-H	0.083	0.65	1.10	1.04	0.37	0.73	1.00	60.5	0.016
9-H	0.084	0.69	1.10	1.04	0.31	0.73	1.00	66.8	0.015
10-H	0.082	0.66	1.10	1.04	0.36	0.73	1.00	61.9	0.016
11-H	0.082	0.68	1.10	1.04	0.32	0.73	1.00	65.7	0.015
12-H	0.083	0.70	1.10	1.04	0.30	0.73	1.00	68.4	0.015
13-H	0.085	0.70	1.10	1.04	0.30	0.73	1.00	67.3	0.015
14-H	0.085	0.66	1.10	1.04	0.34	0.73	1.00	61.6	0.016
15-H	0.084	0.68	1.10	1.04	0.32	0.73	1.00	65.6	0.015
16-H	0.080	0.69	1.10	1.04	0.32	0.73	1.00	68.5	0.015
17-H	0.081	0.59	1.10	1.04	0.46	0.73	1.00	54.6	0.018
18-H	0.080	0.65	1.10	1.04	0.37	0.73	1.00	62.3	0.016
19-H	0.078	0.64	1.10	1.04	0.40	0.73	1.00	60.2	0.017
20-H	0.080	0.65	1.10	1.04	0.37	0.73	1.00	62.3	0.016
21-H	0.075	0.63	1.10	1.04	0.42	0.72	1.00	60.9	0.016
22-H	0.077	0.66	1.10	1.04	0.37	0.73	1.00	64.8	0.015
23-H	0.081	0.63	1.10	1.04	0.40	0.73	1.00	58.8	0.017
24-H	0.079	0.64	1.10	1.04	0.39	0.73	1.00	60.7	0.016
25-H	0.079	0.62	1.10	1.04	0.42	0.73	1.00	58.2	0.017
26-H	0.083	0.70	1.10	1.04	0.29	0.73	1.00	70.0	0.014
27-H	0.083	0.68	1.10	1.04	0.32	0.73	1.00	66.2	0.015
28-H	0.082	0.70	1.10	1.04	0.30	0.73	1.00	69.2	0.014
29-H	0.082	0.71	1.10	1.04	0.28	0.73	1.00	72.0	0.014

Table B.3. (contd)

Pellet	G_p	F_d	F_{ba}	F_{col}	F_{ca}	F_a	E_o	M (dpm/ cpm)	1/M (cpm/ dpm)
30-H	0.081	0.70	1.10	1.04	0.30	0.73	1.00	69.8	0.014
31-H	0.084	0.67	1.10	1.04	0.33	0.73	1.00	63.5	0.016
32-H	0.085	0.66	1.10	1.04	0.35	0.73	1.00	61.0	0.016
33-H	0.083	0.67	1.10	1.04	0.33	0.73	1.00	64.0	0.016
34-H	0.087	0.63	1.10	1.04	0.38	0.74	1.00	57.6	0.017
35-H	0.081	0.65	1.10	1.04	0.37	0.73	1.00	61.3	0.016
36-H	0.079	0.66	1.10	1.04	0.37	0.73	1.00	62.8	0.016
37-H	0.081	0.63	1.10	1.04	0.39	0.73	1.00	59.0	0.017
38-H	0.085	0.70	1.10	1.04	0.30	0.73	1.00	67.4	0.015
39-H	0.080	0.65	1.10	1.04	0.38	0.73	1.00	61.5	0.016
40-H	0.081	0.63	1.10	1.04	0.39	0.73	1.00	59.0	0.017
41-H	0.082	0.66	1.10	1.04	0.36	0.73	1.00	61.7	0.016
42-H	0.081	0.66	1.10	1.04	0.35	0.73	1.00	63.1	0.016
43-H	0.081	0.66	1.10	1.04	0.36	0.73	1.00	62.7	0.016
44-H	0.084	0.64	1.10	1.04	0.38	0.73	1.00	59.1	0.017
45-H	0.081	0.63	1.10	1.04	0.40	0.73	1.00	58.4	0.017
46-H	0.082	0.65	1.10	1.04	0.37	0.73	1.00	60.8	0.016
47-H	0.085	0.68	1.10	1.04	0.32	0.73	1.00	65.0	0.015
48-H	0.081	0.66	1.10	1.04	0.36	0.73	1.00	62.7	0.016
49-H	0.081	0.64	1.10	1.04	0.38	0.73	1.00	60.5	0.016
50-H	0.081	0.64	1.10	1.04	0.38	0.73	1.00	59.8	0.017
51-H	0.083	0.69	1.10	1.04	0.31	0.73	1.00	67.4	0.015
52-H	0.084	0.66	1.10	1.04	0.34	0.73	1.00	62.5	0.016
53-H	0.082	0.68	1.10	1.04	0.32	0.73	1.00	65.6	0.015
54-H	0.081	0.70	1.10	1.04	0.31	0.73	1.00	68.6	0.014
55-H	0.083	0.65	1.10	1.04	0.36	0.73	1.00	61.2	0.016
Average	0.082	0.66	1.10	1.04	0.35	0.73	1.00	63.2	0.016
Min	0.075	0.59	1.10	1.04	0.28	0.72	1.00	54.6	0.014
Max	0.087	0.71	1.10	1.04	0.46	0.74	1.00	72.0	0.018

