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UC-707

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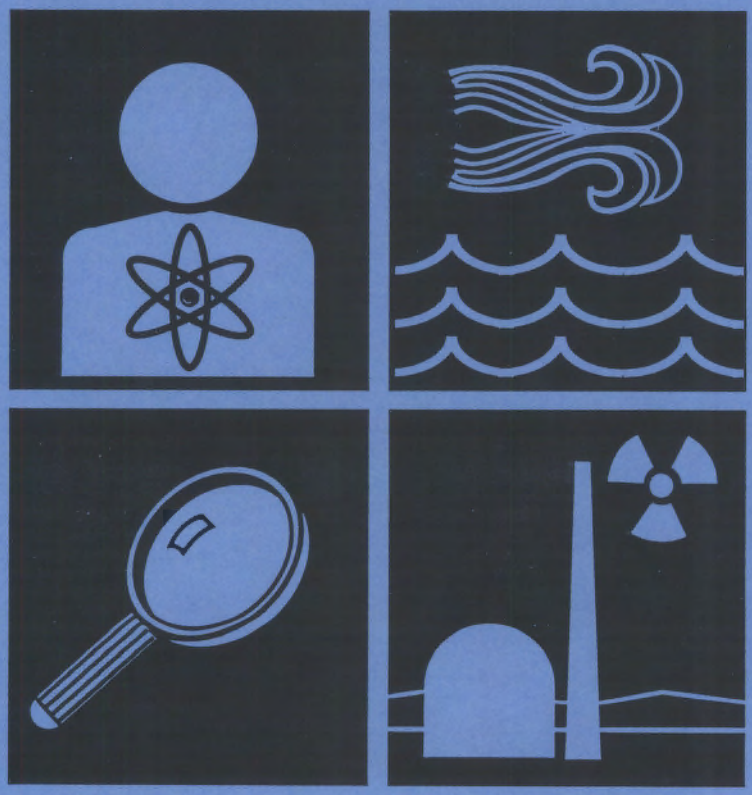
Uncertainties in Source Term Calculations Generated by the ORIGEN2 Computer Code for Hanford Production Reactors

C. M. Heeb

March 1991



Prepared for the Technical Steering Panel

Battelle

PNL-7223 HEDR

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UNCERTAINTIES IN SOURCE TERM CALCULATIONS GENERATED BY THE ORIGEN2 COMPUTER CODE FOR HANFORD PRODUCTION REACTORS

March 1991

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

John E. Till, Chairman
Technical Steering Panel
Hanford Environmental
Dose Reconstruction Project

March 15, 1991
Date

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Hanford Environmental Dose Reconstruction Project

C. M. Heeb

March 1991

Prepared for the
Technical Steering Panel

Pacific Northwest Laboratory
Richland, Washington 99352

FOREWORD

The appendix to this document is a record of TSP comments and Battelle responses to those comments; the TSP has reviewed and approved Battelle's responses. Comment numbers appear in the left margin next to the paragraphs in which the corresponding comments were addressed. Any changed text is shown in italics. In addition to responses to TSP comments, some text has been changed to correct errors and for further clarification.

SUMMARY

The principal sources of uncertainty in the application of the ORIGEN2 computer code to Hanford Environmental Dose Reconstruction (HEDR) Project source term calculations are considered in this report. Comparisons are made between calculated ORIGEN2 values and measurements of light water reactor spent fuel decay heat, isotopic content, and neutron and gamma ray source strengths. Nuclear data uncertainties and code input uncertainties are considered. Because no ORIGEN2 cross-section library of radionuclides exists for the eight original Hanford Production Reactors (HPRs), comparisons are made with various ORIGEN2 libraries, and with computed neutron spectra for the N Reactor and the HPRs in an effort to estimate the maximum uncertainty.

The *range* of uncertainty from using the ORIGEN2 code to calculate source terms for the HEDR Project is estimated to be *within* the following *limits:*

- nuclear data--less than 10%
- code input information--within 10%, *for most isotopes*
- code accuracy compared with measurements--within 10%
- using the N Reactor cross-section library for the older HPRs--generally small uncertainties for most nuclides, but up to a factor of 2 for certain nuclides *(see Table 3.2).*

Uncertainties in some calculated activation products can be much larger than those listed above. Non-fuel activation products--such as reactor stack gas, cooling water activations, and coolant system crud--have highly uncertain neutron flux exposure times (i.e., the uncertainty in code input is larger than 10%). Also, the uncertainty in using the N Reactor cross-section library will be larger for activation products in general because the computed activation rate depends on the value of the spectrum-averaged neutron absorption cross section.

The uncertainties described here are inevitable in the use of the ORIGEN2 code when the N Reactor ORIGEN2 cross-section library of radionuclides is used to calculate source terms for the older HPRs. ORIGEN2 cross-section libraries specific to the HPRs can be generated, thereby reducing the uncertainties, if

such action is determined to be of benefit in reducing the uncertainties in the estimates of radiation dose received as a result of Hanford Site Operations.

GLOSSARY

13	*Actinide*	*The elements thorium, protactinium, uranium, neptunium, plutonium, americium, curium, and those of higher atomic mass.*
	ANSI/ASME NQA - 1	National standard for Nuclear Grade Quality Assurance.
	B	B Reactor, one of the original eight Hanford Production Reactors.
	Barn	Nuclear cross section unit. Equal to 10^{-24} cm^2.
	Burnup	Specific thermal energy generated per mass of nuclear fuel.
	BWR	Boiling water reactor, one of the main types of commercial nuclear reactors.
	CANDU	Canadian heavy water reactor.
	Ci	Curie. A unit of radioactivity equal to 3.7 x 10^{10} disintegrations per second.
13	*Cross Section*	*The apparent cross-sectional area of a nuclide. The units are barns - 10^{-24} cm^2.*
13	*Cross-Section Library*	*A computer file of cross-section numbers.*
33	D	D Reactor. One of the original eight Hanford Production Reactors. *DR, another of the eight original, are also located at D-Area.*
	F	F Reactor. One of the original eight Hanford Production Reactors.
	Gwd/Mtu	Gigawatt-days per metric ton uranium (nuclear fuel burnup unit).
	Gwd/Ton	Gigawatt-days per short ton (2000 lbs) uranium (nuclear fuel burnup unit).
	HEDR	Hanford Environmental Dose Reconstruction (Project).
	HPR	Hanford Production Reactor.
33	K	K Reactor. *Applies to two reactors, KE and KW, two* of the original eight Hanford Production Reactors.

LWR	Light Water Reactor.
MCC	Materials Characterization Center. Pacific Northwest Laboratory center for characterization of nuclear materials.
Mwd/Mtu	Unit of measurement for burnup of fuel in a nuclear reactor; days of reactor operation at a certain power level (megawatt-days) per metric ton of uranium in the fuel.
Mwd/Ton	A unit of measurement for burnup of nuclear fuel; megawatt-days per short ton (2,000 lb) of uranium in the fuel.
N	N Reactor, the ninth Hanford Reactor. Unlike the previous eight Hanford reactors, N Reactor recirculated the primary cooling water.
PNL	Pacific Northwest Laboratory.
PWR	Pressurized Water Reactor.
QA	Quality Assurance.
REA	Ridihalgh, Eggers & Associates. Manufacturers of casks for spent nuclear fuel.
13 *Resonance Integral*	*The integral over neutron energy of the product of normalized neutron flux and the absorption cross section. The integration range is from thermal neutron energy to infinity. It is a measure of the rate of absorption at neutron energies above the thermal range.*

CONTENTS

FIGURES

TABLES

1.0 INTRODUCTION

The ORIGEN2 computer code (Croff 1980) is the primary calculational tool for computing isotopic source terms for the Hanford Environmental Dose Reconstruction (HEDR) Project. The ORIGEN2 code computes the amounts of radionuclides that are created or remain in spent nuclear fuel after neutron irradiation and radioactive decay have occurred as a result of nuclear reactor operation. ORIGEN2 was chosen as the primary code for these calculations because it is widely used and accepted by the nuclear industry, both in the United States and the rest of the world. Its comprehensive library of over 1600 nuclides includes any possible isotope of interest to the HEDR Project. It is important to evaluate the uncertainties expected from use of ORIGEN2 in the HEDR Project because these uncertainties may have a pivotal impact on the final accuracy and credibility of the results of the project.

The work described in this report was conducted in accordance with the requirements of ANSI/ASME NQA-1, 1986 Edition, Quality Assurance Program Requirements for Nuclear Facilities, as interpreted by the Pacific Northwest Laboratory (PNL) Quality Assurance (QA) program.

34 *The ORIGEN2 code was received from Oak Ridge National Laboratory (ORNL) in accordance with software control standards which required extensive comparisons with cases run on the code at ORNL to ensure that results produced with the code are identical to those at PNL.*

All procedures used to support this report were written and controlled in accordance with PNL QA program requirements. Records that support the data in this report were created and stored in accordance with applicable HEDR record control requirements.

Drafts of this document underwent internal independent technical review. Review comments were satisfactorily resolved with no controversial resolutions to the comments.

2.0 ACCURACY OF ORIGEN2

The ORIGEN2 computer code has been used extensively in the commercial light water reactor (LWR) industry to calculate the characteristics of spent nuclear fuel. The results of ORIGEN2 calculations have provided source terms used in the design of spent fuel processing, transportation, and handling facilities as well as LWR design itself. Hence, comparisons of ORIGEN2 predictions with measured quantities are predominantly for LWR fuel. Three types of measured quantities are available for validation purposes:

1. calorimetric measurements of spent fuel decay heat
2. destructive measurements of spent fuel isotopics
3. gamma and neutron source measurements of spent fuel.

These quantities were measured for fuels that underwent higher burnups than the fuel in the Hanford Production Reactors (HPRs).

2.1 SPENT FUEL CALORIMETRIC MEASUREMENTS

28 Calorimetric measurements of irradiated LWR fuel and the corresponding ORIGEN2 predictions of those measurements are shown in Table 2.1. The time period between discharge of spent fuel from the reactor and measurement of its

TABLE 2.1. Measured Versus Predicted LWR Spent Fuel Decay Heat

14, 15

Reactor	Type	Number of Measurements	*(Calc.-Meas.)/Measured (%)* Average	Standard Deviation	Reference
Turkey Point	PWR[(a)]	7	5.4	3.5	Schmittroth (1984)
Cooper Station	BWR[(b)]	81	-0.5	5.4	Wiles and Lombardo (1986)
Monticello	BWR	26	-11.8	12.5	McKinnon et al. (1986)
Overall Measurements		114	-0.85	4.8[(c)]	

(a) Pressurized Water Reactor.
(b) Boiling Water Reactor.
(c) Standard deviation of 114 measurements.

decay heat was from 3 to 10 years. In this time period, the major isotopes contributing to decay heat are strontium-90 (35%) and cesium-137 (48%). Radioactive decay of actinides accounts for less than 10% of the total decay heat. ORIGEN2 calculations predict spent LWR fuel decay heat rather well except when compared with the Monticello *reactor* measurements, where difficulties with calorimeter performance and calibration may have resulted in suspect data.

2.2 SPENT FUEL ISOTOPIC MEASUREMENTS

35 Comparisons of LWR spent fuel isotopic measurements with ORIGEN2 predictions are shown in Table 2.2. These comparisons are a stringent test of the ORIGEN2 code's ability to calculate HPR *isotopic content* with characteristic burnups below 1.0 Gwd/Mtu, because nuclear data used by the calculation determines the computed rate at which radionuclides are formed or removed. If the data contain errors, then as periods of irradiation or decay increase, the discrepancy between the calculated and the measured amount can only increase. The irradiation time and the decay time (i.e., the time of discharge from the reactor to the time of measurement) for this LWR spent fuel is typically 3 years and 5 years, respectively, compared with 75 days and 90 days, respectively, for HPRs.

36 Table 2.2 shows that two fission product isotopes, selenium-79 and tin-126, are grossly overcalculated by ORIGEN2; however, these are not critical isotopes for the HEDR Project *(Napier 1990)*. Carbon-14, which originates primarily from a high-energy n,p reaction on nitrogen-14 that is present in the fuel in trace amounts, is undercalculated by 10%. A lesser source of carbon-14 results from neutron capture by carbon-13, also a uranium fuel impurity. The uncertainty in the initial nitrogen-14 and carbon-13 contents of the fuel contributes substantially to the underestimation of carbon-14 by ORIGEN2.

2.3 GAMMA RAY AND NEUTRON SOURCE MEASUREMENTS

Comparisons of the radiation doses emitted from gamma and neutron sources with measured radiation dose data are rarely done for an unshielded source.

TABLE 2.2. Measured *Isotopic Content* Compared to ORIGEN2 Predictions (averages based on six sample measurements)[a]

Isotope	(Calculated - Measured)/Measured(%) Average	Standard Deviation
Carbon-14	-13.9	4.5
Selenium-79	616.3	88.1
Strontium-90	-1.5	1.1
Technetium-99	19.3	19.7
Tin-126	378.0	52.2
Iodine-129	-1.9	11.7
Cesium-135	4.7	4.8
Cesium-137	-3.8	1.5
Uranium-234	1.1	18.3
Uranium-235	15.9	12.2
Uranium-236	-5.5	1.4
Uranium-238	-1.0	0.6
Plutonium-238	1.6	10.4
Plutonium-239	11.0	7.7
Plutonium-240	1.4	1.6
Plutonium-241	10.2	7.6
Plutonium-242	-10.7	3.6
Neptunium-237	21.5	16.9
Americium-241	-0.8	11.2
All Isotopes Except Selenium-79 and Tin-126	2.5	7.9[b]

(a) Guenther and Blahnik (1988a,b).
(b) Standard deviation over all isotopes except selenium-79 and tin-126.

Usually, measured dose rates at the outside of the shielded structure are compared with ORIGEN2 source calculations plus the calculated attenuation through the radiation shield. It is difficult to separate source term errors from errors in the shielding calculations, which are generally much larger.

In a typical example of this method, gamma ray and neutron dose rates were measured and compared to dose rate predictions using ORIGEN2 to calculate the source terms (Wiles and Lombardo 1986). The transport codes QAD (ORNL 1977) and DOT (Rhoades and Childs 1982) were used to calculate neutron and

gamma ray attenuation through the REA[a] spent fuel shipping cask loaded with spent BWR fuel assemblies (Wiles and Lombardo 1986).

Figure 2.1 shows the calculated and measured gamma dose along the side of the cask (Wiles and Lombardo 1986). Agreement between the calculated and measured value along the body of the cask is within a factor of 2. The major differences arise at the ends of the cask, where the cask geometry in the transport codes is inadequately modeled.