Table B.4. Parameters in M for 1-Gram Reconstructed Press-Formed Pellets

Pellet	G_p	F_d	F_{ba}	F_{col}	F_m	F_a	E_o	M (dpm/ cpm)	1/M (cpm/ dpm)
1-P	0.083	0.70	1.10	1.04	0.30	0.73	1.00	68.7	0.014
2-P	0.082	0.67	1.10	1.04	0.34	0.73	1.00	64.4	0.016
3-P	0.083	0.71	1.10	1.04	0.29	0.73	1.00	71.2	0.014
4-P	0.081	0.75	1.10	1.04	0.24	0.73	1.00	81.8	0.012
5-P	0.082	0.70	1.10	1.04	0.30	0.73	1.00	70.3	0.014
6-P	0.080	0.72	1.10	1.04	0.28	0.73	1.00	73.9	0.014
7-P	0.081	0.70	1.10	1.04	0.30	0.73	1.00	70.5	0.014
8-P	0.080	0.72	1.10	1.04	0.28	0.73	1.00	74.5	0.013
9-P	0.080	0.71	1.10	1.04	0.29	0.73	1.00	72.1	0.014
10-P	0.081	0.70	1.10	1.04	0.30	0.73	1.00	70.8	0.014
11-P	0.087	0.71	1.10	1.04	0.27	0.74	1.00	70.1	0.014
12-P	0.084	0.72	1.10	1.04	0.27	0.73	1.00	71.9	0.014
13-P	0.079	0.69	1.10	1.04	0.32	0.73	1.00	68.9	0.014
14-P	0.081	0.70	1.10	1.04	0.31	0.73	1.00	69.2	0.014
15-P	0.084	0.72	1.10	1.04	0.27	0.73	1.00	72.2	0.014
16-P	0.080	0.70	1.10	1.04	0.31	0.73	1.00	69.4	0.014
17-P	0.080	0.65	1.10	1.04	0.38	0.73	1.00	61.2	0.016
18-P	0.083	0.69	1.10	1.04	0.32	0.73	1.00	66.5	0.015
19-P	0.081	0.68	1.10	1.04	0.32	0.73	1.00	66.6	0.015
20-P	0.079	0.69	1.10	1.04	0.32	0.73	1.00	69.4	0.014
21-P	0.082	0.71	1.10	1.04	0.28	0.73	1.00	72.0	0.014
22-P	0.082	0.71	1.10	1.04	0.29	0.73	1.00	71.1	0.014
23-P	0.082	0.68	1.10	1.04	0.33	0.73	1.00	65.0	0.015
24-P	0.077	0.66	1.10	1.04	0.36	0.73	1.00	64.4	0.016
25-P	0.077	0.72	1.10	1.04	0.28	0.73	1.00	76.1	0.013
26-P	0.084	0.71	1.10	1.04	0.28	0.73	1.00	70.9	0.014
27-P	0.081	0.71	1.10	1.04	0.29	0.73	1.00	71.6	0.014
28-P	0.082	0.70	1.10	1.04	0.30	0.73	1.00	69.5	0.014
29-P	0.082	0.67	1.10	1.04	0.33	0.73	1.00	64.8	0.015

Table B.4. (cont)