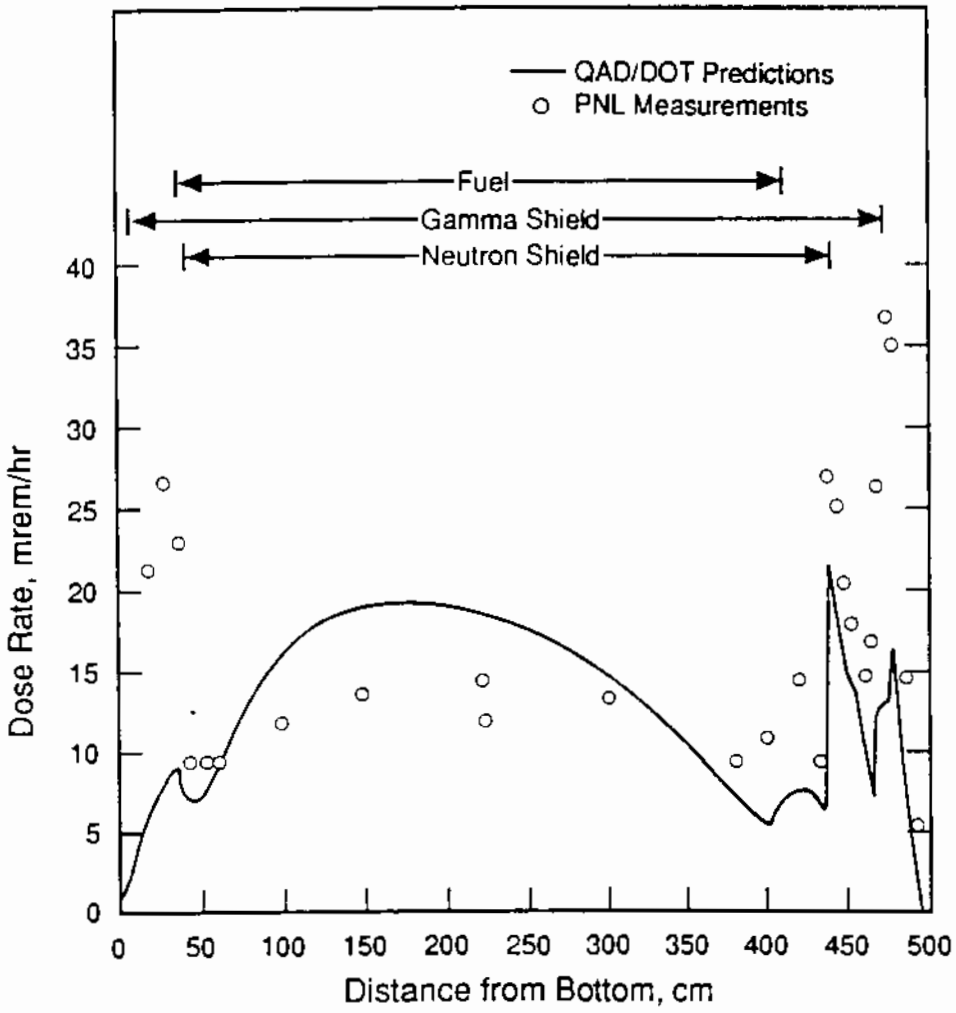

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FIGURE 2.1. Measured and Calculated Gamma Dose Rates for the REA Cask

(a) Ridihalgh, Eggers, and Associates, manufacturers of casks for spent nuclear fuel.

Figure 2.2 shows the neutron dose rate. Again agreement is within a factor of 2 along the body of the cask, with major differences at the end due to transport modeling inaccuracies. The comparisons show that there are no gross errors for the gamma-emitting fission products, or for the actinide neutron emitters in the ORIGEN2 calculations. The cooling times of the spent fuel in the REA cask are of the order of a few years; therefore, short half-life emitters are not validated by this comparison. The tested accuracy of ORIGEN2 in these very stringent (high burnup) comparisons is generally considered to be acceptable by the nuclear industry.

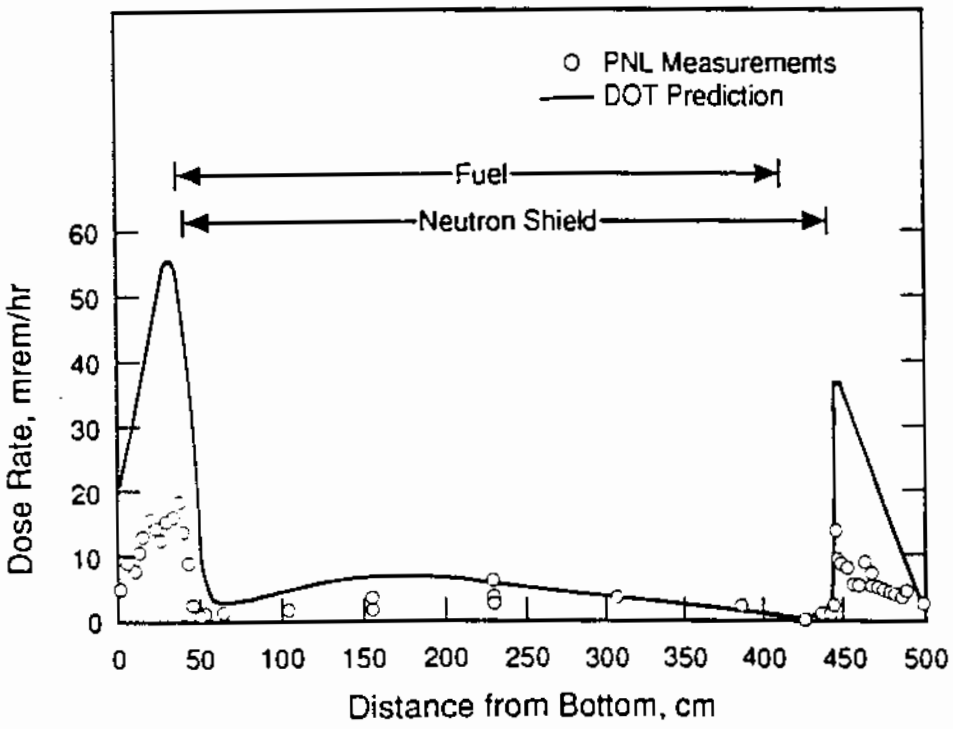

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FIGURE 2.2. Measured and Calculated Neutron Dose Rates for *the* REA Cask

3.0 SOURCES OF UNCERTAINTY IN ORIGEN2 CALCULATIONS

There are three primary sources of uncertainty in an ORIGEN2 calculation:

- basic nuclear data uncertainty in neutron cross sections, radioactive decay constants, energy per fission, and fission product yields
- calculational uncertainty due to input data
- code uncertainties (i.e., numerical approximations, and neutron spectrum-averaged cross-section values from the code library).

3.1 NUCLEAR DATA

18, 19 The nuclear data uncertainty is generally quite small compared with the calculational uncertainty. Table 3.1 shows the estimated uncertainty in the key neutron cross sections and resonance integrals for uranium-235, uranium-238, and plutonium-239 (Mughabghab and Garber 1972). Table 3.1 includes both resonance integrals and thermal neutron cross sections to cover the complete neutron energy range of reactor neutrons. *Experimental error estimates are shown.* The average uncertainty *(ratio of experimental error to the experimental value)* over all entries in Table 3.1 is 2.5%, and the largest is 10%.

Measurements of radioactive decay constants are relatively precise; hence, the uncertainty in the decay constant value is low. For 26 isotopes of

TABLE 3.1. Resonance Integrals and Thermal Neutron Cross Sections for Key Isotopes

	Resonance Integral (barns)		Thermal Cross Section (barns)	
Isotope	Capture	Fission	Capture	Fission
Uranium-235	144 ± 6	275 ± 5	98.6 ± 1.5	582.2 ± 1.3
Uranium-238	275 ± 5		2.7 ± 0.02	
Plutonium-239	200 ± 20	301 ± 10	268.8 ± 3.0	742.5 ± 3.0

primary interest to the HEDR Project, the standard deviation of measured values of the decay constant was 0.6% of the average measured value. The measured values were reported by Lederer and Shirley (1978).

20 The quoted fission product yields are known to be within 6% (Unik and Gindler 1971). ORIGEN2 uses the regression on atomic number and mass reported by Unik and Gindler (1971) to calculate energy per fission. The results of this calculation are valid to within 1%. Generally, most of the nuclear data uncertainties result from random errors. Individually, the degree of these errors is substantially less than 6%, and because the errors are *independent*, they are not cumulative.

3.2 INPUT DATA

37, 38 The most important input parameters are 1) burnup, 2) cooling time, and 3) power density. Each HPR process tube was equipped with a flow-measuring orifice, or venturi, and an individual outlet temperature thermocouple. Thus, the tube power (the product of flow and temperature increase) was known at all times during operation, and individual tube burnups were determined by integration of the tube power over time. *The tube power uncertainty is essentially the measurement uncertainty of flow and temperature, which would be in the 5% range*. Within the process tube, fuel element exposures depended on the average front-to-rear (axial) neutron flux distribution during the time of residence. Although the axial distribution was not well known, it is irrelevant because all of the fuel elements within a process tube were discharged into the storage basin and all position information within the process tube was lost at that point. Thus, the burnup used as input to ORIGEN2 reflects a well-known average around which the degree of burnup for the fuel elements within the batch is widely distributed. For isotopes with linear rates of increase with increasing burnup, the distribution of fuel element burnup around the mean is of little concern *because the product of this value and the batch weight accounts for the isotope inventory accurately, even though parts of it were well above and below the mean prior to dissolution.* In other

cases, severe departures from linearity may introduce some small departures from the computed average isotopic inventory upon dissolution of the irradiated fuel.

39 The cooling time of the discharged fuel batch, the second important input parameter listed above, *is of importance only for short-lived isotopes (half-lives substantially less than a year). Uncertainty in cooling time can have a substantial effect. For example, iodine-131 (8-day half-life) content changes by 8% for each day of change in cooling time.*

12, 21, 24 *Power density uncertainty is less than burnup uncertainty inherently. However, unlike burnup, the average power of a batch of fuel discharged was never recorded. Hence the power density of a discharged fuel batch must be calculated from a knowledge of the pile power history. The extra step involves an additional uncertainty. This uncertainty would apply not only to ORIGEN2, but to <u>any</u> code or algorithm used to calculate the isotopic content of spent fuel. If ORIGEN2 is used to calculate the isotopic content of material in a given process tube, then the input uncertainty is less than 5%. If ORIGEN2 is used to calculate the isotopic content of a batch of fuel, then the uncertainty is larger than 5%. A general figure of merit for all applications would be 10%. However, each application could vary and would have to be evaluated on an individual basis.*

3.3 ORIGEN2 CODE UNCERTAINTIES AS APPLIED TO HEDR CALCULATIONS

25, 40 ORIGEN2 uses a variety of numerical techniques to solve the *linear* system of coupled differential equations that determine the buildup and burnout of nuclides over time. With appropriately chosen input time intervals, the solutions generated by the code are accurate *approximations to the analytic solutions.*

The remaining source of calculational uncertainty is the spectrum-averaged neutron cross-section library. In ORIGEN2, the user is presented with a choice of various cross-section libraries. Each library contains spectrum-averaged cross-section values for a specific reactor core type. The

neutron spectrum is very sensitive to the type of reactor. Figures 3.1 and 3.2 show the neutron spectrum of a PWR fueled with slightly enriched uranium (Croff et al. 1978), and a CANDU heavy water reactor (Croff and Bjerke 1980), respectively. There is a vast difference between the neutron spectra exhibited by these two types of reactors. The heavy water reactor spectrum has a peak at the neutron thermal equilibrium energy around 0.025 eV. The PWR spectrum has a much smaller thermal peak and a larger maximum near the fission neutron source energy of 1 MeV. The neutron spectrum for an HPR is between these two extremes.

22 There are no ORIGEN2 libraries for the original eight HPRs; however, a set of ORIGEN2 cross-section libraries has been generated for N Reactor. Comparison calculations were performed for several isotopes of potential