Pellet	G _p	F _d	F _{bo}	F _{col}	F _m	F _s	E _o	M (dpm/ cpm)	1/M (cpm/ dpm)
30-P	0.085	0.66	1.10	1.04	0.34	0.73	1.00	61.6	0.016
31-P	0.081	0.69	1.10	1.04	0.32	0.73	1.00	67.7	0.015
32-P	0.082	0.71	1.10	1.04	0.29	0.73	1.00	70.8	0.014
33-P	0.082	0.69	1.10	1.04	0.31	0.73	1.00	68.2	0.015
34-P	0.081	0.72	1.10	1.04	0.28	0.73	1.00	73.0	0.014
35-P	0.083	0.67	1.10	1.04	0.34	0.73	1.00	63.3	0.016
36-P	0.079	0.70	1.10	1.04	0.31	0.73	1.00	70.1	0.014
37-P	0.079	0.68	1.10	1.04	0.33	0.73	1.00	66.6	0.015
38-P	0.081	0.70	1.10	1.04	0.31	0.73	1.00	68.9	0.014
39-P	0.079	0.69	1.10	1.04	0.32	0.73	1.00	68.9	0.014
40-P	0.079	0.66	1.10	1.04	0.36	0.73	1.00	63.6	0.016
41-P	0.083	0.69	1.10	1.04	0.32	0.73	1.00	66.6	0.015
42-P	0.081	0.68	1.10	1.04	0.32	0.73	1.00	66.8	0.015
43-P	0.082	0.67	1.10	1.04	0.34	0.73	1.00	63.8	0.016
44-P	0.079	0.70	1.10	1.04	0.31	0.73	1.00	70.1	0.014
45-P	0.080	0.68	1.10	1.04	0.32	0.73	1.00	67.0	0.015
46-P	0.081	0.70	1.10	1.04	0.31	0.73	1.00	68.9	0.014
47-P	0.081	0.68	1.10	1.04	0.32	0.73	1.00	66.6	0.015
48-P	0.082	0.71	1.10	1.04	0.29	0.73	1.00	70.8	0.014
49-P	0.081	0.70	1.10	1.04	0.31	0.73	1.00	68.9	0.014
50-P	0.082	0.69	1.10	1.04	0.32	0.73	1.00	67.0	0.015
51-P	0.083	0.71	1.10	1.04	0.28	0.73	1.00	71.8	0.014
52-P	0.083	0.71	1.10	1.04	0.29	0.73	1.00	70.4	0.014
53-P	0.081	0.70	1.10	1.04	0.31	0.73	1.00	68.9	0.014
54-P	0.083	0.69	1.10	1.04	0.32	0.73	1.00	67.0	0.015
55-P	0.084	0.69	1.10	1.04	0.31	0.73	1.00	67.0	0.015
Average	0.081	0.70	1.10	1.04	0.31	0.73	1.00	68.9	0.014
Min	0.077	0.65	1.10	1.04	0.24	0.73	1.00	61.2	0.012
Max	0.087	0.75	1.10	1.04	0.38	0.74	1.00	81.8	0.016

Appendix C

Summary of Technical Steering Panel Comments and Battelle, Pacific Northwest Laboratories Responses

Appendix C

Summary of Technical Steering Panel Comments and Battelle, Pacific Northwest Laboratories Responses

Document Number PNWD-2133 HEDRDocument Title *Conversion and Correction Factors for
Historical Measurements of Iodine-131
in Hanford-Area Vegetation, 1945-1947*

Comment Number	Commenter	Page, Paragraph	Comment Summary	Resolution
	B. Shleien	Summary Comment	<p>This document is important, well organized and adequately written, however, several TSP members expressed confusion as to how it fit into the HEDR Project task plan. The document PNWD-2176 and the subject document together appear to cover the period 1945-51. Your letter of May 19, 1993 does not clarify this nor does PNWD-2133. Correspondence between B. Shleien and D. Shieler did elucidate this matter and I suggest that it be included in a cover letter or in the preface to PNWD-2133.</p> <p>Other comments by TSP members should be considered from an editorial point of view although some concern matters of substance.</p>	Agree. Revised preface addresses the question. The September 1992 HEDR monthly report explained that the Integrated Task Plans (PNWD-2020 HEDR) definition of Milestone 0502A would be changed to denote that there would be two reports: Milestone 0502A and Milestone 0502B.
1	S. Davis	Full Report	As an editor, I would suggest that all references to completed work should be in the past tense. For example, write "Healy (1948) <u>stated</u> ", not "states." Healy wrote the report in the past, so references should be in the past tense. Only about one-third of the references in the document are correctly given in the past tense.	Tenses changed where applicable. General rule is to use the present tense for concepts that are established knowledge (see, for example, Robert Day's, <i>How to Write and Publish a Scientific Paper</i>).
2	G. Roessler	General Comment	This is an important document and the depth of the work that went into it is obvious. However, it would be helpful to mention somewhere how the iodine-131 vegetation data will be used to "support this work" (page iii, paragraph 5). It is stated in PNWD-1978, "Uncertainty and Sensitivity Analysis of Historical Measurements of Iodine-131 for Vegetation in 1945-1947," in the preface; however, each report should stand alone. Therefore, it seems like it should be repeated in this one.	Agree. Revised preface addresses the question.