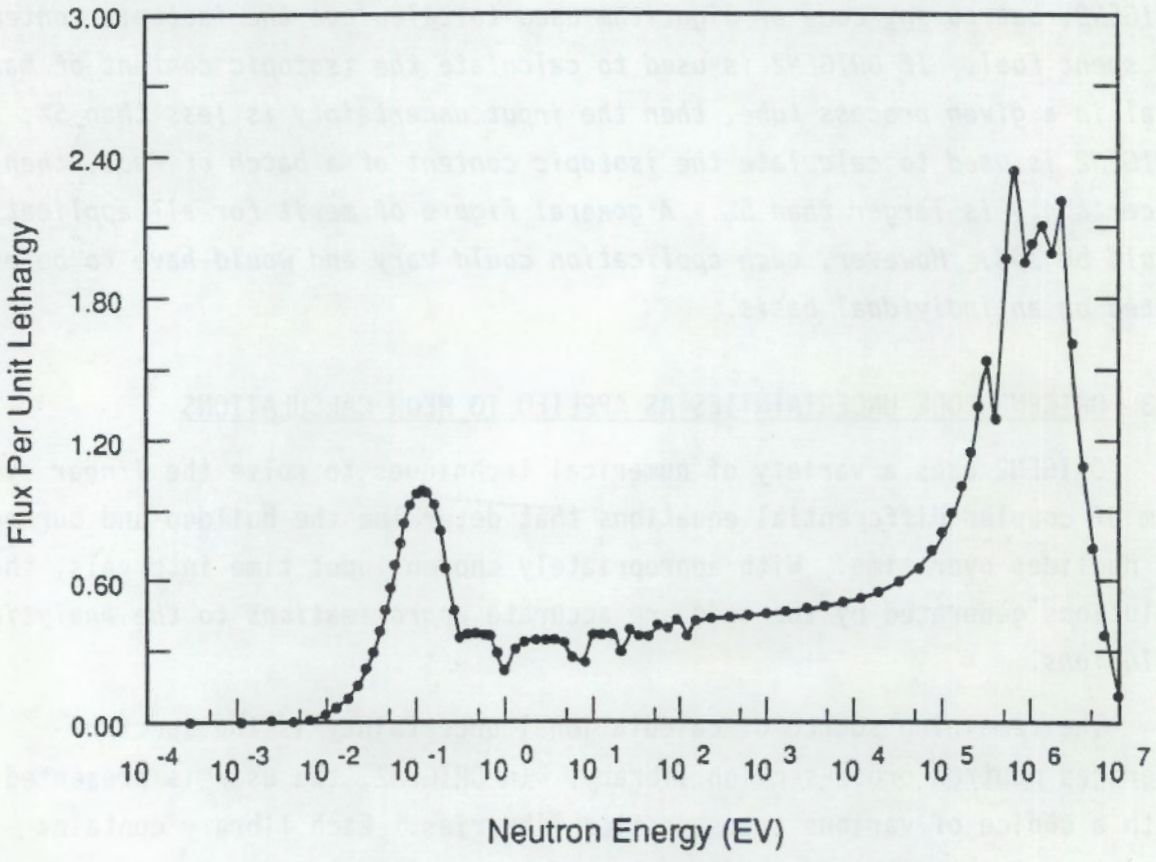

FIGURE 3.1. Neutron Spectrum of a PWR Core

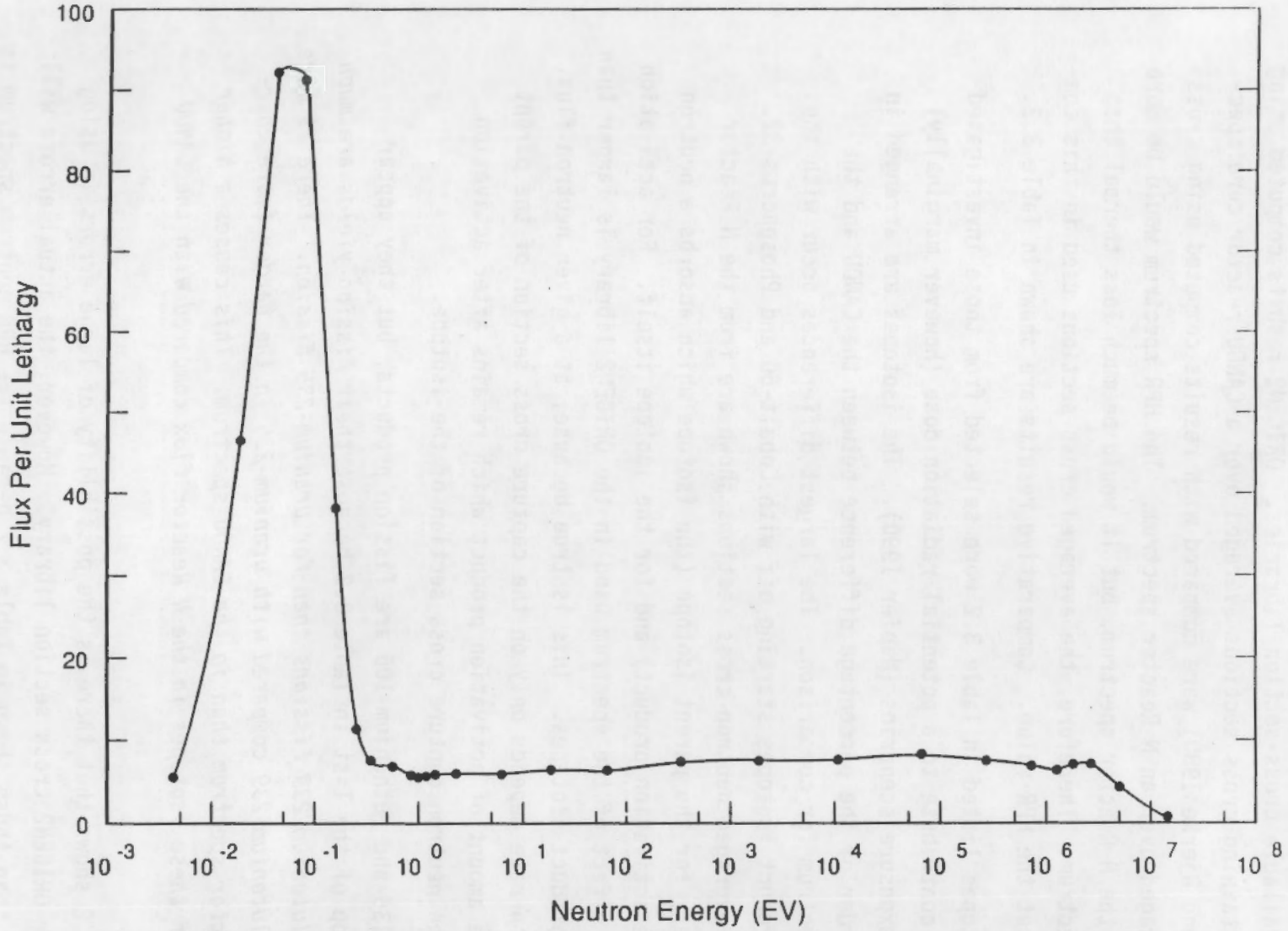

FIGURE 3.2. Neutron Spectrum of a Heavy Water (CANDU) Reactor Core

interest to the HEDR Project to evaluate the *differences* that would result from using available cross-section libraries. ORIGEN2 results computed using a library containing cross sections averaged over a CANDU reactor core spectrum (Croff and Bjerke 1980) were compared with results computed using cross sections averaged over an N Reactor spectrum. The HPR spectrum would be more thermal than the N Reactor spectrum, but it would be much less thermal than the CANDU spectrum. Therefore, the averaged cross sections used in this comparison bracket the HPR value. Comparative results are shown in Table 3.2.

The isotopes listed in Table 3.2 were selected from those investigated and found to contribute to a potential radiation dose (however marginally) for various exposure scenarios (Napier 1990). The isotopes are arranged in decreasing order of the percentage difference between the CANDU and the N Reactor spectrum for comparison. The largest differences occur with the activation product isotopes starting off with Cobalt-60 and Phosphorus-32. The spectrum-averaged neutron cross sections shown are from the N Reactor ORIGEN2 library for the parent isotope (the isotope which absorbs a neutron to create the activation product) and for the isotope itself. For activation products the effect of the spectrum used in the ORIGEN2 library is larger than for fission product isotopes. This is true because, at a given neutron flux, the activation rate depends only on the capture cross section of the parent isotope. The amount of activation product which remains after activation depends on the neutron capture cross section of the isotope.

26 Cesium-136 and ruthenium-106 are fission products, but they appear nearer the top of the list in Table 3.2 because *their fission yields are much higher for plutonium-239 fissions than for uranium-235 fission. There is more fission in plutonium-239 compared with uranium-235 in the harder (more energetic) N Reactor spectrum than in the CANDU spectrum. This causes a higher production of these isotopes in the N Reactor flux compared with the CANDU flux.*

29 Table 3.2 shows that there is the possibility of large errors in using the N Reactor ORIGEN2 cross section library. However, the actual errors will be much less than those shown in Table 3.2 because the HPR neutron spectrum is

TABLE 3.2. Percent Differences in Ci Content Averaged over Discharged Burnups Between 100 and 1000 Mwd/Ton for N Reactor and CANDU Reactor Spectrum ORIGEN2 Libraries

	Isotope	(C-N)/N (%)[a]	Source[b]	Half-Life[c]	N Reactor ORIGEN2 Library Neutron Capture Cross Section (barns) Parent	Isotope
	Cobalt-60	104.9	A	5.3 yr	2.990	0.375
	Phosphorus-32	53.0	A	14.28 da	0.017	0.000
	Scandium-46	52.9	A	83.8 da	1.544	0.736
	Chromium-51	50.9	A	27.7 da	1.470	0.074
	Arsenic-76	-40.0	A	26.3 hr	2.150	0.000
	Copper-64	36.5	A	12.7 hr	0.485	0.000
	Cesium-136	-33.8	FP	13.1 da		1.340
	Iron-59	32.4	A	44.5 da	0.112	0.000
	Ruthenium-106	-28.3	FP	373 da		0.089
	Manganese-56	25.1	A	2.6 hr	1.520	0.000
	Gallium-72	-20.0	A	13.9 hr	1.125	0.000
	Zinc-65	19.5	A	243.8 da	0.098	0.000
	Neptunium-239	-19.2	ACT	2.35 da		11.300
	Sodium-24	-17.9	A	14.97 hr	0.053	0.000
	Plutonium-239	17.3	ACT	24,000 yr		106.700
	Tellurium-129m	-12.2	FP	33.4 da		0.290
	Samarium-153	-9.6	FP	46.7 hr		89.800
	Ruthenium-103	-7.6	FP	39.2 da		2.290
	Strontium-90	7.0	FP	29.0 yr	0.053	0.087
	Strontium-89	6.6	FP	50.5 da	0.012	0.001
	Yttrium-91	5.9	FP	64.0 hr	0.433	0.165
	Thorium-232	-5.2	ACT	1.4×10^{10} yr		3.050
6, 41	*Iodine-129*	*4.4*	*FP*	*1.6×10^{7} yr*		*3.220*
	Zinc-69m	4.3	A	13.8 hr	0.157	0.000
	Iodine-131	-3.6	FP	8.04 da		0.322
	Cerium-144	3.6	FP	284 da		0.149
	Cerium-143	3.3	FP	33.0 hr		1.760
	Zirconium-95	2.7	FP	64.0 da	0.015	1.392
	Lanthanum-140	2.5	FP	12.8 da		2.210
	Tellurium-132	-2.3	FP	78.2 hr		0.000
	Barium-140	2.3	FP	40.3 hr		0.528
	Iodine-132	-1.7	FP	83 min		0.000
	Zirconium-97	0.8	FP	16.8 hr	0.177	0.000
41	*Hydrogen-3*	*-0.8*	*FP*	*12.3 yr*		*0.000*
	Cesium-137	0.7	FP	30.2 yr		0.026
	Molybdenum-99	0.6	FP	65.9 hr	0.237	1.013
	Xenon-133	0.2	FP	5.25 da		24.400

TABLE 3.2. (contd)

Isotope	(C-N)/N (%)[a]	Source[b]	Half-Life[c]	N Reactor ORIGEN2 Library Neutron Capture Cross Section (barns) Parent	Isotope
Iodine-133	0.1	FP	5.25 da		0.000
41 *Technicium-99*	*-0.1*	*FP*	*2.1×10^5 yr*		*8.193*

(a) C = CANDU, N = N Reactor.
(b) A = Activation Product, FP = Fission Product, ACT = Actinide.
(c) Walker, F.W. et al. 1977. Chart of the Nuclides, 12th Edition, General Electric Company.

much closer to the N Reactor spectrum than *to* the CANDU spectrum, which was used for Table 3.2 precisely because it does provide an estimate of the maximum error. *Of most concern are the activation product nuclides important to the water pathway. Calculations using ORIGEN2 for these source term nuclides must include an estimate of the irradiation time that the parent isotope was exposed to the reactor neutrons. Comparison with measurements will be required to do this type of calculation. ORIGEN2 errors will be taken into account in the normalization to measurement process.*

Although no ORIGEN2 library is available for HPRs, an estimate for how large the uncertainties might be within the maximum range shown in Table 3.2 can be obtained by comparing the computed neutron spectrum for N Reactor with various HPR neutron spectra. Neutron spectra for N Reactor, the older HPR lattices, and K Reactor were computed using the WIMS-D code (Askew et al. 1966).

The single most important reactor design parameter in determining the form of the reactor neutron spectrum is the moderator-to-fuel ratio. The original Hanford reactors (i.e., B, D, DR, F, and H Reactors) were built with a 8.375-inch spacing between process tubes (lattice pitch). The K Reactors were built with a 7.5-inch pitch. The N Reactor was built with a 8.462-inch lattice, but with 10% volume void space in the graphite stack, which, along

with an increase in the fuel volume, resulted in a lower moderator-to-fuel ratio than for the other HPR lattices. Thus, in terms of the relative amount of graphite moderator to fuel, the B, D, and F Reactor lattices would have the most moderator, the K Reactor lattice would be next, and the N Reactor lattice would have the least. Accordingly, the B, D, and F Reactor lattices would have the most thermal spectrum, the K Reactor lattice would be less thermal, and the N Reactor lattice would be the least thermal (most energetic) of all the HPRs.

The WIMS-D computed spectra are shown in Figure 3.3. They are plotted as functions of neutron lethargy, u, which is a convenient variable to cover the energy range of 10 MeV to 0 eV. Neutron lethargy is defined as the natural logarithm of the ratio of the highest energy to the energy at which the lethargy is being calculated: $u = \ln(E_0/E)$. Thus, fission neutrons are born at energies up to 10 Mev (taken as the base of the lethargy scale, E_0) or zero lethargy, and slow down to thermal energies around 0.005 eV at 21.4 lethargy units. Figure 3.3 shows spectra for the predominant natural uranium loadings of the B, D, and F Reactor lattices, the K Reactor lattice, and the 0.947 wt% enriched N Reactor lattice. They are typical reactor spectra with a maximum at the upper neutron energies due to fission source neutrons, and another maximum at 18 lethargy units (0.152 eV) at thermal neutron energy. The N Reactor lattice spectrum is the hardest (least thermal) with the highest maximum at source neutron energy, and the lowest maximum at thermal neutron energies. The B, D, and F Reactor lattices' spectra are softer (more thermal) than the K Reactor lattice and much softer than the N Reactor lattice. All spectra shown in Figure 3.3 are normalized to unit area for direct comparison. There is little difference between the B, D, and F Reactor lattices spectra and the K Reactor lattice spectrum.

In order to arrive at a single comparison figure for each of the spectra, a spectrum-averaged cross section was calculated for a unit 1/v absorber (unit cross section value at 0.0253 eV and proportional to the reciprocal neutron velocity). The results are shown in Table 3.3. There is a 5% difference between the two older reactor lattice spectra, while the N Reactor lattice is almost half the B, D, and F Reactor lattice values. This

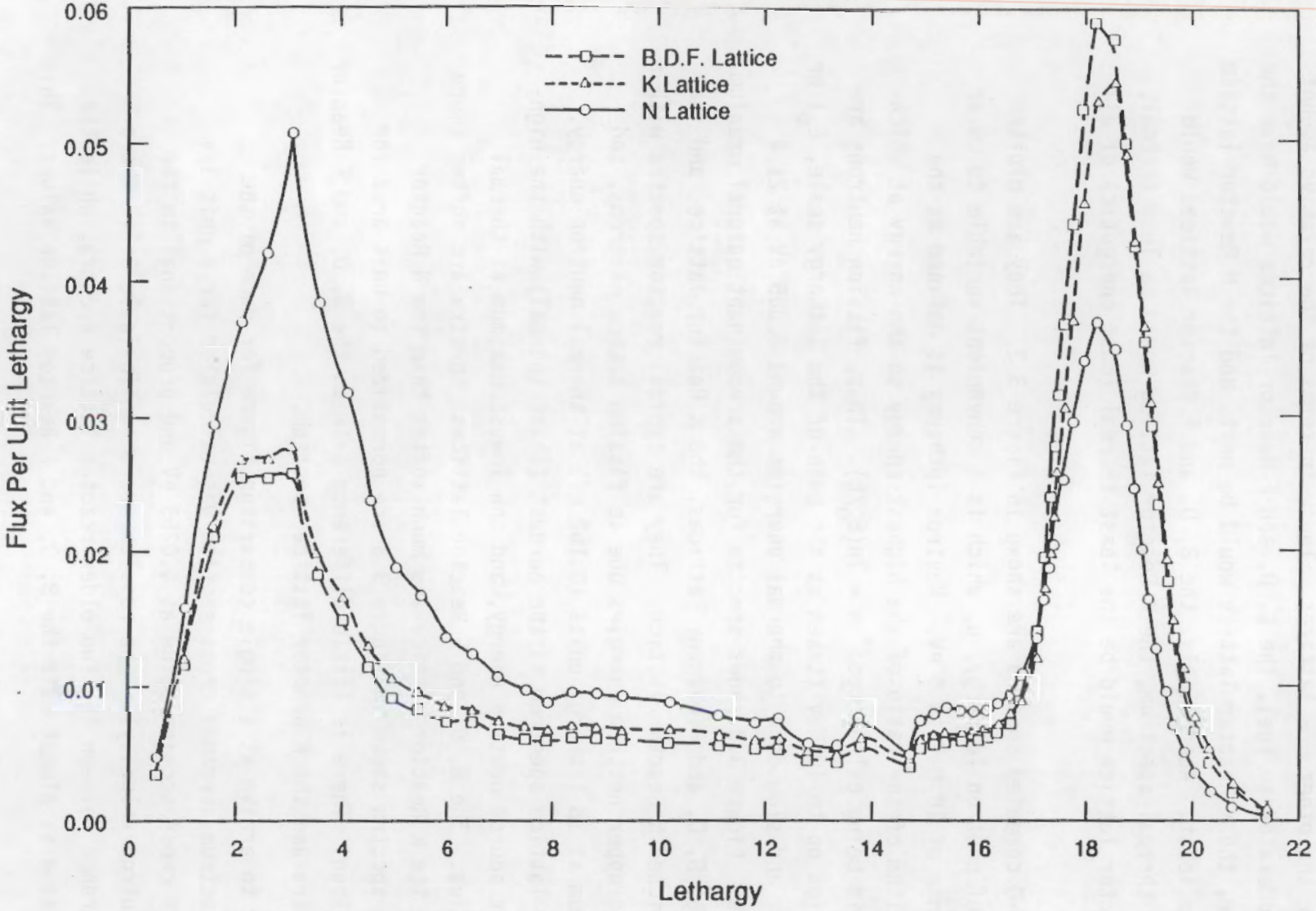

FIGURE 3.3. WIMS-D Calculated Neutron Spectra

TABLE 3.3. Spectrum-Averaged Unit 1/v Cross Section

Spectra	Spectrum-Averaged Cross Section (barns)
B, D, F Reactor Lattice	0.2109
K Reactor Lattice	0.2017
N Reactor Lattice	0.1094

result indicates that while the activation product differences shown for cobalt-60 and phosphorus-32 in Table 3.2 may not be as large, they are still substantial.

4.0 IMPACT OF USING ORIGEN2 TO CALCULATE HEDR SOURCE TERMS

The results shown in Table 3.2 indicate that the uncertainty due to using the N Reactor cross-section library to calculate HEDR source terms is greatest for activation products and fission products with high neutron absorption cross sections. The plutonium isotopes in particular, and other actinides in general, are next in uncertainty. Fission products that exhibit low neutron absorption are the least uncertain.