C.1

Document Number PNWD-2133 HEDRDocument Title Conversion and Correction Factors for
Historical Measurements of Iodine-131 in
Hanford-Area Vegetation, 1945-1947

Comment Number	Commenter	Page, Paragraph	Comment Summary	Resolution
3	G. Caldwell	General Comment	My major question is whether or not this is supposed to satisfy the plan requirements for "Vegetation Monitoring Data and Correction Factors, 1945-1951" (pages 6.5, 6.7, and 6.11). It seems to me that this report covers only part of the 0502A task and only the first five years. Page 6.11 of the Integrated Task Plans (PNWD-2020 HEDR) seems to indicate that 0502A is a Vegetation Data Report for 1945-1951. This may be part of 0502B which includes bias and correction factors (page 6.7). I do not believe this is acceptable to fulfill 0502A as listed in the plan or all of 0502B.	Agree. See response to summary comment.
4	J. Till	General Comment	<p>Please note that a response is needed to the discussion below as there seems to be a major discrepancy between what is described in the work plan and the contents of this report. Perhaps I have missed something here but it appears we have missed the primary purpose of the work related to vegetation monitoring data.</p> <p>I am very concerned that this is the extent of the vegetation data work. The work plan describes task 0502A as "A Summary Report". The description is as follows: "Vegetation monitoring data and conversion factors (1945-1951). This report documents the vegetation monitoring data and conversion/correction factors for the period 1945-1948 and 1948-1951 (combined reports from Phase I)." Yet this report deals with <u>only</u> the correction factor for historical monitoring. Very little vegetation data are summarized in this report. I was expecting a report that listed all monitoring data for vegetation for the period of concern and some discussion of the correction factors.</p> <p>If this is the extent of the vegetation data, we have significantly under-emphasized this important part of the work. These data should help form what we call the proof in dose reconstruction.</p>	Agree. See response to summary comment.

C.2

Document Number PNWD-2133 HEDRDocument Title Conversion and Correction Factors for
Historical Measurements of Iodine-131 in
Hanford-Area Vegetation, 1945-1947

Comment Number	Commenter	Page, Paragraph	Comment Summary	Resolution
5	B. Shleien	General Comment	<p>I am aghast at the statement in your letter of May 19, 1993, indicating your opinion that PNWD-2133 fulfills the requirements of Task 0502A. If this is the case, then I do not understand SUBTASK 0502: TERRESTRIAL MONITORING DATA (PNWD-2020 INTEGRATED TASK PLANS FOR THE HEDR PROJECT, FY 1992 THROUGH MAY 1994). It has been my understanding that 0502A would include "<u>Vegetation Monitoring Data and Correction Factors, 1945-1951.</u>" Page 6.11 of PNWD-2020 specifically indicates that 0502A, Vegetation Data Report (1945-1951) would be completed by November 1992. Thus far I have not received any report on vegetation monitoring data that states the relationship between I-131 on the vegetation, its location, and the date of sample collection. This is particularly perturbing because of its absence for the years 1945-1951. Until this information appears in PNWD-2133 I will encourage the TSP not to accept it as a complete fulfillment of 0502A.</p> <p>The entire collection of environmental monitoring data for the HEDR Project appears to be in a very sorry state to me. Task 0501A, Environmental Monitoring Data Final Report was due October 1992, and has as yet not been delivered. 0502B, Letter Report: Vegetation bias/data collection (which I thought by the way was the subject of the document I just received, PNWD-2133) is also overdue by a half-year.</p> <p>I am particularly perturbed by the lack of progress on vegetation monitoring data, because it still remains a major method for verification of RATCHET. I cannot agree to the employment of RATCHET to calculate atmospheric dispersion prior to its validation and verification. From what I see, the HEDR Project still does not have the vegetation monitoring data available to test the model. I and others on the TSP have emphasized the need for vegetation monitoring data to check the HEDR models from almost the first meeting of the Panel.</p>	Agree. See response to summary comment. See also Napier et al. (1993).
6	S. Davis	Page v, line 6 from the top	Isn't the period supposed to be 1945-1951, not 1945-1947?	See response to summary comment.
7	S. Davis	Page xi	Activity is the number of nuclear transformations (add) <u>per unit time</u> .	Recommended wording incorporated.