The source-term nuclides listed in Table 3.2 are those analyzed and reported by Napier (1990). Of those nuclides reported, iodine-131, ruthenium-106, and ruthenium-103 contribute greater than 99% of the potential radiation dose from releases to the atmosphere. Phosphorus-32, neptunium-239, zinc-65, arsenic-76 and copper-64 contribute over 70% of the potential radiation dose resulting from releases to the Columbia River. For most exposure scenarios, the sensitivity of the fission products to the neutron spectra is low, except for cesium-136 and ruthenium-106, which have a large *difference in the uranium-235 and plutonium-239 fission product yield.*

To increase the source-term accuracy, ORIGEN2 HPR libraries could be computed just as they were developed for N Reactor. This process would improve the accuracy of activation product calculations, which are the most sensitive to the particular neutron spectra used in the ORIGEN2 cross-section libraries. The calculation of ORIGEN2 cross-section libraries for HPRs would start with a high-order calculation of the HPR lattice neutron spectrum using the WIMS code. The spectrum would then be used with neutron cross sections as functions of neutron energy from the appropriate ENDF library[a] to produce cross-section values averaged over the computed HPR lattice neutron spectrum in the ORIGEN2 library format.

(a) ENDF/B-IV Library Tapes 401-411 and 414, available from the National Neutron Cross Section Center, Brookhaven National Laboratory (December 1974); ENDF/B-V Library Tapes 514, 521, and 522, available from the National Neutron Cross Section Center, Brookhaven National Laboratory (July 1979).

5.0 CONCLUSIONS

Uncertainties in the estimated radiation doses that people could have received from nuclear operations at the Hanford Site since 1944 originate, in part, from the uncertainty in the source terms used. ORIGEN2 calculations of activation product sources in the Columbia River pathway may be used to supplement the extensive river monitoring data. ORIGEN2 calculations may also be used to estimate air pathway sources. The uncertainties to be expected when applying ORIGEN2 to HEDR source terms have been discussed separately and are on the order of 10%, except for uncertainty in cooling time estimates for short-lived nuclides, and in using the N Reactor cross-section library in place of an HPR library. Table 3.2 provides an estimate of the maximum uncertainty from this source.

43, 44 *The uncertainty in the input parameters (specific power, burnup, and cooling time) would be common to any code or algorithm used to estimate radionuclide source terms. However, the uncertainties in using the N Reactor cross-section library in HPR calculations are specific to the ORIGEN2 code. These uncertainties will be addressed on a nuclide-by-nuclide basis or by creating an HPR ORIGEN2 library, whichever represents the most efficient use of HEDR resources. If an HPR library is not created, then the uncertainties in Table 3.2 provide a basis for determining whether a particular nuclide requires special treatment in the ORIGEN2 source term estimation.*

For example, the important pathway nuclides iodine-131, ruthenium-103, and ruthenium-106 have maximum uncertainties from Table 3.2 of 3.6%, 7.6% and 28.3%. Compared with uncertainties in the release fractions and atmospheric transport, iodine-131 and ruthenium-103 uncertainties are negligible, and special provisions would not be required in using ORIGEN2 to calculate source terms for these nuclides. However, the 28.3% uncertainty for ruthenium-106 could be eliminated by a correction factor applied to the ORIGEN2 input.

Important water pathway nuclides such as phosphorus-32 and neptunium-239 are activation products. Although their Table 3.2 uncertainties are rather large (53% and 19%, respectively), a correction factor would not be needed because activation product calculations require normalization to measured data; the reactor holdup time of the parent nuclide is not well known. The

normalization process involves finding an effective holdup time that provides an ORIGEN2 answer which agrees with the measured value. The process water traversed the central region of an HPR in less than 2 seconds. The effective holdup time will generally be longer because it includes time for the parent nuclide to lodge and be released from films on process tubes and fuel elements. Using different activation cross sections from different ORIGEN2 libraries will change these holdup times but cannot alter the fact that the results of the calculation agree with the measured values by definition.

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APPENDIX

SUMMARY OF TSP COMMENTS AND BATTELLE RESPONSES

APPENDIX
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Document Number PNL-7223 HEDR Document Title Uncertainties in Source Term Calculations Generated by the ORIGEN2 Computer Code for Hanford Production Reactors

Comment Number	Commenter	Page, Paragraph	Comment Summary	Resolution
1.	J. Stohr		Are measurements of iodine-131 or ruthenium-103/106 available for comparison with the ORIGEN2 predictions of Table 2.2?	NA - No measurements are available that would allow us to check the total amount of iodine-131 in the fuel under dissolution. ORIGEN2 would be used to provide an iodine-131 content in terms of Ci/T, which, when used with the number of metric tons being dissolved, would give an amount of iodine-131. The amount released would then be calculated assuming a release factor. The uncertainty inherent in using ORIGEN2 without an HPR library is shown to be less than 3.6% (Table 3.2). This is too small to be tested in view of the larger uncertainties in the amount dissolved and in the release factor.
2.	JS		Add info on time/money necessary to convert libraries for activation products.	NA - A past estimate is that ~$70K would be required to provide an HPR library for all nuclides in the ORIGEN2 library.

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Comment Number	Commenter	Page, Paragraph	Comment Summary	Resolution
3.	D. Barth		Add section summarizing production amounts by year with uncertainties for the most important radionuclides: I-131, Ru-106, Ru-103, P-32, Np-239, Nz-65, As-76, and Cu-64.	NA - Out of the scope of this document.
4.	R. Cook		Make units in Figures 3.1, 3.2 and 3.3 consistent, energy or lethargy. If energy, express flux on the vertical scale as "flux per unit energy," not flux per unit lethargy. Add units for flux.	NA - The units of Figures 3.1 and 3.2 were taken directly from ORNL documents. To change into consistent units would require reducing the figure to numbers and recalculating them as flux per unit lethargy versus lethargy. A visual comparison of Figures 3.1 and 3.2 suffices to show that the neutron spectra are very different regardless of the units. Figure 3.3 is based on our WIMS calculation and was done with consistent units.
5.	RC		Review magnitude of fluxes on Figures 3.1, 3.2, and 3.3 to ensure accuracy; e.g., peak thermal flux for CANDU reactors in Figure 3.2 is nearly 100 times that for PWRs in Figure 3.1.	NA - The ORNL references are believed to be accurate. The CANDU spectrum is very thermal.
6.	RC		Supplement Table 3.2 to include information on iodine-129.	Iodine-129 information was added.

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Comment Number	Commenter	Page, Paragraph	Comment Summary	Resolution
7.	RC		Modify cross section library for ORIGEN2 for the HPRs for key isotopes in the generation of iodine-131 and iodine-129 and their precursors.	NA - If an HPR library is generated in Phase III, all isotopes will be updated, including precursors.
8.	RC		Estimate atom ratios of iodine-131 to iodine-127 and iodine-129 at discharge of the fuel from the reactor as a function of burnup and the uncertainties, as well as the ratios at the time of discharge out the stacks. Ratios help determine uptake of isotopes into thyroids and enable calculation of iodine-129 dose to thyroid given iodine-131 concentrations in food and water.	NA - Out of the scope of this document.
9.	RC		List data on burnup from operating data for each reactor for each irradiation cycle during the period up to 1949.	NA - Out of the scope of this document. Our conclusions are valid for the entire range of Hanford HPR burnups (0-1000 Mwd/Ton).
10.	K.J. Kopecky		Define uncertainty; used in report in two different contexts [see comments 11, 18, and 22].	NA - We chose not to define the term "uncertainty" because it is used in many different technical senses. In most instances, it means the maximum range of a difference. For example, in paragraph 2 of the Summary, "nuclear data--less than 10%" means that if

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Comment Number	Commenter	Page, Paragraph	Comment Summary	Resolution
				you go to Table 3.1, the maximum percentage uncertainty is for plutonium-239. It is 20 parts out of 200, or 10%. More precise definitions of ORIGEN2 uncertainty are beyond the scope of the document--to provide an idea of the relative uncertainty to be expected from using ORIGEN2 to calculate HEDR source terms.
11.	KJK	Summary, para. 2	Define uncertainty. Do the phrases "less than 10%", "within 10%" and "up to a factor of 2" refer to coefficients of variation, standard deviations, ranges, or what?	Text was clarified.
12.	KJK	Summary, para. 2	How was "within 10%" obtained for "code input information?" Section 3.2 only states that after uncertainties in isotopic inventories after 1948 "should be low." And what about before 1948?	Section 3.2 was revised to clarify.
13.	KJK	Glossary	Add actinides, cross-section, cross-section library, resonance integral.	Added.