C.3

Document Number PNWD-2133 HEDRDocument Title Conversion and Correction Factors for
Historical Measurements of Iodine-131 in
Hanford-Area Vegetation, 1945-1947

Comment Number	Commenter	Page, Paragraph	Comment Summary	Resolution
8	G. Roessler	Page xi, 2nd definition--Activity	The number of nuclear transformations (disintegrations) <i>per unit time</i> .	See response to comment No. 7.
9	G. Roessler	Page xiii, Four-Pi Counting	I'd replace the first sentence with: Counting techniques in which the geometry approaches <i>that of a sphere, i.e., 4 steradians</i> .	Recommended wording incorporated.
10	G. Roessler	Page xiii, Half-life	I'd recommend the following: <i>The time required for an initial number of radioactive atoms to be reduced to half that number by transformations.</i>	Recommended wording incorporated.
11	G. Roessler	Page 1.1, last para.	If someone wanted to "address the detector systems themselves," are there any still in operation so that this could be done?	Historical detector systems not available for commercial use.
12	G. Roessler	Page 1.2, para. 2	Did the veteran employees recall why only reported iodine-131 values were available from August 1946 through December 1947? Is there any hope of finding the original beta counting data? Is it worth looking or are your adjustment factors good enough so that further searching for the original data is not justified?	Question was addressed in discussions with veteran employees, but no one knew where the data were. Further data searches are not recommended, as 1) such searches would probably not uncover the original beta counting data and 2) the adjustment factors identified for the data are adequate considering that during this period of time most of the data were at or near the detection limit (see Denham et al 1993a). Net counting rates ranged from 0 to 20 cpm/g and reported activities were in the 0 to 0.4 mCi/kg range. (Most of the data were reported either in cpm/g or in mCi/kg, but not both.) Section 6.1.1 of this report lists the average concentration of potassium-40 at about 3.5 cpm/g. This represents from 15 to as much as 100 percent of all the beta activity detected on samples from the 1946-1947 period.
13	G. Caldwell	Page 2.3, entry 8/45	"... counted on second shelf ..." and similar elsewhere. Need to indicate from top or bottom of counter. Pages 2.11 and 2.12 seem to indicate from top by measurements but it might be clearer to the persons not knowledgeable about ancient equipment to indicate exact location during first mention.	Agree. Words "from the top" added when discussing location of Geiger-Mueller detector shelves.

C.4

Document Number PNWD-2133 HEDRDocument Title Conversion and Correction Factors for
Historical Measurements of Iodine-131 in
Hanford-Area Vegetation, 1945-1947

Comment Number	Commenter	Page, Paragraph	Comment Summary	Resolution
14	G. Caldwell	Page 2.9, item 6A	Indicates "... thistle thorns.", but the report in general seems that mostly sagebrush was counted. How much of the data are "sagebrush" and how much are other plants (thistle, grass, etc.)? In addition, were the various kinds of vegetation comparable?	See paragraph 4 of Section 2.1 and paragraph 5 of Section 1.0, page 1.2.
15	G. Caldwell	Page 2.9, items 1, 7B and 7F	Was a different sleeve used for each sample to avoid cross-contamination?	See number 1 of paragraph 3, Section 2.1.3.
16	G. Roessler	Page 2.14, last para.	It would be clearer if the 2nd sentence showed that the 12.5 comes from dividing by 0.08.	Agree. Sentence changed
17	S. Davis	Page 4.3, item 2	It might be important to note that H-3 was probably not detected by the earliest counters because of the low average energy of emission (0.019 MeV). Also would Ru-106 (0.039 MeV) have been detected?	Disagree. The scope of work was to determine the accuracy of the calibration for iodine-131. Ruthenium-106 would not have been detected.
18	G. Roessler	Page 4.13, last para.	A diagram would sure help here. I am having a hard time picturing those betas getting deflected toward the detector. I believe that the count rates were higher since Schwendiman (1954) made the empirical measurements, but I wonder if he had the right phenomenon.	Clarified in text.
19	G. Roessler	Page 4.19, para. 4	The iodine referred to here, I assume, entered the somata of leaves by deposition on the plant and then absorption into the plant rather than by uptake from the soil.	Agree. Clarified in text.
20	G. Roessler	Page 4.21, para. 2	This would be more reassuring if you were able to say: <i>Few environmental samples during this time period had counts high enough to result in significant coincidence losses.</i> The releases during this time period were high, were the activities on vegetation actually quite low for most or all samples?	Agree. Recommended wording incorporated.
21	G. Roessler	Page 6.3, para. 2	It seems like potassium-40 would have contributed about 1%. Perhaps this number should be mentioned so the "minimal" contribution by potassium-40 can be compared to the apparently significant contribution by other parameters which received a lot of attention.	We agree that the contribution from potassium-40 was minimal. The overall count rate from potassium-40 was minimal. However, the percentage to the overall count rate varied depending on the total amount of contamination. With no contamination, the count rate was low but the percentage from potassium-40 could have approached 100 percent.

C-5

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