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Comment Number	Commenter	Page, Paragraph	Comment Summary	Resolution
14.	KJK	Section 2.1	State that Table 2.1 entries are arithmetic means. Mean discrepancy seems to differ significantly between the three reactors. "Overall" standard deviation in Table 2.1 is simply the standard deviation of the 114 values, not based on the "within reactor" variation. If so (which should be stated), "overall" standard deviation of 4.8 may actually overstate the uncertainty.	Table was clarified.
15.	KJK	Table 2.1	Relabel columns as in Table 2.2.	Columns were relabeled.
16.	KJK	Section 2.2, Table 2.2	State how standard deviation of 7.9 was calculated.	Footnote was added to Table 2.2.
17.	KJK	Section 2.3	Define "elevation" in Figures 2.1 and 2.2.	Axis label was revised in each figure.
18.	KJK	Section 3.1, para. 1	State that "uncertainty" refers to coefficient of variation, i.e., the ratio of standard deviation to mean, expressed as percentage.	Uncertainty was defined.
19.	KJK	Section 3.1	State that Table 3.1 entries are arithmetic means ± standard deviations.	Text was revised to clarify.
20.	KJK	Section 3.1, last para.	Should sentence say that errors are not cumulative because they are independent (rather than "random")?	Yes - sentence was changed.

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Comment Number	Commenter	Page, Paragraph	Comment Summary	Resolution
21.	KJK	Section 3.2, last para.	Implies that before 1949, reactor power levels and tube power distributions were unstable and/or poorly defined. Implications for uncertainty of source terms for this period?	Text was revised to clarify.
22.	KJK	Section 3.3, para. 3	Define uncertainty here as unknown but largely systematic error, as opposed to the primarily random errors in preceding sections.	Replaced "uncertainty" with "differences."
23.	G. Caldwell	Page v, para. 1, line 4-5	Is a light water commercial reactor equal to the one-pass Hanford Production Reactors?	No.
24.	M.A. Robkin	Page 3.3, para. 3	Rephrase paragraph to address the current status of the estimating procedure for isotopes such as iodine-131. When the practice of discharging each tube at approximately equal plutonium inventory is taken into account, the inventory of a shorter-lived isotope in the dissolved fuel is larger than the amount estimated from core average specific inventory.	Paragraph was revised.
25.	MAR	Page 3.3	The set of coupled differential equations for a generalized isotopic chain is linear, i.e., there are not products or powers greater than unity of the concentrations of isotopes. Also,	Text was revised.

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Comment Number	Commenter	Page, Paragraph	Comment Summary	Resolution
			coefficients are discretely constant in time, i.e., they are slowly varying at most and held constant in each time step.	
26.	MAR	Page 3.6	Text refers to high neutron capture cross section of cesium-136 and ruthenium-106 as reasons that they are destroyed during irradiation at a rate sensitive to cross-section values used in the ORIGEN2 calculation. Yet calculations show that burnup is a trivial contributor to the isotope inventory, and the error in the capture cross-section is irrelevant.	Text was revised.
27.	B. Shleien	Page v	Are error terms additive? Does equation [in original comments] hold?	$\sum E = \sqrt{(E_1)^2 + (E_2)^2 + (E_3)^2}$ does not hold because the uncertainties in nuclear data, code input information, and code accuracy are not standard deviations or statistical variances. Rather they are just rough estimates of the maximum range of the variation seen in examples used in the text.
28.	BS	Page 2.2	Define "Monticello" measurements.	Text was clarified.

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Comment Number	Commenter	Page, Paragraph	Comment Summary	Resolution
29.	BS	Page 3.8	Point out that variation in code is not very large for iodine-131. Yet for the water pathway, isotopes of interest show a very large difference. However, during the Phase I study period, measurements in the Columbia are available.	Text was added on page 3.8 to clarify.
30.	J.E. Till	Page v	Relate statement about uncertainties in source term to the dominant radionuclides discussed in Napier's report. In Phase I, report our best estimate of uncertainty for each of these: I-131, Ru-106, Ru-103, I-132, Xe-133, Co-60, Te-129, Ce-144, Pu-239, and Y-91.	The uncertainties in the Summary apply to <u>any</u> isotope calculated using ORIGEN2, except for the use of a non-specific cross-section library. Table 3.2 examines the uncertainty to be expected from this source for each of the dominant radionuclides (which include all of those on John Till's list). Table 3.2 gives an isotopic-dependent estimate of the <u>maximum</u> possible uncertainty from this source. The document does not include an estimate of the overall statistical uncertainty in using ORIGEN2.
31.	JET	Page v	Since iodine-131 gives more than 95% of the dose, what is its uncertainty in the ORIGEN2 code for Hanford reactors?	NA - The uncertainty in iodine-131 will be addressed in the Phase II iodine closure task. ORIGEN2 is not used.

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Comment Number	Commenter	Page, Paragraph	Comment Summary	Resolution
32.	JET	Page v	How will uncertainties in the study be reported? Be consistent in describing uncertainty and reporting values (variability, factors of two, etc.).	No change to document required.
33.	JET	Page vii	Glossary - explain that there were two D reactors (D&DR) and two K reactors (KE&KW).	Definitions for D and K were revised.
34.	JET	Page 1.1	Describe additional QA measures taken to make sure ORIGEN was used correctly, e.g., were ORIGEN runs compared with those made at other locations for similar reactor configurations and spectra?	Paragraph was added.
35.	JET	Page 2.2 and Table 2.2	Term "isotopics" is unusual -- do you mean isotopes?	Term was replaced with "isotopic content."
36.	JET	Page 2.2, para. 3, sentence 1	Refer to Napier report here.	Reference was added.
37.	JET	Page 3.2, para. 3, sentence 3	Describe reliability of instruments.	Sentence was added.

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Comment Number	Commenter	Page, Paragraph	Comment Summary	Resolution
38.	JET	Page 3.2, para. 3, 5th line from bottom	Explain why the distribution of fuel element burnup around the mean is of little concern.	Explained.
39.	JET	Page 3.3, para. 2	Add references to support this statement.	Paragraph was revised.
40.	JET	Page 3.3, para. 4	"... solutions are accurate and the uncertainties are negligible." How is "negligible" defined here? Seems to support the conclusion that uncertainties associated with ORIGEN2 generated source terms are small. Reference?	Statement was revised to clarify.
41.	JET	Page 3.7, Table 3.2	Do not understand rationale for order of the table. Are abbreviations used in Column 4 coexistent with what we want in the study, e.g., "y" and "h"? Several nuclides are not included, notably H-3, Tc-99, and I-129.	Page 3.6, paragraph 2, line 3, explains the rationale for the order of the table (i.e., "arranged in decreasing order of the percentage difference between the CANDU and the N Reactor spectrum"). The abbreviation for the time unit of the half-life is used for clarity for a lay audience (i.e., yr instead of y, da for d, min for m). It is consistent with the HEDR editorial decision to use "iodine-131"

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Comment Number	Commenter	Page, Paragraph	Comment Summary	Resolution
				instead of I-131, etc. I-129, H-3, and Tc-99 were added to Table 3.2.
42.	JET	Page 4.1, para. 3	Are there specific radionuclides in the list of dominant nuclides for which uncertainty would be greatly reduced by doing this activity? If so, identify expected reduction in uncertainty.	NA - Table 3.2 provides the degree of uncertainty for each of the dominant nuclides from using the N Reactor library.
43.	JET	Page 5.1, para. 2, sentence 1	List contains radionuclides being considered in the HEDR Project based on a preliminary analysis of their contribution to dose. List may be augmented as study progresses.	Section 5.0 was rewritten.
44.	JET	Page 5.1, para. 2	Conclusions should summarize what we know about the dominant radionuclides for the air pathway and make a statement about uncertainties for the radionulcides in the surface water pathway had we decided to use ORIGEN2 to generate those source terms. Future work should focus on other factors inherent in source term development rather than further analysis of ORIGEN2 uncertainties.	Section 5.0 was rewritten.

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